

# *Synopsis of Research*

Conducted under the 2008-2009  
Northern Contaminants Program



Indian and Northern  
Affairs Canada

Affaires indiennes  
et du Nord Canada

Canada



# **Synopsis of Research Conducted under the 2008-2009 Northern Contaminants Program**

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## Foreword

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This report provides a summary of the progress to date of research and monitoring studies on contaminants in northern Canada, and related education, communications and policy activities that were conducted in 2008-2009 under the auspices of the Northern Contaminants Program (NCP). The projects cover all aspects of northern contaminants issues, as outlined in the NCP blueprints, including human health, monitoring the health of Arctic peoples and ecosystems and the effectiveness of international controls (abiotic monitoring and modeling, and biotic monitoring), education and communications, international policy and program management.

These projects were evaluated as proposals, by external peer reviewers, technical review teams, territorial/regional contaminants committees, and the NCP Management Committee to ensure that they support the overall Northern Contaminants Program objectives.

Further information about the Northern Contaminants Program is available on the NCP website at [www.ainc-inac.gc.ca/ncp](http://www.ainc-inac.gc.ca/ncp).

### Official Languages Disclaimer

These synopsis reports are published in the language chosen by the researchers. The full reports have not been translated. The Abstracts are available in English and French at the beginning of each report. Complete individual project synopses are available in either official language, upon request.

Requests for individual reports can be made to: [PLCN-NCP@ainc-inac.gc.ca](mailto:PLCN-NCP@ainc-inac.gc.ca).

## Avant-propos

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Le présent rapport comporte un résumé des progrès réalisés à ce jour dans le cadre des projets de recherche et des études de contrôle sur les contaminants dans le Nord canadien ainsi que des activités de sensibilisation, de communication et d'orientation menées en 2008-2009 sous l'égide du Programme de lutte contre les contaminants dans le Nord (PLCN). Les projets portent sur tous les aspects du dossier des contaminants dans le Nord, décrits dans les plans directeurs du PLCN, soit la santé humaine, la surveillance de la santé des résidents et des écosystèmes de l'Arctique et de l'efficacité des mécanismes internationaux de contrôle (surveillance abiotique et modélisation; surveillance biotique), l'éducation et les communications, la politique internationale et la gestion de programme.

Des pairs examinateurs de l'extérieur, des équipes d'examen technique, des comités territoriaux sur les contaminants, un comité régional sur les contaminants et le comité de gestion du PLCN ont évalué les propositions de projet, afin de s'assurer de la réalisation des objectifs du programme.

Pour obtenir d'autres renseignements, consultez le site Web du Programme de lutte contre les contaminants dans le Nord, à l'adresse [www.ainc-inac.gc.ca/ncp](http://www.ainc-inac.gc.ca/ncp).

### Avertissement lié aux langues officielles

Les chercheurs ont rédigé leurs rapports dans la langue de leur choix. Les rapports n'ont pas été traduits en entier, mais comportent un résumé en anglais et en français, au début. Il est possible d'obtenir sur demande le synopsis d'un projet dans l'une ou l'autre des langues officielles.

Pour obtenir un rapport, envoyez un courriel à l'adresse [PLCN-NCP@ainc-inac.gc.ca](mailto:PLCN-NCP@ainc-inac.gc.ca).



# Introduction

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The Northern Contaminants Program (NCP) was established in 1991 in response to concerns about human exposure to elevated levels of contaminants in fish and wildlife species that are important to the traditional diets of northern Aboriginal peoples. Early studies indicated that there was a wide spectrum of substances – persistent organic pollutants, heavy metals, and radionuclides – many of which had no Arctic or Canadian sources, but which were, nevertheless, reaching unexpectedly high levels in the Arctic ecosystem.

The Program's key objective is *to reduce and, where possible, eliminate contaminants in northern traditional/country foods while providing information that assists informed decision making by individuals and communities in their food use.*

Under the first phase of the NCP (NCP-I), research was focussed on gathering the data required to determine the levels, geographic extent, and source of contaminants in the northern atmosphere, environment and its people, and the probable duration of the problem. The data enabled us to understand the spatial patterns and temporal trends of contaminants in the North, and confirmed our suspicions that the major sources of contaminants were other countries. The data, which included information on the benefits from continued consumption of traditional/country foods, was also used to carry out assessments of human health risks resulting from contaminants in those foods. Results generated through NCP-I are synthesized in the *Canadian Arctic Contaminants Assessment Report*.

Extensive consultations were conducted in 1997-1998 to find the common elements between the concerns and priorities of northern communities and the scientific needs identified as critical for addressing the issue of contamination in Canada's North. As a result, priorities for current and future research are based on an understanding of the

Le Programme de lutte contre les contaminants dans le Nord (PLCN) a été créé en 1991, en réaction aux inquiétudes que suscitait l'exposition des humains à des concentrations élevées de contaminants chez les espèces sauvages aquatiques et terrestres constituant une part importante du régime alimentaire traditionnel des populations autochtones du Nord. Les premières études ont mis en évidence une vaste gamme de substances – polluants organiques persistants (POP), métaux lourds et radionucléides –, substances qui dans de nombreux cas n'avaient pas de source dans l'Arctique, ni même au Canada, mais qui se retrouvaient néanmoins à des concentrations anormalement élevées dans l'écosystème de l'Arctique.

Le Programme a comme principal objectif de *travailler à réduire et, dans la mesure du possible, à éliminer les contaminants présents dans les aliments traditionnels, tout en fournissant de l'information pour aider les individus et les collectivités à prendre des décisions éclairées au sujet de leur alimentation.*

Le premier volet du PLCN (PLCN-I) visait principalement à réunir les données nécessaires pour déterminer les concentrations, l'étendue géographique et la source des contaminants dans l'atmosphère, l'environnement et les habitants du Nord ainsi que la durée probable du problème. L'information recueillie a permis de comprendre les tendances spatio-temporelles de la contamination dans le Nord et de confirmer les soupçons à savoir que les principales sources de contaminants se situent à l'étranger. De plus, ces données, qui portaient notamment sur les bienfaits de la consommation régulière de tels aliments, ont servi à évaluer les risques pour la santé humaine de la présence de contaminants dans les aliments traditionnels. Dans le *Rapport de l'évaluation des contaminants dans l'Arctique canadien*, on présente les résultats obtenus dans le cadre du PLCN-I.

De vastes consultations ont été menées en 1997-1998 en vue de concilier les préoccupations et priorités

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species that are most relevant for human exposure to contaminants in the North, and geographic locations and populations that are most at risk.

In 1998-1999, the NCP began its second phase (NCP-II), which continued until 2002-2003. Results of this phase are synthesized in the *Canadian Arctic Contaminants Assessment Report II (CACAR II)*. NCP-II supported research designed to answer questions about the impacts and risks to human health that may result from current levels of contamination in key Arctic food species. To ensure a balanced assessment of the risks, an emphasis is placed on characterizing and quantifying the benefits associated with traditional diets. Communications activities are also emphasized and supported under NCP-II. Under the leadership of the northern Aboriginal organizations, the dialogue between northerners and the scientific community, which was initiated in NCP-I, continued to build awareness and an understanding of contaminants issues, and helped to support the ability to deal with specific contaminant issues at the local level.

In addition, the NCP effort to achieve international controls of contaminants remained strong in NCP-II. The legally binding POPs protocol, under the United Nations Economic Commission for Europe (UN ECE) Convention on Long-range Transboundary Air Pollution, has been successfully negotiated and was signed by 34 countries (including Canada) at the UN ECE Ministerial Conference in Aarhus, Denmark in June 1998. Canada ratified this agreement in December 1998. Negotiations for a legally binding global instrument on POPs under the United Nations Environment Programme have now also been completed with the signing of the POPs Convention in Stockholm, Sweden, May 23, 2001. The Convention has been signed by more than 100 countries; Canada has signed and ratified the Convention. Cooperative actions under the Arctic Council, including the circumpolar Arctic Monitoring and Assessment Programme (AMAP) and the Arctic Council Action Plan (formally launched in October 2000), are continuing. NCP continues to generate the data that allows Canada to play a leading role in these initiatives.

The NCP is directed by a management committee that is chaired by the Indian and Northern Affairs Canada, and which includes representatives from

des collectivités du Nord et les activités scientifiques nécessaires pour traiter la question des contaminants dans le Nord canadien. Ainsi, on a établi, pour les travaux actuels et à venir, des priorités fondées sur la définition des espèces principales par lesquelles les habitants du Nord se trouvent exposés aux contaminants ainsi que des régions et des populations les plus à risque.

Dans le deuxième *Rapport de l'évaluation des contaminants dans l'Arctique canadien*, on trouve une synthèse des résultats du second volet du Programme (PLCN-II), mis en œuvre de 1998-1999 à 2002-2003. Les recherches menées au cours de ce volet visaient à déterminer les risques et conséquences pour la santé humaine associés au degré actuel de contamination d'aliments clés dans l'Arctique. Pour que l'évaluation des risques soit juste, on a aussi défini et quantifié les avantages des régimes traditionnels. On a aussi mis l'accent sur les activités de communication dans le cadre du PLCN-II. Sous la conduite des organisations autochtones du Nord, le dialogue entre les habitants de la région et la communauté scientifique, amorcé lors du PLCN-I, a continué d'accroître la sensibilisation aux questions liées aux contaminants et la compréhension de celles-ci et a permis d'améliorer la prise en charge de certains problèmes associés aux contaminants à l'échelle locale. En outre, les efforts déployés dans le cadre du PLCN afin d'assurer l'établissement de mécanismes internationaux de contrôle des contaminants sont demeurés un objectif privilégié du second volet. À l'occasion de la Conférence ministérielle de la Commission économique des Nations Unies pour l'Europe, ayant eu lieu à Aarhus, au Danemark, en juin 1998, trente-quatre pays (dont le Canada) ont signé le protocole sur les POP, ayant force obligatoire, dans le cadre de la Convention sur la pollution atmosphérique transfrontalière à longue distance. Le Canada a ratifié l'entente en décembre 1998. Les négociations visant la conclusion d'un accord global sur les POP, ayant force obligatoire, dans le cadre du Programme des Nations Unies pour l'environnement, ont abouti à la signature de la Convention sur les polluants organiques persistants, à Stockholm, en Suède, le 23 mai 2001, par plus de cent pays. Le Canada a signé et ratifié la Convention. Les efforts concertés déployés dans le cadre des travaux du Conseil de l'Arctique, ce qui comprend le Programme de surveillance et d'évaluation de l'Arctique et le Plan d'action du Conseil de



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four northern Aboriginal organizations (Council of Yukon First Nations, Dene Nation, Inuit Tapiriit Kanatami, and Inuit Circumpolar Conference), the Yukon, Northwest Territories and Nunavut Territorial Governments, Nunavik, and four federal departments (Environment, Fisheries and Oceans, Health, and Indian Affairs and Northern Development). The management committee is responsible for establishing NCP policy and research priorities and for final decisions on the allocation of funds. Three territorial contaminants committees in the Yukon, Northwest Territories and Nunavut (established in May 2000), and a regional contaminants committee in Nunavik support this national committee. Funding for the NCP's \$4.4 million annual research budget comes from INAC and Health Canada.

The NCP Operational Management Guide, available on the NCP website ([www.ainc-inac.gc.ca/ncp](http://www.ainc-inac.gc.ca/ncp)), provides a summary of the management structures and review processes used to effectively implement the NCP. The Guide explains the overall management structures currently used, the proposal review process and outlines a protocol to be used to publicly disseminate health and harvest information generated by the NCP. Background information on all NCP committees and review teams is also provided.

In 1998, the NCP Management Committee redesigned the NCP-Phase II for application under the 1999-2000 funding year. The two main initiatives undertaken were: 1) the development of blueprints that represent the long-term vision and strategic direction for NCP-II; and 2) the implementation of a more open and transparent proposal review process. This new management structure is designed to ensure that the NCP remains scientifically defensible and socio-culturally aware, while at the same time, achieving real progress in terms of the Program's broad policy objectives.

Blueprints were developed for each of the three main NCP subprograms: i) Human Health, ii) Monitoring the Health of Arctic People and Ecosystems and the Effectiveness of International Controls, and iii) Education and Communications. The blueprints are used to provide the necessary guidance to project proponents for the development of proposals as well as to peer reviewers, review teams and the NCP Management Committee

l'Arctique (lancé officiellement en octobre 2000), se poursuivent. Les données recueillies dans le cadre du PLCN permettent au Canada de jouer un rôle de premier plan dans ces initiatives.

Le PLCN est dirigé par un comité de gestion, présidé par Affaires indiennes et du Nord Canada (AINC), qui compte des représentants de quatre organisations autochtones du Nord (soit le Conseil des Premières nations du Yukon, la Nation dénée, Inuit Tapiriit Kanatami et la Conférence circumpolaire inuite), des gouvernements du Yukon, des Territoires du Nord-Ouest et du Nunavut, du Nunavik et de quatre ministères fédéraux (Environnement Canada, Pêches et Océans Canada, Santé Canada et Affaires indiennes et du Nord Canada). Le comité de gestion est chargé de l'établissement des priorités du PLCN en matière de politique et de recherche et de la prise des décisions définitives en matière de financement. Trois comités territoriaux sur les contaminants (Yukon, Territoires du Nord-Ouest et Nunavut), mis sur pied en mai 2000, et un comité régional sur les contaminants (Nunavik) appuient les travaux du comité national. Les fonds pour le budget annuel de recherche de 4,4 millions de dollars du PLCN-II proviennent d'AINC et de Santé Canada.

*Le Guide de gestion des opérations du Programme de lutte contre les contaminants dans le Nord*, que l'on peut consulter sur le site Web du Programme ([www.ainc-inac.gc.ca/ncp](http://www.ainc-inac.gc.ca/ncp)), comprend un résumé des structures de gestion et des processus d'examen utilisés pour assurer la mise en œuvre efficace du PLCN. On y présente les structures globales de gestion actuelles, le processus d'examen des projets et la marche à suivre pour la diffusion publique d'informations sur la santé et la récolte recueillies dans le cadre du Programme. Le Guide comporte aussi des renseignements généraux sur tous les comités et équipes d'examen du PLCN.

En 1998, le comité de gestion du PLCN a réorienté le second volet du Programme en vue de l'exercice financier 1999-2000. Deux initiatives clés ont été mises en œuvre, soit : 1) l'élaboration de plans directeurs représentant la vision à long terme et l'orientation stratégique du second volet; 2) la mise en œuvre d'un processus ouvert et transparent d'examen des propositions. Cette nouvelle structure de gestion visait à assurer le caractère défendable du Programme du point de vue scientifique et la prise en compte des aspects socioculturels, tout en assurant la réalisation de progrès réels en ce qui a trait aux objectifs stratégiques généraux du PLCN.

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for evaluating proposals. They are evolving documents that are reviewed at least annually.

Under a revamped proposal review process, the NCP Technical Committee was replaced with an external peer review process facilitated by review teams. The review of proposals is a two pronged approach involving a scientific review by external peer reviewers, facilitated by technical review teams, and a socio-cultural review facilitated by the Regional Contaminants Committees (RCCs). Both sets of recommendations are considered by the management committee in making final funding decisions. Proposals submitted under the Education and Communications subprogram are evaluated by a technical review team. All peer reviewers, review teams and RCCs use evaluation criteria and the blueprints to review and rate proposals. Written consent from the appropriate northern community authority or national-level Aboriginal organization is required for all projects involving field work in the North and/or analyses of samples as a condition of approval for funding.

This report provides a summary of the progress to date of research and activities funded by the Northern Contaminants Program in 2008-2009. It is a compilation of reports submitted by project teams, emphasizing the results of research and related activities that took place during the 2008-2009 fiscal years. The report is divided into chapters that reflect the broad scope of the NCP: Human Health; Environmental Trends Related to Human Health and International Controls (including abiotic monitoring and biotic monitoring), and Education and Communications.

On a créé un plan directeur pour chacun des trois sous-programmes principaux : i) santé humaine; ii) surveillance de la santé des habitants et des écosystèmes de l'Arctique et de l'efficacité des mécanismes internationaux de contrôle; iii) éducation et communications. Ces plans fournissent les directives nécessaires aux promoteurs de projet, pour l'élaboration de leurs propositions, ainsi qu'aux pairs examinateurs, aux équipes d'examen et au comité de gestion du PLCN, pour l'évaluation des projets. Il s'agit de documents évolutifs, examinés au moins une fois par année.

Par suite de changements apportés au processus d'examen, un processus d'examen externe par les pairs, facilité par des équipes d'examen, a remplacé le comité technique du PLCN. L'évaluation des projets se fonde sur une approche à deux volets comprenant un examen scientifique par des pairs examinateurs de l'extérieur (facilité par des équipes d'examen technique) ainsi qu'un examen des aspects socioculturels, mené par les comités sur les contaminants. Le comité de gestion se penche sur les deux types de recommandation en vue de la prise de décisions définitives en matière de financement. Un comité d'examen technique évalue les projets soumis dans le cadre du sous-programme sur l'éducation et les communications. Les pairs examinateurs, les équipes d'examen et les comités sur les contaminants se servent des critères d'évaluation et des plans directeurs pour évaluer et noter les projets. Le consentement écrit d'une autorité compétente de la collectivité nordique ou d'une organisation autochtone nationale est requis pour tous les projets comportant des travaux sur le terrain dans le Nord ou des analyses d'échantillons, comme condition d'approbation du financement.

Le présent rapport comporte un résumé des progrès accomplis à ce jour relativement aux travaux de recherche et aux activités financés par le Programme de lutte contre les contaminants dans le Nord, en 2008-2009. Il s'agit d'une compilation des données provenant des rapports soumis par les équipes de projet. On met l'accent sur les résultats des travaux de recherche et des activités connexes menés au cours de l'exercice 2008-2009. Les chapitres du rapport rendent compte de la portée étendue du programme : soit la santé humaine; les tendances environnementales liées à la santé humaine et les mécanismes internationaux de contrôle (y compris la surveillance abiotique et biotique) et l'éducation et les communications.

# ***Human Health***





# Nunavik cohort study on exposure to environmental contaminants and child development

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## Abstract

We have conducted three studies in Nunavik during the last 15 years: monitoring of prenatal exposure based on cord blood sampling, and two effect studies, one with infants up to 12 months of age, and one at preschool age. The proposed study will follow-up these three cohorts of children at 10 years of age. The aims are to document the long-term effects of pre- and postnatal exposure to environmental contaminants and to evaluate the degree to which omega-3 fatty acids and selenium may protect against adverse effects. This 5-year study will involve 300 school-age children from the 14 Nunavik communities. Year 2008/2009 was devoted to data collection, data analyses and publications as well as presentation of a portion of the study results to the Nunavik Nutrition and Health Committee (NNHC).

## Résumé

Nous avons réalisé trois études au Nunavik au cours des 15 dernières années : la surveillance de l'exposition prénatale d'après des échantillons de sang ombilical et deux études sur les effets chez des nourrissons allant jusqu'à l'âge de 12 mois et chez des enfants d'âge préscolaire. L'étude proposée fera un suivi de ces trois cohortes d'enfants à l'âge de 10 ans. Les buts sont de documenter les effets à long terme de l'exposition pré et postnatale aux contaminants environnementaux et d'évaluer le degré de protection que les acides gras omega-3 et le sélénium peuvent apporter contre les effets nocifs. Cette étude d'une durée de cinq ans portera sur 300 enfants d'âge scolaire de 14 collectivités du Nunavik. Durant l'année 2008-2009, nous avons recueilli des données, nous les avons analysées, nous avons publié des articles et nous avons présenté une partie des résultats de l'étude au Comité de la nutrition et de la santé du Nunavik.

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## Key Messages

- The data collection for this ongoing study started in October 2005 and to date, 277 Nunavik children have been successfully tested at 11 years of age.
- Year 2008/2009 was devoted to training of new research staff, data collection, updating of the database, data analyses and publications.
- A portion of the results involving the follow-up at age 11 years was presented to the NNHC, along with results from previous follow-ups.
- The data collection will end in 2009/2010 with all study results available in 2010/2011.

## Messages clés

- La collecte de données pour cette étude permanente a commencé en octobre 2005 et à ce jour, 277 enfants du Nunavik ont passé les tests avec succès à l'âge de 11 ans.
- Durant l'année 2008-2009, nous avons formé du nouveau personnel de recherche, recueilli des données, mis à jour la base de données, analysé des données et publié des articles.
- Nous avons présenté une partie des résultats incluant le suivi des enfants à l'âge de 11 ans au Comité de la nutrition et de la santé du Nunavik, de même que les résultats de suivis antérieurs.
- La collecte de données prendra fin en 2009-2010 et tous les résultats de l'étude seront disponibles en 2010-2011.

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## Objectives

The overall objectives of this prospective study are:

- To document the long-term effects of pre- and postnatal exposure to environmental contaminants such as PCBs, MeHg and lead in multiple domains.
- To evaluate if nutrients, such as polyunsaturated fatty acids (PUFAs) and selenium (Se), can protect against adverse effects of exposure to environmental contaminants.
- To document the impact of lifestyle factors, such as smoking, alcohol and drug use during pregnancy, on multiple domains of child development and behaviour and to document the specific contributions of exposure to environmental contaminants and these other substances to development and behaviour of Inuit children.

## Activities in 2008/2009

- A) Two data collection trips in Nunavik to test about 100 children.

The data collection trip conducted during the fall was four weeks long instead of six, due to staff-related difficulties: the maternal

interviewer working on this project since 2005 resigned and was replaced by a PhD student with limited availability for field work. As a result, 25 children were successfully tested during that trip. After that trip, the research nurse responsible, since the beginning of the study, for the coordination of the field work, blood sampling, growth, auditory and visual testing also resigned. We successfully find a new research nurse with enough coordinating experience and knowledge of the Nunavik region. She was trained in January. The PhD student hired as maternal interviewer was also replaced by a research professional who has been involved in the data collection of the Infant cohort. The second four weeks long data collection trip allowed testing an additional 34 children. A total of 59 children were tested during the 2008/2009 period.

- B) Data coding, data entry and update of the data base.

Data coding and data entry are completed for the participants seen during fall and is underway for those seen during the most recent data collection trip.



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C) Presentation of preliminary results to the NNHC.

We met the NNHC in Kuujuaq in November 2008. The following study results were presented:

- Developmental trajectory of exposure to environmental contaminants from birth to 10 years of age and the determinants of exposure at age 11 (Muckle);
- Effects of contaminants on vision and visual evoked potentials (Saint-Amour);
- Effects of contaminants on memory and attention assessed through event-related potentials (Jacobson);
- Effects of contaminants on child behaviour at preschool aged (Plusquellec).

Muckle also presented to the NNHC the results communicated from the infancy cohort as well as the public health recommendations provided by the NNHC and the public health director to the population in 2003, which allowed opening the discussion regarding how the new results presented impact on the conclusions and recommendations made previously. The NNHC agreed with the researchers that those results could be submitted to peer-reviewed journals without being communicated to the population.

D) Preliminary data analyses on two third of the cohort.

Extensive data analyses were conducted and the following articles were published in peer-review journals:

E) Behavioural outcomes assessed through video recordings.

In 2008/2009, analysis of behavioural data collected in preschoolers was completed. Current exposure to Pb was related to behaviour indicators of inattention and to examiner's ratings of impulsivity. Prenatal PCB exposure was significantly associated with emotionality assessed from direct observation of video recordings. This result was corroborated with data from examiners' ratings. All these effects are sub-clinical and have not been shown to impair the day-to-day functioning of

preschoolers, but the full impact will be known only when data from 10 year-old children will be analyzed. Those results have been presented to the NCP result workshop in September 2008 and to the NNHC in November 2008.

In 2008/2009, collection of video recordings has also been done in 131 eleven-year-old children and behavioural coding of emotional reactivity has been completed for those children. Behavioural coding of children's attention levels in these video recordings will be completed during the summer of 2009.

## Conclusions

To date, we have succeeded in collecting data from 277 children. Year 2008/2009 was devoted to data collection, data scoring, data entry and statistical analyses. The analyses conducted so far allowed to present a significant portion of results to the NNHC, to submit manuscript to scientific journals and present study results in national and international conferences.

## Expected Project Completion Date

2009/2010

Boucher, O., Muckle, G., Saint-Amour, D., Dewailly, É., Ayotte, P., Jacobson S.W., Jacobson J.L., Bastien C.H. (2009) The relation of lead neurotoxicity to the event-related potentials P3b component in Inuit children from Arctic Quebec. *Neurotoxicology*. doi:10.1016/j.neuro.2009.06.008

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- Fraser, S.L., Belkacem, A., Muckle, G., Jacobson, J., Jacobson, S.W. The validation of a composite indicator of prenatal alcohol exposure. *Alcohol and Alcoholism*.
- Fraser, S.L., Muckle, G., Julien, P., Jacobson, J.L., Jacobson, S.W., Dewailly, É., Holub, B. The relation of prenatal exposure to acute doses of alcohol to maternal and umbilical cord fatty acid composition. *Neurotoxicology and Teratology*.
- Fraser, S.L., Muckle, G., Jacobson, J.L., Jacobson, S.W., Julien, P., Dewailly, É. Evaluating the role of Polyunsaturated Fatty Acids in the relationship between prenatal alcohol exposure and infant development. *Alcoholism: Clinical and Experimental Research*.
- Plusquellec, P., Dewailly, E., Ayotte, P., Begin, G., Desrosiers, C., Després, C., Saint-Amour, D., Muckle, G. The Relation of Environmental Contaminants Exposure to Behavioral Indicators in Inuit Preschoolers in Arctic Quebec. *Neurotoxicology*.
- The following manuscripts will be submitted to international peer reviewed journals during the summer of 2009:**
- Boucher, O., Bastien, C.H., Saint-Amour, D., Dewailly, É., Ayotte, P., Jacobson J.L., Jacobson S.W., Muckle, G. Auditory oddball in 11-year-old Inuit children from Arctic Québec exposed to seafood contaminants: An event-related potentials study. *Environmental Health Perspectives*.
- Burden, M.J., Westerlund, A., Muckle, G., Dewailly, E., Nelson, C.A., Jacobson, S.W., Jacobson, J.L. The effects of maternal binge drinking during pregnancy on neural correlates of response inhibition and memory in childhood. *Alcoholism: Clinical & Experimental Research*.
- Furthermore, study results were presented in the following conferences:**
- Fraser, S.L., Fortin, S., Muckle, G., Gagnon, J., Laflamme, D. (03/2009) Infrequent Exposure to Acute Doses of Alcohol on the Cognitive Development of Inuit. 3rd International meeting on Indigenous Child Health: Many Voices Into One Song. Albuquerque New Mexico.
- Burden, M.J., Westerlund, A.J., Muckle, G., Ayotte, P., Dewailly, É., Nelson, C.A., Jacobson, S.W., Jacobson, J.L. (2008/11). The N2-P3 and ERN-Pe complexes in a go/no-go task: Implications for response inhibition and conflict monitoring. Society for Neuroscience, Washington, USA.
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- Dubé-Linteau, A., Dewailly, É., Muckle, G., Ayotte, P. (2008/05). Gene-environmental interactions and Inuit's health in Nunavik. Annual Workshop of the International Network for Circumpolar Health Research, Tromsø, Norway.
- Fraser, S.L, Muckle, G., Déwailly, É., Jacobson, J.L., Jacobson, S.W., Julien, P. (2008/05). The association between foetal/maternal DHA ratio and pre- and postnatal development in Inuit children. 8th Meeting of the International Society for the Study of Fatty Acid and Lipids, Kansas City, Missouri, USA.

# Assessment of contaminant and dietary nutrient interactions in Inuit Health Survey: Nunavut, Nunatsiavut and Inuvialuit

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## **Abstract**

The Assessment of contaminant and dietary nutrient interactions in the Inuit Health Survey (IHS) seeks to incorporate contaminants research within the context of a broader health research study. The IHS is a major study that will provide a snapshot and baseline data on the health status of Inuit People in Nunavut, Inuvialuit, and Nunatsiavut for the first time. In 2008 the IHS team travelled to four coastal Inuvialuit communities, six communities in the Kitikmeot Region of Nunavut, and five communities in Nunatsiavut aboard the CCGS *Amundsen*. There were 359 ship based participants with a total of 280 blood samples collected for contaminant and biomarker analysis in Inuvialuit, 577 participants with a total of 220 blood samples collected in Kitikmeot and 309 participants with 263 blood samples collected in Nunatsiavut. A separate land team completed the land based surveys in Inuvik, Aklavik, and Baker Lake during this time period as well with a

## **Résumé**

Dans le cadre de l'Enquête sur la santé des Inuits (Inuit Health Survey – IHS), l'évaluation des interactions entre les contaminants et les nutriments alimentaires vise à incorporer la recherche sur les contaminants dans le contexte d'une étude plus vaste sur la santé. L'IHS est une étude majeure qui fournira pour la toute première fois des données de base et un aperçu de l'état de santé de la population inuite du Nunavut, de la région désignée des Inuvialuit et du Nunatsiavut. En 2008, l'équipe de l'IHS à bord du NGGC *Amundsen* s'est rendue dans quatre collectivités côtières de la région désignée des Inuvialuit, dans six collectivités de la région de Kitikmeot au Nunavut et cinq collectivités du Nunatsiavut. Dans la région désignée des Inuvialuit, 359 participants sont embarqués à bord permettant la collecte d'un nombre total de 280 échantillons sanguins aux fins d'analyse des contaminants et des marqueurs biologiques. Dans la région de Kitikmeot, 577 personnes ont participé à l'enquête et l'équipe a

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total of 332 participants with 264 blood samples collected. Detailed health, dietary, and lifestyle questionnaires were also completed, as well as bone density measurements in women over 40 years of age and carotid artery ultrasounds in men and women over 40 years of age. The risks and benefits associated with the traditional food diet and the relationship between contaminants and health outcomes of the participants will be analyzed. We are developing the results communication plans with the public health professionals and community leaders.

recueilli un nombre total de 220 échantillons sanguins. Au Nunatsiavut, il y a eu 309 participants et l'équipe a recueilli 263 échantillons sanguins. Pendant la même période, une équipe distincte au sol a complété des sondages dans les collectivités d'Inuvik, Aklavik et Baker Lake. Au total 332 personnes y ont participé et 264 échantillons sanguins ont été recueillis. Les participants ont également répondu à des questionnaires détaillés sur la santé, l'alimentation et les habitudes de vie. L'équipe a pris des mesures de la densité osseuse chez les femmes de plus de 40 ans et réalisé des ultrasons de la carotide chez les hommes et les femmes de plus de 40 ans. Nous analyserons les risques et les avantages associés au régime alimentaire traditionnel et le rapport entre les contaminants et les résultats en matière de santé chez les participants. Nous sommes à élaborer des plans de communication des résultats avec les professionnels de la santé publique et les dirigeants communautaires.

## Key Messages

- 2008 was the second and final year of data collection for the project. A total of 359 participants from Inuvialuit, 577 participants from the Kitikmeot Region of Nunavut, and 309 participants from Nunatsiavut took part in the ship based portion Inuit Health Survey between August 6<sup>th</sup> and October 16<sup>th</sup> 2008. In addition 205 participants from Aklavik and Inuvik, and 124 from Baker Lake took part in the land based survey. Not all participants completed all portions of the survey.
- Information on diet and health status and blood samples for nutrient and contaminant analysis was collected.
- Analysis of the data collected in 2007 is ongoing.
- Communication plans are being developed with health professionals and community leaders.

## Messages clés

- L'année 2008 était la deuxième et dernière année de collecte de données pour le projet. Au total, 359 personnes de la région désignée des Inuvialuit, 577 personnes de la région de Kitikmeot au Nunavut et 309 participants du Nunatsiavut ont participé à la portion de l'Enquête sur la santé des Inuits se déroulant à bord du navire, du 6 août au 16 octobre 2008. De plus, 205 participants d'Aklavik et d'Inuvik et 124 participants de Baker Lake ont participé au sondage réalisé à terre. Ce ne sont pas tous les participants qui ont rempli toutes les sections du sondage.
- Nous avons recueilli des renseignements sur l'état de santé et le régime alimentaire des participants, ainsi que des échantillons sanguins pour analyser les contaminants et les éléments nutritifs.
- L'analyse des données recueillies en 2007 est en cours.
- Nous sommes à élaborer des plans de communication avec les professionnels de la santé et les dirigeants communautaires.

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## Short-term Objectives

- a. Measure the body burden of environmental contaminants including persistent organic pollutants (POPs) and mercury (Hg);
- b. Study the relationship between diet intake of contaminants and body burden;
- c. Evaluate the complex interactions between lifestyle factors (obesity, smoking, physical

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activity, alcohol use) and contaminants exposure including POPs and Hg exposures as determinants of health;

- d. Investigate the interactive effects between dietary nutrients such as vitamin D, iron, selenium, and fatty acids with POPs and Hg on health status of the participants;
- e. Study the relationship between contaminant exposures, nutrient intakes, lifestyle factors and their relationship with markers of thyroid function, blood pressure, insulin resistance, lipid profiles, markers of oxidative stress and inflammation, neurotoxicity and bone mineral density.
- f. Develop health prevention and health promotion policy and communication strategy for contaminants in partnership with the regional Inuit organizations and health authorities.

## Long-term Objectives

- a. To close some of the existing gaps in knowledge that affect communities undergoing acculturation by providing them with information on the benefits and risks for adults associated with the consumption of traditional and market food.
- b. To collect baseline data in a format compatible with work in Greenland, Nunavik and Alaska to allow for prospective evaluation of factors associated with new emerging disease cases to enable improved evaluation of contaminant risks and nutrient benefits.
- c. To build capacity for the communities and local health authorities in public health. To engage in knowledge translation of findings in communities, with the larger network of NCP colleagues, and with scientific audiences including peer-review publications.

## Introduction

Emerging research suggests linkages between contaminant exposure and chronic diseases and the potential for nutrient and lifestyle factors to either exacerbate or reduce risks associated with contaminant exposures on various outcomes. For example, the heart is one of the target organs for mercury (Hg), and there is considerable interest in evaluating Hg's association with heart disease as

well as with blood pressure. However, the evidence of Hg effect on heart disease to date is inconsistent and may be attributed to the heart healthy beneficial exposures that co-occur with Hg exposure through the food chain (Chan and Egeland, 2004). One of the key mechanisms for Hg toxicity involves the oxidative stress pathway, and antioxidants such as selenium (Se) and possibly vitamin E may ameliorate Hg's toxic effects (Beyrouty et al 2006). Oxidized low density lipoprotein (LDL) levels in Inuit were significantly lower than in a Caucasian population in preliminary analyses suggesting that dietary factors, such as selenium, vitamin E, and omega-3 fatty acids may play a protective role against cardiovascular disease among the Inuit (Dewailly et al 2001).

Serum concentrations of POPs are known to be influenced by body mass index, breastfeeding history, place of residence, and food habits. Also, weight loss may increase serum concentrations of POPs. Given the increasing prevalence of obesity in the Arctic, evaluation of the extent to which obesity and other lifestyle factors are associated with contaminant exposure levels are warranted. Similarly, research in Greenland found that smoking status was significantly associated with contaminant exposure levels. While cigarettes are known to contain cadmium and inhaled cadmium is readily absorbed, the relationship between smoking and serum POP levels needs to be further investigated to help evaluate possible synergistic smoking-contaminant effects as well as rule out smoking as a potential confounder in epidemiologic research exploring the health implications of contaminant exposure.

Substances structurally related to 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (TCDD) such as non- and mono-ortho chloro-substituted polychlorinated biphenyls (PCBs) as well as 2,3,7,8-chloro-substituted polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs), bind the aryl hydrocarbon receptor (AHR) with varying affinities. The ligand-receptor complex triggers the expression of genes that are involved in cell proliferation and differentiation. These compounds referred to as dioxin-like compounds (DLCs), are ubiquitous pollutants that are especially associated with the aquatic food chain. DLCs elicit a number of species-specific, toxic responses in laboratory and wildlife species,

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including hepatotoxicity, body weight loss, thymic atrophy, and impairment of other immune responses, dermal lesions, reproductive toxicity, alterations in vitamin A and thyroid hormone metabolism, teratogenicity, and carcinogenesis (Safe, 1990, 1994). Other possible effects linked to DLC exposure in humans include modification of the sex ratio at birth, alterations in thyroid function, and increased risk for diabetes (Kogevinas et al., 2001).

Under-nutrition is also a concern for selected micronutrients in the Arctic. The research will evaluate the extent to which nutrient exposure (including inadequate exposure) inter relate with contaminant exposure in order to guide priorities for future research. For example, vitamin D deficiency is a problem throughout Northern populations as evidenced by the occurrence of rickets in children and studies evaluating plasma concentrations of 25(OH) D in mothers, newborns, and children in the Arctic. Vitamin D deficiency results in hypocalcemia, which in turn increases parathyroid (PTH) concentrations. PTH is important as it stimulates bone resorption to mobilize bone calcium in order to maintain plasma calcium levels. Vitamin D is also known to influence the immune system. As contaminant research suggests immune, bone, and thyroid function as well as other potential effects of organochlorines, characterizing the extent to which contaminant exposure correlates with vitamin D status and the interplay of both nutrient status and the various POP contaminants as they relate to selected outcomes is warranted. We will develop a model to weigh the relative risk of contaminant versus other nutrients as determinants/risk factors for chronic diseases.

Measuring biomarkers of effect in the Inuit population can be an effective strategy to detect sub-clinical/early effects induced by exposure to POPs and MeHg. The two proposed biomarkers have been tested in other populations and have been measured in over 400 participants in the Nunavik study; therefore, this proposed study is for validation, specifically for the Inuit population, and not for development.

Monoamine oxidase (MAO) activities in platelet samples have been shown to be potential biomarker for neurotoxic effects, such as decline

in colour vision (Stamler et al, 2005; 2006a,b). Preliminary studies from the Nunavik study, in 2004, with over 400 participants (pending results for all OC and PCB analysis) have shown that MAO activities showed a significant negative association with MeHg (in men not in women) in addition to other lifestyle factors such as alcohol consumption and smoking. Another biochemical marker, the activity of paraoxonase 1 (PON1) in serum for cardiovascular effects, has been measured in the course of the Nunavik Health Survey. This enzyme which is isolated in the high density lipoprotein (HDL) fraction of blood lipids may prevent cardiovascular diseases by metabolizing toxic oxidized lipids associated with both low density lipoprotein (LDL) and HDL (Aviram, 2000; Gur et al., 2006; Mackess et al., 1998). A preliminary analysis of results obtained in 900 Inuit adults from Nunavik indicated that selenium is positively associated with PON1 activity, whereas mercury negatively associated with the activity of this enzyme.

## Results

### Inuvialuit and Nunavut

In August and September of 2008 the Inuit Health Survey travelled to four coastal Inuvialuit communities and six communities in the Kitikmeot Region of Nunavut aboard the CCGS Amundsen. A separate land team completed the land based surveys in Inuvik, Aklavik, and Baker Lake during this time period as well. The participation targets were reached for each region.

For the 2008 Inuvialuit Settlement Region portion of the survey there were 359 ship based participants with a total of 280 samples collected for OCs, 281 samples for metals, 283 for PON1, 283 for ChE, 128 samples for MAO, and 113 for DR-CALUX. For the Kitikmeot Region of Nunavut there were a total of 577 ship based participants, with a total of 220 samples collected for OCs, 228 for metals, 233 for PON1, 233 for ChE, 388 for MAO, and 45 for DR-CALUX. Detailed health, dietary, and lifestyle questionnaires were also completed, as well as bone density measurements in women over 40 years of age and carotid artery ultrasounds in men and women over 40 years of age.

For the land based surveys completed in Aklavik and Inuvik there were a total of 205 participants with 154 samples collected for OCs, 153 for metals, 153 for ChE, 153 for PON1, and 98 for DR-CALUX. In Baker Lake there were a total of 127 participants with 107 samples collected for OCs and 107 for heavy metals, 110 samples for both ChE and PON1, and 33 samples for DR-CALUX.

Country food samples were also collected in the Inuvialuit region for contaminant analysis, see table two.

### Nunatsiavut

In 2007 and early 2008 the Nunatsiavut Regional Inuit Health Survey Steering Committee participated in the planning process for the 2008 Survey. In October of 2008, the Inuit Health Survey travelled to five Inuit communities in the Nunatsiavut Region aboard the CCGS Amundsen. There were a total of 309 participants in Nunatsiavut with 263 samples collected for OCs, 264 samples collected for metals, 264 for PON1, and 98 for DR-CALUX. Because of logistics constraints, MAO samples were not collected in Nunatsiavut. Detailed health, dietary, and lifestyle questionnaires were also completed, in addition to bone density measurements in women over 40 years of age, and carotid artery ultrasounds in men and women over 40 years of age.

**Table 2 – Inuvialuit Country Food Samples 2008**

Caribou Meat	3
Caribou Liver	1
Arctic Char	1
Whitefish	2
Swan	2
Goose	4
Musk Ox	3
Beluga	10
Ringed Seal	10

## Project Plan and Expected Completion Date

This is the fourth year of a five year study. Preliminary results of participant profiles and health status, excluding contaminant results and contaminant related biomarkers, are being reported back to individual participants by the Inuit Health Survey Team. Biomarkers for neurological and cardiac effects and contaminants in blood samples have been measured. A communication plan continues to evolve with planning including the Department of Health and Social Service of Nunavut Government and the NAC, the Inuvialuit Settlement Region Inuit Health Survey Committee, and the Nunatsiavut Inuit

**Table 1 – Current status of sample analysis**

	Nunavut 2007	Inuvialuit, Western Nuavut and Baker Lake 2008	Nunatsiavut 2008
<b>Blood Samples</b>			
OCs	N = 1069 Completed	N = 761 In Progress	N = 263 In Progress
Metals	N = 1178 Completed	N = 769 In Progress	N = 264 In Progress
PON1	N = 1184 Completed	N = 779 In Progress	N = 264 In Progress
DR-CALUX	N = 292 Completed	N = 257 In Progress	N = 98 In Progress
MAO	N = 1127 Completed	N = 516 Completed	N = 0
ChE	N = 949 Completed	N = 779 In progress	N = 264 In Progress
<b>Food Samples</b>			
OCs	Completed	In Progress	NA
Mercury	Completed	In Progress	NA



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Health Survey Committee. Data analysis for the 2007 data will be completed in mid to late 2009, and the data analysis for the 2008 survey data is expected to be completed in mid to late 2010.

Results delivery will begin with a results delivery planning meeting between the regions and the researchers in mid November 2009. The entire project is expected to be completed by 2011.

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# Contaminant Nutrient Interaction Issues as part of a Public Health Intervention Study of Inuit Children in Nunavik: third year of data collection

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## **Abstract**

The present study on contaminant nutrient interactions is part of the Nutrition Program in Nunavik Childcare Centres, which aims to improve children's health by providing healthy meals rich in iron and containing country/traditional foods and selected market foods. This study aims to provide essential information about dietary patterns and intakes, nutritional status and contaminant exposure of young Inuit children, and about the risks/benefits concerning the use of traditional foods during preschool age. So far, 155 children have been recruited from the Ungava and the Hudson coasts, and 50 of them have been seen for a follow-up visit one year later. Blood contaminant levels (mercury and lead), dietary intakes and nutritional status (fatty acids, vitamins A, E, D, B<sub>12</sub>, folate, selenium and iron) are measured at recruitment and twelve months later for each participating child. Blood samples obtained from

## **Résumé**

La présente étude sur les interactions entre contaminants et éléments nutritifs fait partie du Programme de nutrition des Centres de soins pour enfants du Nunavik qui vise à améliorer la santé des enfants en leur fournissant des repas riches en fer et contenant des aliments du pays/aliments traditionnels et certains aliments achetés au marché. Cette étude a pour but de fournir des renseignements essentiels sur les habitudes et les apports alimentaires, l'état nutritionnel et l'exposition aux contaminants des jeunes enfants inuits et sur les risques et avantages concernant l'usage d'aliments traditionnels lorsque les enfants sont en âge préscolaire. Jusqu'à maintenant, nous avons recruté 155 enfants le long des côtes des baies d'Ungava et d'Hudson et 50 d'entre eux ont été revus un an plus tard pour un suivi. Les taux de contaminants dans le sang (mercure et plomb), les apports alimentaires et l'état nutritionnel (acides gras, vitamines A, E, D, B<sub>12</sub>, folate, sélénium



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2006-2008 have been stored for persistent organic pollutants (POPs) analysis which is planned for 2009-2010. The results of this study will support Nunavimmiut and the Public Health authorities in their effort to prevent contaminant toxicity and nutritional deficiencies in a way that maximizes the benefits of traditional/country foods without putting at risk the health of children. The present report presents preliminary results (data collection is ongoing) on the relationship between the iron and folate status and blood mercury and lead levels.

## Key Messages

- The data collection for this study started in 2006 and to date, 155 Nunavimmiut children have been recruited from both coasts with 50 of them seen a second time at one year interval (2 out of 3 cohorts have been completed so far).
- The activities for 2008-2009 were mainly related to data collection, data entry and some statistical analyses.
- Our analysis indicated that 12% of participating children at recruitment were anemic, but a higher proportion suffered from iron deficiency (13 to 50% according to the biochemical indicator used).
- The folate and iron status improved in children who were seen twice at one-year interval (n=50).
- Correlations revealed that a higher vitamin B<sub>12</sub> status was associated with lower blood mercury and lead levels. Low hematocrit levels (an indicator of anemia) were also related to higher mercury levels. On the contrary, a higher folate status was related to higher mercury levels, while higher levels of red cells indices (MCV, MCH, MCHC) were related to higher lead levels.
- In the fall 2009, more children will be recruited, increasing statistical power and allowing us to better document the contaminant nutrient interactions.

et fer) sont mesurés lors du recrutement et 12 mois plus tard, chez chaque enfant participant. Nous avons stocké des échantillons sanguins obtenus de 2006 à 2008 en vue de l'analyse des polluants organiques persistants (POP) prévue en 2009-2010. Les résultats de cette étude aideront les autorités du Nunavimmiut et de la santé publique dans leurs efforts pour prévenir la toxicité causée par les contaminants et les lacunes nutritionnelles de façon à maximiser les avantages des aliments du pays/traditionnels sans mettre en danger la santé des enfants. Le présent rapport fait état des résultats préliminaires (la collecte de données se poursuit) sur la relation qui existe entre les bilans de fer et de folate et les taux de mercure et de plomb dans le sang.

## Messages clés

- La collecte de données pour cette étude a commencé en 2006 et à ce jour, nous avons recruté 155 enfants du Nunavimmiut des deux côtes et revu 50 d'entre eux un an plus tard (2 cohortes sur 3 terminées à ce jour).
- Les activités en 2008-2009 étaient principalement liées à la collecte de données, la saisie de données et certaines analyses statistiques.
- Notre analyse a indiqué que 12 % des enfants participants étaient anémiques au moment du recrutement, mais une proportion plus élevée souffrait de déficience en fer (13 à 50 % selon l'indicateur biochimique utilisé).
- Il y a eu amélioration des bilans de folate et de fer chez les enfants revus à un an d'intervalle (n = 50).
- Les corrélations révèlent qu'un bilan plus élevé en vitamine B<sub>12</sub> était associé à des taux sanguins inférieurs en mercure et en plomb. De faibles valeurs d'hématocrites (un indicateur de l'anémie) étaient également reliées à des taux plus élevés de mercure. Par ailleurs, un bilan élevé en folate était relié à des taux de mercure plus élevés, tandis que des indices érythrocytaires élevés (VGM, TCMH, CCMH) étaient reliés à des taux de plomb plus élevés.
- À l'automne 2009, nous recruterons d'autres d'enfants, ce qui augmentera le pouvoir statistique de l'étude et nous permettra de mieux documenter les interactions entre les contaminants et les éléments nutritifs.

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## Objectives

The overall objectives of the present study are:

- i) To document the contaminant nutrient interactions in Nunavik children of preschool age.
- ii) To assist daycare directors, cooks and parents in making informed decisions concerning the benefits/risks of traditional/country foods among preschool children using dietary intakes, biochemical/haematological parameters and clinical information.

The specific objective of the analysis presented in this report is to explore the relationship between iron status, and blood mercury and lead levels, based on data collected during the first three years of the project.

## Introduction

Publications have pointed out a poor health status among Nunavimmiut children that could be related to a poor nutritional status (Hodgins, 1997; Willows *et al.*, 2000; Dallaire *et al.*, 2003). Also there is some evidence, based on results from mechanistic, experimental and observational studies, that various nutrients and diet components can protect against or attenuate the adverse effects of contaminants on health (Van Oostdam *et al.*, 2003; Van Oostdam *et al.*, 2005). For example, it has been suggested that a low micronutrient diet might predispose to toxicity from metals (Peraza *et al.*, 1998; Schell *et al.*, 2004).

Mineral and some heavy metals share the same transport divalent receptors for absorption and transport (Ballatori, 2002). Iron carrier in iron deficiency is upregulated and may also handle toxic elements. It has been reported that iron deficiency can increase the risk of metal poisoning in children (World Health Organization, 2001). Several authors have observed inverse associations between an abnormal iron status and blood concentrations of metals such as cadmium (Barany *et al.*, 2005) and lead (Ahamed *et al.*, 2007; Willows & Gray-Donald, 2002), whereas other authors did not find such a relationship (Després *et al.*, 2005). In Nunavik, cohort data from Muckle *et al.* showed that about 10-13% of Inuit infants were anemic at 6 months old (Plusquellec *et al.*, 2007) and 5% of children about 5 years of age had a low hemoglobin level

(Hb  $\leq$  105 g/L) (Després *et al.*, 2005). Also the last Inuit Health Survey still reported a high prevalence of iron deficiency among Nunavimmiut women of childbearing age (about 40%) even though iron intake seems relatively high in this age group (Plante *et al.*, 2007).

Other studies suggest associations between contaminant exposure and nutritional factors. **Vitamin A** homeostasis is altered by organochlorines in many species, including primates (Zile, 1992 cited by Van Oostdam *et al.*, 2005; Mos *et al.*, 2007). Vitamin A is important for normal vision, growth and immune function. In Nunavik, prenatal exposure to PCBs (Dallaire *et al.*, 2006) and neonatal vitamin A deficiency (Cameron *et al.*, 2008) have been related to the incidence of lower respiratory tract infections and acute otitis media in Inuit children. However, the understanding of the relationship between vitamin A, infections and contaminants is limited.

Organochlorines have also been related to bone diseases like osteoporosis and periodontitis, a teeth disorder, in animals (Sonne *et al.*, 2004) and in humans (Van Oostdam *et al.*, 2005). As **vitamin D** is known to have many functions including a role in bone health and teeth condition, it is of interest to take into account the vitamin D status when examining blood contaminant levels and its effects on human health. Moreover there is evidence that vitamin D deficiency exists in the Arctic population including children. Between July 2002 and June 2004, 104 confirmed cases of vitamin D-deficiency rickets have been reported in children in Canada, with the highest incidence rates among children who reside in the north (Yukon Territory, Northwest Territories and Nunavut) (Ward *et al.*, 2007). Along with the upcoming results from the Inuit Health Survey 2007-2008, this study will document the vitamin D status of Inuit children and describe the relationship between vitamin D and contaminants.

In addition, results from experimental studies suggest that **selenium**, **vitamin E** and **polyunsaturated fatty acids** could have a protecting effect against mercury toxicity (Van Oostdam *et al.*, 2005). Also some authors have recently suggested that essential nutrients derived from seafood such as iodine may prevent the negative effects of organochlorines on the thyroid economy during

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foetal development (Dallaire *et al.*, 2008). Still the nutrient contaminant interactions have seldom been examined in humans.

## Activities for 2008-2009

### Data collection

- Three data collections have been completed so far (during fall 2006, 2007 and 2008). The 2008 data collection took place in 7 communities: Kangiqsualujuaq, Kuujuaq, Quaqtaq, Kangiqsujuaq, Ivujivik, Akulivik and Inukjuak.

### Laboratory analysis and data entry

- A complete blood count (CBC), many iron status indicators, selenium, vitamins (A, D, E, B<sub>12</sub>, folate), C-reactive protein, red blood cells fatty acids and heavy metals (Hg, Pb) blood analyses have been carried out from 2006-2008. Blood samples obtained from 2006-2008 have been stored for persistent organic pollutants (POPs). Their analysis is planned for 2009-2010.
- All 24-h dietary recalls obtained from 2006-2007 have been computed by the Institut national de santé publique du Québec (INSPQ) using the Micro Gesta software. The computerization of all the food frequency questionnaires started in April 2009. Also, all the general questionnaires have been coded and computerized. Statistical analyses using SAS has started with the support of the INSPQ.

### Communication

- Results of the work completed during the first two years of the project have been presented to the NCP meeting in Yellowknife in September 2008 (poster session) and were published in the Synopsis of Research conducted under the 2007-2008 Northern contaminants program.
- Preliminary results were also presented at the daycare directors' annual meeting in Kuujjuraapik in February 2009, and to the director and the coordinator of the KRG Childcare Department.
- Contacts with public health authorities in Nunavik were made to inform them about high mercury levels observed in some participants (mainly children recruited in communities

included in the study in 2008). Also participating children diagnosed with anemia continued to be referred to local doctors.

## Results and discussion

Parents of children attending childcare centres were asked to enroll their child for two data collections, at one year interval. So far, 155 children have been recruited from the Ungava and Hudson coasts: 69 in 2006, 35 in 2007 and 51 in 2008. Fifty (50) of the 104 children recruited in 2006 and 2007 returned for a second visit, respectively in 2007 and 2008 (retention rate: 48%). Some of the results obtained on the iron, folate and vitamin B<sub>12</sub> status, as well as the blood Hg and Pb levels of Inuit children recruited from 2006-2008 are presented here. These results allow us to get a better picture of the blood nutrient and contaminant levels of Inuit children attending childcare centres in Nunavik.

Characteristics of the study sample are summarized in Table 1. A slightly higher proportion of children recruited were male and the majority of children were aged between 11-35 months, which is consistent with our inclusion criteria. Fifty-four percent came from the Hudson coast, the rest from the Ungava coast. The vast majority of children attended daycare five days a week. The biological mother was the main respondent for the child. More than three-quarters of the respondents were married or had a partner. Nearly half of them had completed a secondary level of education or more. The high rate of smoking observed in the Inuit population was evident as only 13% of households had no smokers, although 83% mentioned that they restricted smoking inside their home. Finally, almost 10% of respondents said they did not have enough food to eat until satisfied during the month prior to the interview.

Parameters of nutrient status and blood contaminant levels among children at recruitment (T1) from 2006-2008 are shown in Table 2. According to the reference cut-off for hemoglobin levels, anemia was present in 12% of participating children. This is consistent with cohort data from Muckle *et al.* who showed that about 10-13% of Inuit infants were anemic at 6 months old (Plusquellec *et al.*, 2007) and 5% of children about 5 years of age had a low hemoglobin level (Hb  $\leq$  105 g/L) (Després *et al.*, 2005). This proportion is also lower than the level of 25% observed

**Table 1. Characteristics of participants at recruitment (T1) from 2006-2008**

Characteristics	n	%
<b>Child characteristics</b>		
Gender (% male)	155	51.0
Age		
11-35 months	155	86.5
36-61 months		13.5
Coast of origin (% Hudson)	155	53.5
Breastfeeding status (% yes)	153	74.5
Child's health – subjective evaluation by the respondent (% Excellent or Very good)	154	59.1
Daycare attendance (% 5 days/week)	155	93.5
<b>Respondent characteristics</b>		
Relation to the child (% biological mother)	155	76.8
Marital status (% married or partner/Common Law)	155	72.9
Employment status (% currently employed)	155	81.9
Education (% secondary completed or more)	155	43.9
<b>Household characteristics</b>		
Number of smokers (% none)	155	12.9
Restriction about smoking (% forbidden inside the home)	149	85.2
Enough food to eat until satisfied in the month prior to the interview (% yes)	152	90.8

in 2002, among 9 months old Aboriginal Cree infants living in northern Quebec (Willows & Gray-Donald, 2002). Low hematocrit levels which are also used to establish the presence of anemia were observed in 4% of children. Red cells indices characteristic of iron deficiency anemia were also observed: MCV (size of red blood cells) and MCH and MCHC (content of red blood cells) were decreased (9-31%), while an elevated RDW (anisocytosis) was observed in 27% of children. Concurrent folate/B<sub>12</sub> deficiencies (16 and 2.5%) with its increasing red cells volume could partly explain the increase in RDW. Iron deficiency measured by abnormal levels of serum transferrin receptors, serum iron, TIBC and transferrin saturation varied between 13 and 50%, depending on the biochemical parameter used. Depleted iron stores (low ferritin levels) were observed in 20% of children. This last result is very similar to the level of 22.7% reported in Cree infants from northern Quebec (Willows & Gray-Donald, 2002). Thus, although the prevalence of anemia was relatively

low, iron and folate deficiencies at recruitment (T1) were of concern in these daycare children.

Almost 20% of subjects had Hg levels above the cut-off point (28.9 nmol/L), but no children were above the cut-off limit ( $>0.48 \mu\text{mol/L}$ ) for Pb levels (Table 2). The mean blood Hg and Pb concentrations observed in the present study (8.9 nmol/L and  $0.08 \mu\text{mol/L}$ ) were much lower than what has been observed in the 1990s for 110 five years old preschoolers from Nunavik (47.8 nmol/L and  $0.26 \mu\text{mol/L}$ ) (Després *et al.*, 2005).

The folate and iron status of children improved between Time 1 and Time 2 (2006-7, 2007-8 cohorts). More specifically, MCV, MCH and MCHC increased and high levels of serum transferrin receptors (indicative of a poor iron status) decreased (Table 3). However, we observed an increased in RDW possibly caused by the presence of a mixed iron and folate/vitamin B<sub>12</sub> deficiency which respectively produces a mix of both small and large cells hence elevating the RDW. Pb levels

**Table 2. Mean and proportion of abnormal values of nutrient and contaminant blood levels among children at recruitment (T1) from 2006-2008**

Parameters	n	Mean <sup>1</sup>	95% IC (lower–upper limit)	Cut-off value <sup>2</sup>	Proportion of values below or above the cut-off point (% (n))	
Iron status						
Hemoglobin (g/L)	121	121.0	(119.0–123.0)	< 110 or < 112	12.4	(15)
Hematocrit	121	0.37	(0.36–0.37)	< 0.32	4.1	(5)
MCV (fL)	121	74.7	(73.6–75.8)	< 67 or < 73	9.1	(11)
MCH (pg)	118	24.6	(24.1–25.1)	< 22 or < 25	15.3	(18)
MCHC (g/L)	118	328.3	(325.3–331.4)	< 320	30.5	(36)
RDW (%)	118	13.5	(13.2–13.9)	> 14.5	27.1	(32)
sTfR (mg/L)	106	2.0	(1.9–2.1)	> 2.5	13.2	(14)
Serum Iron (μmol/L)	119	7.7	(7.0–8.5)	< 5.4	25.2	(30)
TIBC (μmol/L)	119	80.6	(78.1–83.0)	> 86.0 or > 84.2	28.6	(34)
Transferrin saturation (%)	119	9.7	(8.7–10.7)	< 10 or < 12	49.6	(59)
Serum ferritin (μg/L)	119	17.9	(15.6–20.5)	< 10	20.2	(24)
Other nutrient status						
Vitamin B <sub>12</sub> (pmol/L)	119	765.0	(713.8–816.2)	< 288	2.5	(3)
Folate (nmol/L)	119	22.9	(20.9–24.8)	< 11.0	16.0	(19)
Contaminants						
Mercury (nmol/L)	129	8.9	E (7.1–11.3)	> 28.9	19.4	(25)
Lead (μmol/L)	129	0.08	(0.07–0.09)	> 0.48	0	(0)

MCV: mean corpuscular volume; MCH: mean corpuscular hemoglobin; MCHC: mean corpuscular hemoglobin concentration; RDW: red cell distribution width; sTfR: soluble transferrin receptor; TIBC: total iron binding capacity.

<sup>1</sup> Arithmetic mean, except for sTfR, serum iron, transferrin saturation, serum ferritin, mercury and lead (geometric mean).

<sup>2</sup> When two cut-off values are presented, they apply to children < 3 and ≥ 3 years old respectively.

E Interpret with caution.

obtained at one year interval, did not differ significantly, while Hg levels produced an unreliable estimate and cannot be presented.

Correlations were performed to identify the relationship between folate and iron status indices and blood Hg and Pb levels at recruitment (T1). As indicated in **Table 4**, inverse associations were observed between vitamin B<sub>12</sub> and hematocrit (an indicator of anemia) and blood Hg levels, while positive associations were observed with MCHC and folate levels. Thus, as vitamin B<sub>12</sub> and hematocrit levels increased, blood Hg decreased.

On the contrary, blood Hg levels increased with the improvement in MCHC and folate levels.

Blood Pb levels were also negatively associated with vitamin B<sub>12</sub> (**Table 4**). Unexpectedly, a negative association was also observed between blood Pb levels and total iron binding capacity (TIBC), whose level is usually higher-than-normal when the body's iron stores are low. Finally, positive associations were observed between blood Pb levels and red cell indices (MCV, MCH, MCHC). Important correlations have been reported between some indices of iron status (serum iron [ $r=-0.552$ ,

**Table 3. Nutrient and contaminant blood levels among children who participated in two data collections (2006-2007 and 2007-2008)**

Parameters	Recruitment (T1)			Follow-up (T2)			P-value <sup>2</sup>
	n	Mean <sup>1</sup>	95% IC (lower–upper limit)	n	Mean <sup>1</sup>	95% IC (lower–upper limit)	
Iron status							
Hemoglobin (g/L)	48	120.2	(116.7–123.7)	44	121.9	(117.8–125.9)	0.5968
Hematocrit	48	0.37	(0.36–0.38)	44	0.37	(0.36–0.38)	0.0581
MCV (fL)	48	74.1	(72.5–75.6)	44	77.0	(75.3–78.6)	<0.0001
MCH (pg)	48	23.9	(23.2–24.7)	38	25.5	(24.7–26.3)	<0.0001
MCHC (g/L)	48	322.5	(317.9–327.1)	38	331.7	(326.7–336.7)	0.0095
RDW (%)	48	13.3	(12.7–13.9)	38	14.1	(13.5–14.7)	0.0155
sTfR (mg/L)	42	2.001	(1.8–2.2)	36	1.8	(1.7–1.9)	0.0018
Serum Iron (μmol/L)	46	7.7	(6.6–9.1)	39	8.7	(7.4–10.3)	0.2957
TIBC (μmol/L)	46	84.2	(80.7–87.7)	39	81.9	(77.9–85.8)	0.1391
Transferrin Saturation (%)	46	9.3	(7.8–11.0)	39	10.8	(9.0–12.9)	0.1693
Serum Ferritin (μg/L)	46	16.9	(13.2–21.6)	39	16.5	(12.8–21.4)	0.5372
Other nutrient status							
Vitamin B <sub>12</sub> (pmol/L)	46	819.6	(735.3–903.9)	39	784.9	(690.0–879.7)	0.1242
Folate (nmol/L)	46	18.7	(15.8–21.5)	39	22.8	(20.6–24.9)	0.0027
Contaminants							
Mercury (nmol/L)	50	F	F	40	F	F	
Lead (μmol/L)	50	0.08	(0.07–0.09)	40	0.07	(0.06–0.09)	0.6677

MCV: mean corpuscular volume; MCH: mean corpuscular hemoglobin; MCHC: mean corpuscular hemoglobin concentration; RDW: red cell distribution width; sTfR: soluble transferrin receptor; TIBC: total iron binding capacity.

<sup>1</sup> Arithmetic mean, except for sTfR, serum iron, transferrin saturation, serum ferritin, mercury and lead (geometric mean).

<sup>2</sup> Paired-Samples T Test (Student), with log-transformed data for sTfR, serum iron, transferrin saturation, serum ferritin and lead.

F Unreliable estimate.

$p < 0.05$ ] and hematocrit [ $r = -0.427$ ,  $p < 0.05$ ]) and blood lead levels by Ahamed *et al.* (2007) in 75 Indian children aged 1 to 7. However, the proportion of children with elevated blood lead levels and anemia was high: 75% had blood Pb levels  $\geq 0.48$  μmol/L and 66% were anemic. Traditional/country foods are rich in iron, but also contain contaminants. Muckle *et al.* (2008) reported in 218 Nunavik children at 10 years of age that food items more strongly associated with blood Hg levels were marine mammal meat and fat, arctic char and goose. Consumption of goose and eggs from game birds, although not frequent, were the

food items related to higher child blood Pb concentrations (Muckle *et al.*, 2008). Twenty-four dietary recalls obtained in children at recruitment and one year later, will help us to better understand the impact of traditional/country foods on blood Hg and Pb levels. Statistical analyses are presently undergoing and will be available soon.

## Conclusions

To date, we have conducted three data collections (2006-2008) to obtain blood mercury and lead levels, nutritional status and dietary intakes in



**Table 4. Spearman correlations between blood nutrient and contaminant levels among children at recruitment (T1) from 2006-2008**

Parameters	n	Correlation coefficients (P-value)	
		Mercury	Lead
Iron status			
Hematocrit	119	-0.28955 <b>(0.0014)</b>	0.06617 (0.4746)
MCV	119	0.01564 (0.8659)	0.19975 <b>(0.0294)</b>
MCH	116	0.12291 (0.1887)	0.24751 <b>(0.0074)</b>
MCHC	116	0.18357 <b>(0.0486)</b>	0.18365 <b>(0.0484)</b>
TIBC	119	-0.14858 (0.1068)	-0.18327 <b>(0.0460)</b>
Other nutrient status			
Vitamin B <sub>12</sub>	119	-0.20557 <b>(0.0249)</b>	-0.42423 <b>(&lt;0.0001)</b>
Folate	119	0.18901 <b>(0.0395)</b>	0.04805 (0.6038)

MCV: mean corpuscular volume; MCH: mean corpuscular hemoglobin; MCHC: mean corpuscular hemoglobin concentration; TIBC: total iron binding capacity.

155 children at recruitment (T1) and 50 children (T2) who came back one year later for a follow-up visit. Blood samples obtained from 2006-2008 have been stored for persistent organic pollutants (POPs) analysis which is planned for 2009-10. Year 2008/2009 was devoted to the data collection, data entry, computerization of the 24-h dietary recalls and general questionnaires and some statistical analyses. The analyses conducted so far revealed that anemia was present in 12% of participating children at recruitment, but the proportion of iron deficiency was higher, varying between 13 and 50%, according to the biochemical indicator used. The folate and iron status improved in children who were seen twice at one-year interval. Correlations revealed that a higher vitamin B<sub>12</sub> status was associated with lower blood mercury and lead levels. Low hematocrit levels (an indicator of anemia) were also related to higher mercury levels. On the contrary, a higher folate status was

related to higher mercury levels, while higher levels of red cells indices (MCV, MCH, MCHC) were related to higher blood lead levels.

A nutrition intervention providing traditional/ country foods and healthy market foods has been implemented in the daycare centres of Nunavik. We studied the nutritional status and blood lead and mercury levels at one-year interval in 50 children who participated in this nutrition program. In the fall 2009, more children will be recruited, increasing statistical power and allowing us to better document the contaminant nutrient interactions.

## Expected Project Completion Date

The expected project completion is 2011. The last data collection will be done in the fall of 2010 and this, only for children recruited in 2009. Communication of the final results will be done in 2011-2012.

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# Contaminant exposure and emergence of cardio-vascular diseases in Inuit

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## Abstract

Some preliminary data suggests that the cardio-vascular system should be considered a potential target for Arctic contaminants. For example, work conducted in the Faeroe Islands in children suggests association between mercury exposure and blood pressure and heart variability which are known risk factors for cardiac health. Other studies conducted elsewhere suggest associations between mercury and heart diseases and between POPs and diabetes. These health conditions are rising in the Arctic and our hypothesis is that this rise is not only due to a changing life style but also to contaminant exposure. This project aims at investigating associations between exposure to Hg and POPs on the emergence of heart diseases and related risk factors using the 3 large epidemiologic studies conducted among adults and children in Nunavik since 1992. Since heart diseases represent the most important causes of death, even a slight negative impact on the cardiovascular system could be of greater public health relevance than any other health effects related to contaminant exposure.

## Résumé

Certaines données préliminaires indiquent que le système cardiovasculaire devrait être considéré comme une cible potentielle pour les contaminants présents dans l'Arctique. À titre d'exemple, le travail réalisé chez des enfants des îles Féroé indique une association entre l'exposition au mercure, la tension artérielle et la variabilité de la fréquence cardiaque, soit des facteurs de risque connus pour la santé du cœur. D'autres études effectuées ailleurs suggèrent des associations entre le mercure et les maladies du cœur et entre les polluants organiques persistants (POP) et le diabète. Ces problèmes de santé sont en croissance dans l'Arctique et notre hypothèse est que cette augmentation n'est pas uniquement due à un style de vie en évolution, mais aussi à l'exposition aux contaminants. Ce projet vise à examiner les associations qui existent entre l'exposition au mercure et aux POP et l'émergence des maladies du cœur et des facteurs de risque connexes par le biais de trois grandes études épidémiologiques réalisées chez des adultes et des enfants du Nunavik depuis 1992. Puisque les maladies du cœur représentent les causes les plus importantes de décès, même une

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répercussion légèrement négative sur le système cardiovasculaire pourrait être d'une très grande pertinence pour la santé publique que tout autre effet sur la santé lié à l'exposition à des contaminants.

## Objectives and Key Messages

The general objective of this project was to evaluate the role of contaminant exposure in the emergence of chronic diseases in the Arctic.

- Mercury exposure is associated with elevated blood pressure in adults and children even after controlling for major confounders. This was assessed in adults using data from 1992 and 2004 as well as in children (age 10).
- Mercury decreases heart rate variability in adults and children
- Mercury is a risk factor for atherosclerosis. This association is counterbalanced by n-3 fatty acids. This was based on 2004 Nunavik data in adults.
- PFOS interact with lipid metabolism: the work was restricted to the 2004 Nunavik data (on adults).

## Objectifs et messages clés

Ce projet avait pour objectif général d'évaluer le rôle de l'exposition aux contaminants dans l'émergence des maladies chroniques dans l'Arctique.

- L'exposition au mercure est associée à une tension artérielle élevée chez les adultes et les enfants, même après le contrôle des principales variables confusionnelles. Cette association a été évaluée chez des adultes d'après des données de 1992 et de 2004, de même que chez des enfants (âgés de 10 ans).
- Le mercure diminue la variabilité de la fréquence cardiaque chez les adultes et les enfants.
- Le mercure est un facteur de risque pour l'athérosclérose. Cette association est contrebalancée par les acides gras n-3. Ce constat est basé sur des données de 2004 chez des adultes du Nunavik.
- Le PFOS interagit avec le métabolisme des lipides : ce travail ne portait que sur des données de 2004 du Nunavik (chez des adultes).

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## Introduction

Many health endpoints such as child neurodevelopment or cardiovascular diseases have multi-factorial causes and environmental stressors can contribute in varying degrees to the aetiology of these diseases. Compared to the role that lifestyle (alcohol, smoking, drug abuse, etc.) and genetic factors play in the aetiology of most diseases, contaminants likely play a modest role. However, these environmental factors are highly preventable.

Our environment interacts with many functions in the body, among them the cardiovascular system. These interactions are often complex, sometimes also indirect and thus difficult to study. So far most of the information comes from epidemiological studies.

### Toxic metals and CVD

Most studies on the relationship between toxic metals and the risk for clinical cardiovascular disease has been on mercury exposure. The first studies reporting a relationship between methylmercury exposure and carotid intima-media thickness came from Finland (Salonen *et al.* 2000). Since that report, several other studies have been published, offering support to varying degrees. The same study group in a follow-up study showed that a high content of mercury in hair may be a risk factor for acute coronary events and cardiovascular disease (CVD), CHD, and all-cause mortality in middle-aged eastern Finnish men, and that mercury also may attenuate the protective effects of fish on cardiovascular health (Virtanen *et al.* 2005). The US Health Professionals' Follow-up Study

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(Yoshizawa *et al.* 2002) showed only a minimal overall risk.

### **Oxidative Stress**

Studies on Nunavik Inuit suggest that the consumption of marine products, a major source of n-3 polyunsaturated fatty acids (n-3 PUFA), is beneficial to cardiovascular health (Dewailly *et al.* 2001). Dewailly *et al.* concluded that the traditional Inuit diet was probably responsible for the low mortality rate from ischemic heart disease in this population (Dewailly *et al.* 2001). In Greenland, however, in spite of their high consumption of n-3 PUFA the Inuit have a similar prevalence and mortality of ischemic heart disease as European populations (Bjerregaard *et al.*, 2003; Jorgensen *et al.*, 2008) while their mortality from other cardiovascular diseases (stroke and other heart diseases) is significantly higher. This could be due to the presence of other active substances in the marine diet or to lifestyle factors such as smoking and decreased physical activity, which counteract the beneficial effect of n-3 PUFA. Fish and sea mammals consumed by the Inuit are also highly contaminated by methylmercury (MeHg) (Wagemann *et al.* 1996) and other potentially prooxidant contaminants such as polychlorinated biphenyls (PCB) (Dewailly *et al.* 1993; Muckle *et al.* 2001). Both MeHg (LeBel *et al.* 1990; Lund *et al.* 1993; Sarafian and Verity 1991; Yee and Choi 1994) and PCB (Ryu *et al.* 2003; Slim *et al.* 1999; Slim *et al.* 2000) are documented sources of oxidative stress. For example, PCB-153, the main PCB congener found in Inuit, was reported to induce concentration-dependent formation of reactive oxygen species (ROS) and death of cerebellar granule cells (Mariussen *et al.* 2002). The mean concentrations of PCB, MeHg and Se in Inuit from Salluit were respectively 16-18-fold, 10-14 fold and 8-15 fold higher than reported for reference Caucasian populations consuming little fish (Bélanger *et al.* 2006). The low risk of CVD observed in an Inuit population highly exposed to MeHg stands in sharp contrast with the increased risk of CVD and acute myocardial infarction associated with mercury exposure among Finnish men (Salonen *et al.* 1995; Virtanen *et al.* 2005). The potentially deleterious effects of MeHg on cardiovascular health have been at the centre of vigorous debates (Guallar *et al.* 2002; Salonen *et al.* 2000; Seppanen *et al.*

2004; Virtanen *et al.* 2005; Yoshizawa *et al.* 2002), and it would be of particular interest to find out which factors may account for the contrasting results obtained in different populations.

The Inuit are heavily exposed to potentially prooxidant contaminants such as methylmercury (MeHg) and polychlorinated biphenyls (PCB) through their traditional diet. This diet is also an abundant source of n-3 polyunsaturated fatty acids (n-3 PUFA), selenium and antioxidants, which might reduce cardiovascular risk. Although preliminary work reported that Inuit from Nunavik have low concentrations of plasma oxidized LDL (OxLDL) and elevated glutathione-related antioxidant defenses, the variance in OxLDL was predicted by PCB and blood glutathione, leaving the issue of contaminant-associated oxidative stress unresolved.

Preliminary results indicate that oxidized LDL was significantly lower in Inuit subjects than the normal Caucasian population (1.6X,  $p < 0.0001$ ), supporting the previous observation that omega-3 fatty acids and selenium could be strong protective factors for cardiovascular diseases among Inuit (Bélanger *et al.*, 2003).

That both mercury and selenium can modulate CHD risk is also suggested by observations in fish-eating coastal populations such as Inuit living in Arctic regions. Inuit consume large amounts of fish and marine mammals, and consequently receive large doses of mercury. Contrary to the situation in eastern Finland, however, the mortality rate from CHD in Inuit is low. Although, it was reported that omega-3 fatty acids are strong protective factors for cardiovascular diseases among Inuit (Dewailly *et al.*, 2001), the protection could also result from a high intake of selenium, (Bélanger *et al.*, 2003), through the consumption of traditional/country food such as muktuk (beluga and narwhal skin) and sea mammal liver which are rich in selenium.

### **Mercury and PON1 activity**

Methylmercury (MeHg) exposure has been linked to an increased risk of cardiovascular diseases, in particular myocardial infarction. Paraoxonase 1 (PON1) is an enzyme located in the high density lipoprotein (HDL) fraction of blood lipids. It metabolizes toxic oxidized lipids associated with

both low density lipoprotein (LDL) and HDL. MeHg and various metals (Cd, Co, Cu, Fe, Mn, Ni, Zn) have been shown to inhibit PON1 activity in vitro but the relation between metal exposure and PON1 activity has *not been studied in human populations*. Our hypothesis is that blood concentrations of mercury and other metals are linked to decreased serum PON1 activity and in turn to atherosclerosis.

### Mercury and blood pressure

Intake of mercury through food items from sea mammals and fish has been suggested to be involved in cardiovascular disease, and the relationship between mercury in blood and 24-h ambulatory blood pressure (BP) was studied in Greenland. Mercury was measured in blood and 24-h BP in four groups of healthy subjects: group 1, Danes living in Denmark consuming European food; group 2, Greenlanders living in Denmark consuming European food; group 3, Greenlanders living in Greenland consuming European food; and group 4, Greenlanders living in Greenland consuming mainly traditional Greenlandic food. Mercury in blood was highest in Greenlanders and increased when they lived in Greenland and consumed traditional Greenlandic food (group 1: 2.2 µg/L (median), group 2: 4.8 µg/L, group 3: 10.8 µg/L, and group 4: 24.9 µg/L microg/L). The 24-h BP was the same in all three groups of Greenlanders. However, 24-h diastolic BP was lower among Greenlanders than Danes (71 v 76 mm Hg,  $P < .000$ ) and 24-h pulse pressure was higher (54 v 50 mm Hg,  $P < .000$ ). Mercury in blood was significantly and positively correlated to pulse pressure ( $r = 0.272$ ,  $P < 0.01$ ). Pulse pressure was higher and diastolic BP was lower in Greenlanders than Danes. Pulse pressure increased with higher mercury content in the blood. Although genetic factors may be responsible to some extent for the difference in pulse pressure between Greenlanders and Danes, the present results seem to support the hypothesis that mercury intake from marine food is involved in cardiovascular disease (Pedersen *et al.*, 2005).

Blood pressure in childhood is an important determinant of hypertension risk later in life, and methylmercury exposure is a potential environmental risk factor. A birth cohort of 1,000 children from the Faeroe Islands was examined

for prenatal exposure to methylmercury, and at the age of 7, blood pressure, heart rate and heart rate variability were determined (Sorensen *et al.*, 1999). After adjustment for body weight, diastolic and systolic blood pressure increased by 13.9 mmHg [95% confidence limits (CL) = 7.4, 20.4] and 14.6 mmHg (95% CL = 8.3, 20.8), respectively, when cord blood mercury concentrations increased from 1 to 10 µg/L. Above this level, which corresponds to a current exposure limit, no further increase was seen. Birth weight acted as a modifier, with the mercury effect being stronger in children with lower birth weights. In boys, heart rate variability decreased with increasing mercury exposure, particularly from 1 to 10 µg/L cord blood, at which the variability was reduced by 47% (95% CL = 14%, 68%). These findings suggested that prenatal exposure to methylmercury might affect the development of cardiovascular homeostasis.

It should be pointed out that more sensitive health end-points other than neurotoxicity could be of greater relevance in understanding some areas of cardiovascular effects. For example, the low incidence of CHD in Inuit suggested to be due to the fatty acid composition of their diet could be attenuated by high mercury exposure since recent studies indicate that Hg can have a negative effect on the cardiovascular system. The reason is still unknown, but Hg may inhibit important antioxidative mechanisms in humans and could promote the peroxidation of unsaturated fatty acids such as DHA and DPA. Regarding cardiovascular toxicity at low-level methylmercury exposures, the first Faeroes cohort showed that blood pressure had a tendency to increase and the heart rate variability to decrease when prenatal mercury exposures increased in the low-dose range (Sorensen *et al.*, 1999). Alkyl-mercury poisoning is associated with increased blood pressure and children with mercury poisoning often have increased heart rate and blood pressure. Experimental evidence shows that methylmercury toxicity results in hypertension that remains many months after cessation of exposure.

### Mercury and heart rate variability

MeHg is well known for its toxic activity on the central nervous system (CNS) but recently, some authors have suggested that it can also interfere with the normal functioning of the cardiovascular



system (Guallar *et al.*, 2002; Sorensen *et al.*, 1999; Pedersen *et al.*, 2005; Oka *et al.*, 2002; Grandjean *et al.*, 2004). Some studies have reported an association between mercury and myocardial infarction (Guallar *et al.* 2002), elevated blood pressure (Sorensen *et al.* 1999; Pedersen *et al.* 2005), and reduced heart rate variability (HRV) (Sorensen *et al.* 1999; Oka *et al.*, 2002; Grandjean *et al.*, 2004). This latter effect reflects the balance between the cardiac parasympathetic and sympathetic activities of the autonomic nervous system (ANS) but information concerning the influence of MeHg on HRV is sparse. Reduced HRV can be a marker for an increased arrhythmia risk, causing among other things ventricular fibrillation which can lead to sudden cardiac death (Makikallio *et al.*, 2001; Kataoka *et al.*, 2004; Galinier *et al.* 2000; La Rovere *et al.*, 2003; Kruger *et al.* 2002). The decrease of HRV has also been associated with increased cardiac mortality and all causes of mortality (Seccareccia *et al.*, 2001, Tsuji *et al.*, 1996; Tsuji *et al.*, 1994).

The effect of MeHg on HRV has only been studied in subjects exposed *in utero* (Sorensen *et al.*, 1999; Oka *et al.*, 2002; Grandjean *et al.*, 2004). In a cohort study of 7-year-old children from the Faroe Islands, the coefficient of variance of the heart rate in boys decreased by 47% in a range of cord blood concentrations from 1 to 10 µg/L (5–50 nmol/L). This was depicted as an indication of parasympathetic nervous dysfunction in children exposed to low doses of MeHg during the prenatal period (Sorensen *et al.* 1999). This cohort was examined at 14 years old and a decrease of the coefficient of variation of heart rate of 2.7 % was detected as well as a decrease in parasympathetic and sympathetic activity of 6.7 % (Grandjean *et al.*, 2004).

On the other hand, Oka *et al.* (2002) conducted a case-control study to assess the chronic effect of exposure to high doses of MeHg on HRV in subjects suffering from fetal Minamata disease (FMD). Some HRV indices such as the RR interval (NN) and high frequency (HF) were lower in FMD patients while SDNN and CVRR tended to be lower in FMD group but this difference did not reach statistical significance.

Up to now, the influence of background mercury levels on HRV in adults environmentally exposed has not been studied. Preliminary work was conducted to assess the impact of MeHg levels on

HRV in an adult population of Nunavik. Several indices of HRV from the time and frequency domains were derived from a 2-hour Holter monitoring assessment. Simple linear regression was used to analyse the relation between mercury levels and Holter parameters. MeHg was weakly associated with low frequency in simple correlations but these associations need to be confirmed after full adjustments are made on confounding factors (Valera, pers. comm.).

### **POP exposure and metabolic effects in Inuit**

Legacy POPs found in the traditional Inuit diet include organochlorines (OCs), such as polychlorinated dibenzo *p*-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs), polychlorinated biphenyls (PCBs) and various chlorinated pesticides or industrial products. More recently, new POPs have emerged as potential environmental health threats to the Arctic people. Brominated flame retardants (BFR) such as polybrominated diphenyl ethers (PBDEs) (de Boer *et al.* 1998; Meirionyte *et al.* 1999; Noren *et al.* 2000; Ikononou *et al.* 2002) show similar physico-chemical and toxicological properties with PCBs, PCDDs and PCDFs (Darnerud *et al.* 2001; McDonald 2002; Zhou *et al.* 2002; Birnbaum *et al.* 2004; Gill *et al.* 2004), and have been measured in human milk (Pereg *et al.* 2003) and plasma (Dewailly *et al.* 2005) from Nunavik Inuit. Perfluorinated compounds (PFCs) such as perfluorooctanesulfonate (PFOS) and perfluorooctanoate (PFOA) are found in several wildlife species of the Arctic (Giesy *et al.* 2001; Kannan *et al.* 2001; Kannan *et al.* 2002; Tomy *et al.* 2004; Bossi *et al.* 2005; Smithwick *et al.* 2005; Smithwick *et al.* 2006), including some that are part of the traditional Inuit diet (Tittlemier *et al.* 2006) as well as in human blood from several populations (Kannan *et al.* 2004), including Nunavik Inuit (Dewailly *et al.* 2005; Dewailly *et al.* 2007). Contrarily to BFRs, PFCs are not structurally similar to OCs, but rather tend to resemble the structure of essential polyunsaturated fatty acids (PUFAs) that are abundant in the traditional Inuit diet, and have been shown to exert similar hypolipidemic effects in Nunavik Inuit (Pereg *et al.*, in preparation). However, despite their structural similarity with PUFAs, the inability of PFCs to be converted to Acyl-CoA esters and to enter lipids intermediary metabolism suggests different mechanisms of action that could possibly lead to

toxic effects (Vanden Heuvel *et al.* 2006). While POPs have been shown to interact with steroids (De Rosa *et al.* 1998; Sonnenschein *et al.* 1998; Soto *et al.* 1998; Davis *et al.* 2005; Turyk *et al.* 2006) and thyroid hormones (Koopman-Esseboom *et al.* 1994; Langer 2005), to affect neurodevelopment and behaviour (Jacobson *et al.* 1985; Gladen *et al.* 1988; Rogan *et al.* 1991; Jacobson *et al.* 1992; Huisman *et al.* 1995a; Huisman *et al.* 1995b; Jacobson *et al.* 1996; Koopman-Esseboom *et al.* 1996; Jacobson *et al.* 1997; Winneke *et al.* 1998; Despres *et al.* 2005; Saint-Amour *et al.* 2006) and to disrupt immune functions (Dewailly *et al.* 2000; Weisglas-Kuperus *et al.* 2000; Dallaire *et al.* 2004; Dallaire *et al.* 2006) in many populations including Nunavik Inuit, the higher risk of developing diabetes in relation to POPs exposure has recently received more attention from the scientific community and is now emerging as a potential additional adverse effect related to POPs exposure. Indeed, the association between POPs exposure and type II diabetes has been reported in several populations exposed through different routes, including dietary ones (Henriksen *et al.* 1997; Pesatori *et al.* 1998; Calvert *et al.* 1999; Cranmer *et al.* 2000; Longnecker *et al.* 2000; Longnecker *et al.* 2001; Steenland *et al.* 2001; Remillard *et al.* 2002; Fierens *et al.* 2003; Glynn *et al.* 2003; Rylander *et al.* 2005; Vasiliu *et al.* 2006), but this issue has not been investigated yet in the Inuit of Nunavik.

Although insufficient for risk assessment purposes, some preliminary data suggests that the cardiovascular system should be considered a potential target for Arctic contaminants. Even a slight negative impact on the cardiovascular system could be of greater public health relevance than any other health effects related to contaminant exposure.

## Preliminary Results

### a. Effect of PFOS on lipids.

In the Qanuippitaa study, women were slightly overrepresented in the population sample (54%) and mean age was 36.8 yrs with no significant gender difference ( $p = 0.286$ ). BMI showed a mean of 27.4 kg/m<sup>2</sup> with significant gender difference, and data ranged from 14 to 43. EPA and DHA

(expressed as percent total polyunsaturated fatty acids) were summed to approximate circulating n-3 polyunsaturated fatty acids (EPA+DHA) and were significantly higher in women than in men. Plasma lipids were compared with a double stratification for age and gender (Figure 1). Total cholesterol significantly increased with age in both genders and showed significant differences between genders only within the age groups of 18-24 yrs and 25-44 yrs, while the older age group did not show any significant gender difference. HDL showed significant gender differences within all three age groups but a significant increase in plasma levels was observed only in the older age group. LDL levels showed significant differences between all age groups in men, but for women, only the younger age group differed significantly from others. Significant gender differences were observed within the older age group only for LDL. For triglycerides, a significant gender difference could be observed only in people aged between 25-44 yrs. Regarding age dependency, only the younger age group differed significantly from the others.

### Exposure to PFOS

In all 739 participants, mean PFOS plasma concentration was  $18.3 \pm 0.42$  µg/L (geometric mean  $\pm$  SE). Levels were significantly higher in men ( $20.5 \pm 0.66$  µg/L) than in women ( $16.3 \pm 0.50$  µg/L) when no age stratification was considered ( $p = 0.001$ ). Figure 2 shows PFOS concentrations stratified by gender and age category (geometric means and 95% confidence intervals are shown). In the 18-24 yrs group, mean levels (geometric mean  $\pm$  SE) were significantly lower in women ( $12.1 \pm 0.6$  µg/L) than in men ( $16.7 \pm 0.9$  µg/L). The latter were very close to those observed in the 25-44 yrs group ( $14.0 \pm 0.6$  µg/L for women, and  $18.4 \pm 0.7$  µg/L for men) where the gender difference was also significant, but much lower than those observed in the older age group ( $28.0 \pm 2.1$  µg/L for women,  $31.2 \pm 2.4$  µg/L for men), where the gender difference was no longer significant. Although PFOS levels increased with age, levels were significantly higher only in people aged above 45 years old, in both genders.



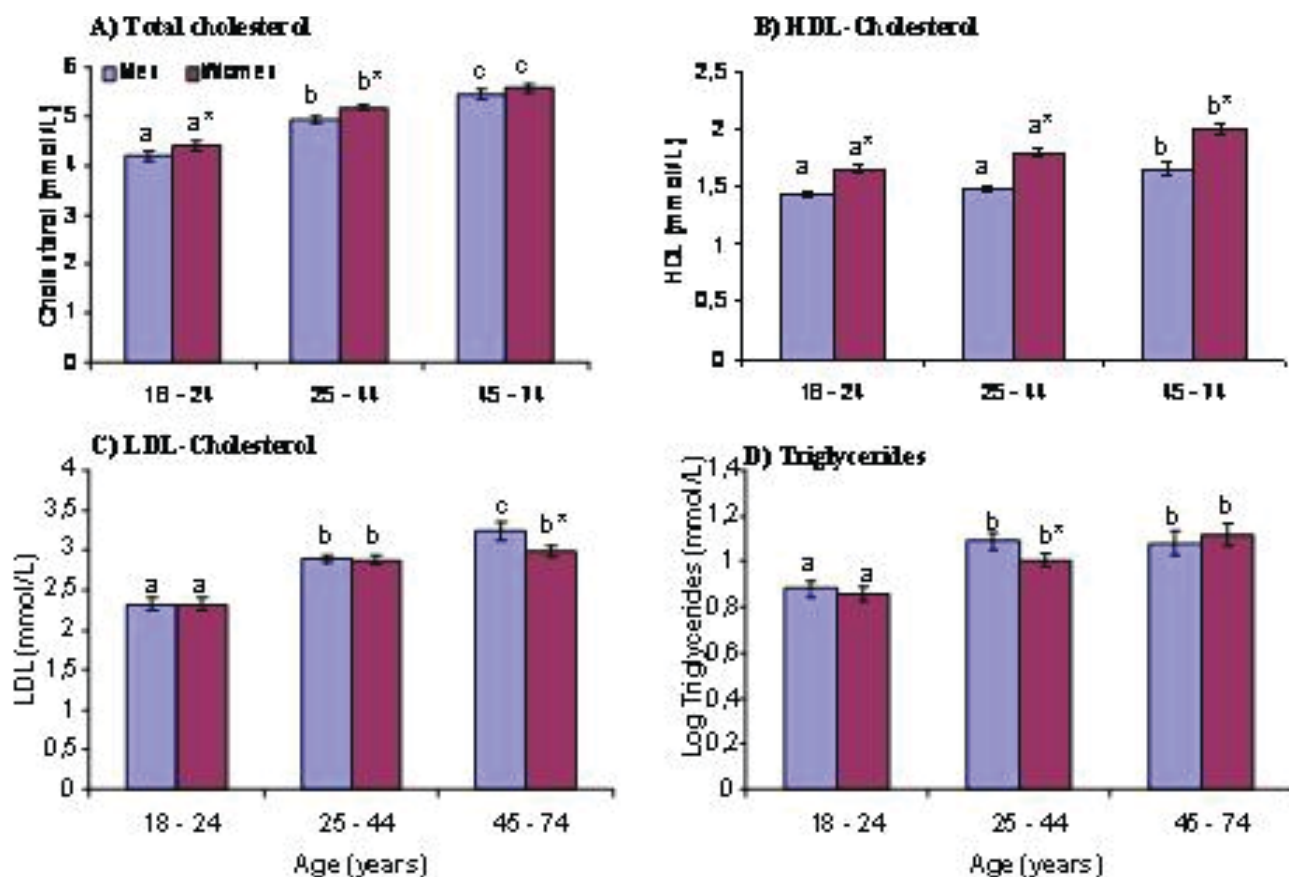


Figure 1. Blood lipid concentrations in Inuit of Nunavik, stratified by gender and age. A) total cholesterol; B) HDL-Cholesterol; C) LDL-Cholesterol; D) Triglycerides. Geometric means with 95% confidence intervals are presented for all, except LDL (arithmetic means and 95% CI). N are indicated inside each bars. Stars indicate statistically significant differences between genders within age group and letters indicate statistically significant differences among age groups within genders.

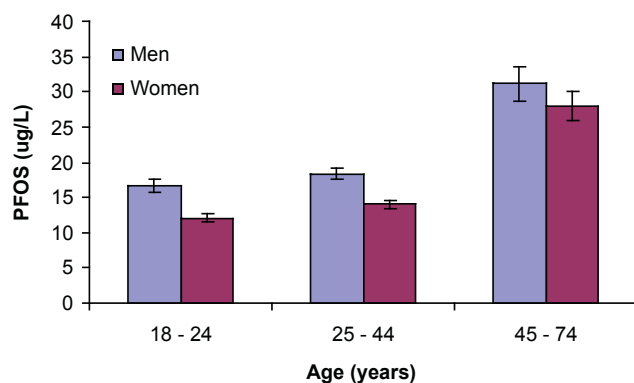


Figure 2. PFOS plasma levels in Inuit of Nunavik, stratified by gender and age (geometric means with 95% confidence intervals). Stars indicate statistically significant differences between genders within age group and letters indicate statistically significant differences among age groups within genders.

#### Association between blood lipids and PFOS exposure

Table 1 shows the results of multiple regression analyses testing the association between blood lipids and PFOS exposure. Triglycerides (TG) were negatively associated with PFOS plasma concentrations and HDL showed a significant positive association with PFOS plasma concentrations after adjusting the models for EPA+DHA, as well as for the interaction between gender and PFOS. Other plasma lipids seemed unaffected by PFOS plasma concentrations.

Fish consumption as well as sea mammal fat consumption were not confounders in the different models tested despite the fact that they are a common source of PFOS and LCPUFAs (data not shown). However, blood concentration of LCPUFAs (EPA+DHA) confounded the effect

**Table 1. Effect of PFOS on blood lipid levels in the population of Nunavik**

Blood lipids	N	R <sup>2</sup> model	Adjusted $\beta$ PFOS	<i>p</i> -value
Total cholesterol (mmol/L) <sup>a</sup>	666	0.21	0.0021	0.254
HDL (mmol/L) <sup>b</sup>	735	0.14	0.0065	< 0.001
TG (mmol/L) <sup>c</sup>	735	0.006	-0.0017	0.026
LDL (mmol/L) <sup>d</sup>	651	0.17	-0.0020	0.2420

<sup>a</sup> Multiple: correlation coefficients adjusted for PFOS, PFOS\*gender, standardised BMI, smoking status, insulinemia and  $\omega$ 3-polyunsaturated fatty acids;

<sup>b</sup> Multiple: correlation coefficients adjusted for PFOS, PFOS\*gender and  $\omega$ 3-polyunsaturated fatty acids;

<sup>c</sup> Multiple: correlation coefficients adjusted for PFOS, PFOS\*gender and  $\omega$ 3-polyunsaturated fatty acids;

<sup>d</sup> Multiple: correlation coefficients adjusted PFOS, age, BMI, smoking status, insulinemia and  $\omega$ 3-polyunsaturated fatty acids.

of PFOS on triglycerides and HDL, and was kept in the final regression models. The interaction term between PFOS plasma levels and gender also confounded the effect on triglycerides and HDL and was kept in these models.

After adjusting for confounders, the effect of PFOS on TG and HDL was not very strong, as expressed by the adjusted  $\beta$  coefficients for PFOS in both models. For HDL, an increase of 1  $\mu$ g/L in PFOS plasma concentration was associated with an increase of 0.0065 mmol/L HDL, while it was related to a decrease of 0.0017 mmol/L TG. This effect nevertheless remained significant and distinct from that attributable to LCPUFAs.

#### b. Other metabolic associations

In crude analyses, dioxin activity measured by the Calux assay was associated with triglycerides, LDL oxidation and blood pressure. Further analyses are needed to control for major confounders. (Table 2).

#### c. Mercury exposure and blood pressure in Inuit adults

A complete set of data was obtained from 806 adults of 18 years and older. We excluded 71 individuals who were treated for systemic hypertension during the data collection period. Thus, the final sample was composed of 735 individuals. Systolic blood pressure (SBP) and diastolic blood pressure (DBP) means were 117 mmHg (95%CI: 116-118) and 73 mmHg (95%CI: 72-74) respectively. Mercury was positively correlated with SBP ( $r = 0.11$ ;  $p = 0.0065$ ) and the association remained statistically significant ( $\beta = 2.14$ ;  $p = 0.0004$ ) after considering the effect of confounders [age, gender, obesity, cholesterol levels, insulin sensitivity, smoking, alcohol consumption, physical activity, socioeconomic status, eicosapentaenoic (EPA) and docosahexaenoic (DHA) acids and selenium blood levels]. Similarly, the association with DBP ( $\beta = 1.09$ ,  $p = 0.047$ ) was

**Table 2. PFOS, dioxin and PON-1 in relation to CVD parameters**

Pearson Partial Correlation Coefficients, N = 802								
	Glucose	Insulin	Triglycerides	Cholesterol Total/Hdl	LDL Ox.	HOMA_IR	BP Systolic	BP Diastolic
PFOS	0.00656 0.8530	0.04008 0.2572	-0.08646 0.0144	-0.05681 0.1081	-0.00005 0.9989	0.03331 0.3464	0.03081 0.3839	0.02156 0.5423
Dioxin	0.07935 0.0247	0.02035 0.5653	0.10266 0.0036	0.05163 0.1443	0.07343 0.0377	0.02969 0.4013	0.06843 0.0529	-0.06532 0.0646
Pon-1	-0.08521 0.0159	-0.09351 0.0081	-0.02585 0.4650	-0.10589 0.0027	0.06581 0.0627	-0.10577 0.0027	-0.01575 0.6563	0.01998 0.5723

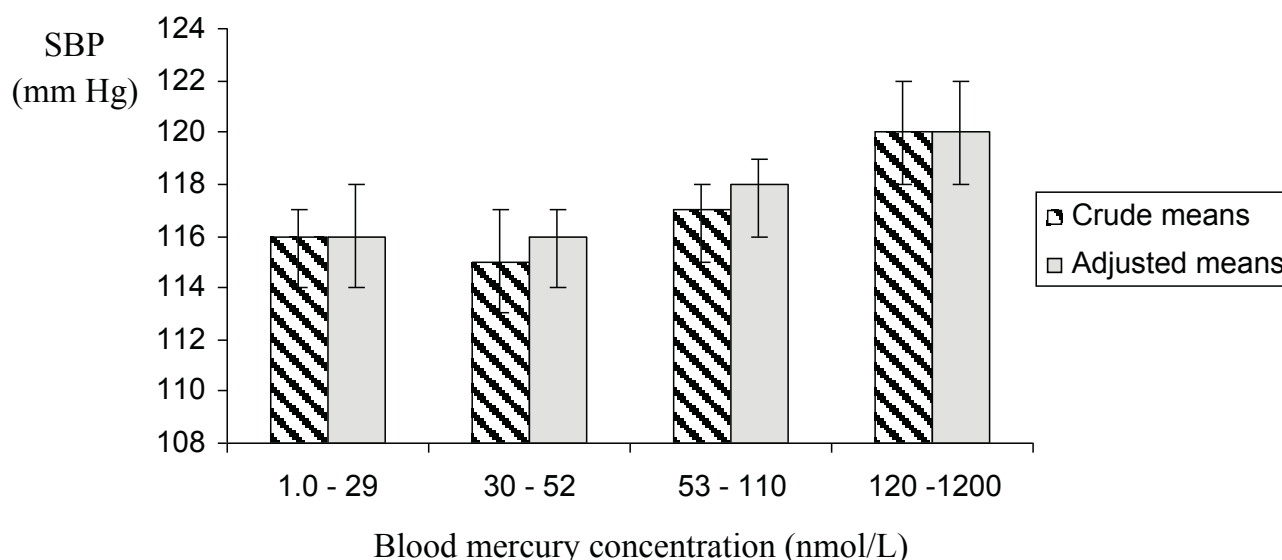


Figure 3. BP crude and adjusted means (95% confidence interval) per quartile of blood mercury concentration

statistically significant after adjusting for confounders. When SBP was compared by quartiles of mercury concentration, crude means were similar in quartiles 1, 2 and 3 but significantly lower than SBP in quartile 4 (Figure 3). After adjusting for confounders, means in quartiles 1 and 2 were significantly lower than SBP in quartile 4. These results suggest a deleterious impact of mercury on systolic blood pressure and diastolic blood pressure among Nunavik Inuit adults.

Significant differences in crude means are: means diff<sub>(1-4)</sub> = 4 ( $p = 0.01$ ), means diff<sub>(2-4)</sub> = 5 ( $p = 0.001$ ), means diff<sub>(3-4)</sub> = 3 ( $p = 0.046$ ). Significant differences in adjusted means are: means diff<sub>(1-4)</sub> = 4 ( $p = 0.02$ ), means diff<sub>(2-4)</sub> = 4 ( $p = 0.006$ ).

#### d. Mercury and heart rate variability: Inuit adults

In the context of the Qanuippitaa Survey, the parameters of heart rate variability (HRV) were derived from a 2-hour Holter monitoring. The ECG was recorded on tapes from 211, 40 year old and over participants that were sent to a centralized laboratory at the Quebec Heart Institute for analysis. To extract the HRV parameters, the ECG was analysed in order to identify the R waves which represent the heartbeats. Afterwards, the RR interval between consecutive heartbeats was calculated and the median RR was used to estimate parameters indicating the variability of the heartbeat. These parameters included time domain

indices which represent the parasympathetic activity (SDNN: standard deviation of RR intervals, SDANN: standard deviation of RR intervals measured over 5-minute periods) and frequency domain indices (LF: low frequency and HF: high frequency which is also an index of parasympathetic activity). Simple linear regression was used to analyse the relationship between mercury levels and Holter parameters while multiple regressions were carried out to control for confounders. In simple regression, mercury was associated with LF ( $\beta = -0.20$ ,  $p = 0.02$ ) and lower SDNN ( $\beta = -0.06$ ,  $p = 0.047$ ). After adjusting for confounders, a negative association was observed between mercury and SDANN ( $\beta = 0.086$ ,  $p = 0.026$ ) while the association with SDNN only approached the significance level ( $r = -0.056$ ,  $p = 0.10$ ). These results suggest a deleterious impact of mercury on HRV.

Further work will analyse these associations in Nunavut:

- Nunavut: 503 Holters in 2007
- Inuvialuit-Nunavut-Nunatsiavut: 399 Holters in 2008

#### e. HRV and blood pressure in children

A cohort of children from Nunavik that was analysed at birth and at 10 years old will be used to assess the impact of mercury exposure on blood pressure (BP) and heart rate variability (HRV). Presently, the data collection is in progress and

data on mercury concentration (hair and blood) as well as cardiac outcomes (BP and HRV) are available from 213 children. HRV indices, which represent the sympathetic and parasympathetic activities of the autonomic nervous system, were derived from a 2-hours Holter monitoring. Mean RR interval (interval between adjacent normal beats) was calculated and indices from the time and the frequency domains were derived. Time domain parameters included the standard deviation of R-R intervals (SDNN), standard deviation of the average R-R intervals calculated over 5-minute periods (SDANN), the square root of the mean squared differences of successive R-R intervals (rMSSD) and the proportion of interval differences of successive NN intervals > 50 ms (pNN50). Time domain parameters are indices of cardiac parasympathetic modulation. The analysis of the frequency domain included the low frequency (LF = 0.04-0.15 Hz) which represents both sympathetic and parasympathetic activity, and high frequency (HF = 0.15-0.40 Hz) which is an index of solely parasympathetic activity. The LF/HF ratio represents the sympatho-vagal balance.

Up to now, only simple analyses have been conducted and preliminary results revealed significant associations between mercury exposure and BP and HRV. Mercury in cord blood was negatively associated with diastolic blood pressure (DBP). Furthermore, blood mercury concentration at 10 years old was negatively associated with systolic blood pressure (SBP) while mercury in hair at 10 years old was negatively associated with SBP and DBP (Table 3). Regarding HRV, mercury in cord blood was negatively associated with LF and SDNN. Blood mercury at 10 years old was negatively associated with most of the HRV parameters (LF, HF, LF/HF, SDANN, rMSSD). Moreover, negative associations between mercury in hair and LF, HF, SDANN, and rMSSD were observed (Table 4). These associations suggest a negative impact of mercury on HRV. However, further analyses including potential confounders are needed before drawing a conclusion about a deleterious impact of mercury on HRV. These analyses will be conducted during summer 2009.

**Table 3. Simple correlation between mercury (at birth and at 10 years old) and blood pressure**

BP parameters	Mercury in cord blood <sup>&amp;</sup>	Mercury in blood <sup>&amp;</sup>	Mercury in hair <sup>&amp;</sup>
	Pearson r (p-value)	Pearson r (p-value)	Pearson r (p-value)
SBP (mmHg)	-0.12 (0.07)	-0.14 (0.04)	-0.15 (0.02)
DBP (mmHg)	-0.14 (0.04)	-0.11 (0.10)	-0.16 (0.02)

<sup>&</sup> Variable log-transformed

**Table 4. Simple correlation between mercury (at birth and at 10 years old) and HRV parameters**

HRV parameters <sup>&amp;</sup>	Mercury in cord blood <sup>&amp;</sup>	Mercury in blood at 10 years <sup>&amp;</sup>	Mercury in hair at 10 years <sup>&amp;</sup>
	r Pearson (p-value)	r Pearson (p-value)	r Pearson (p-value)
LF (ms <sup>2</sup> )	-0.13 (0.05)	-0.19 (0.005)	-0.13 (0.05)
HF (ms <sup>2</sup> )	-0.10 (0.15)	-0.18 (0.008)	-0.15 (0.03)
LF/HF	-0.004 (0.96)	-0.14 (0.045)	-0.12 (0.07)
SDNN (ms)	-0.15 (0.03)	-0.007 (0.92)	0.018 (0.76)
SDANN (ms)	-0.13 (0.07)	-0.21 (0.002)	-0.15 (0.04)
rMSSD (ms)	-0.10 (0.14)	-0.21 (0.002)	-0.14 (0.047)
pNN50 (%)	-0.08 (0.22)	-0.05 (0.50)	-0.07 (0.34)

<sup>&</sup> Variable log-transformed

#### f. Mercury and atherosclerosis in Inuit adults

1319 carotid examination were conducted in Nunavut, Inuvialuit and Nunatsiavut in 2007-2008

More specifically.

- Amundsen I (2004 / Nunavik)–  
284 participants / 284 read
- Amundsen II (2007 / Nunavut)–  
363 participants / 363 read
- Amundsen III (2008/ North West Territories)–  
63 participants / 0 read
- Amundsen III (2008 / Nunavut)–  
197 participants / 0 read
- Amundsen III (2008 / Labrador)–  
131 participants / 0 read

Preliminary results: In order to determine the sub-clinical atherosclerosis burden among Canadian Inuit, we measured the extracranial artery intimal to medial thickness (IMT) by high-resolution ultrasonography of a representative sample of Inuit older than 40 years. Out of three distinctive missions onboard the NGCC/CCGS Amundsen, we visited many communities in Nunavik, Nunavut, North West Territories and Labrador where a total of 1038 participants were scanned. As of now, approximately 60% of all images acquired have been analysed. The remaining analysis (n=391) consist of scans acquired during the mission in Nunavut in 2008 and are scheduled to be completed in late March 2009. Our preliminary analysis shows that the IMT of Inuit is thinner

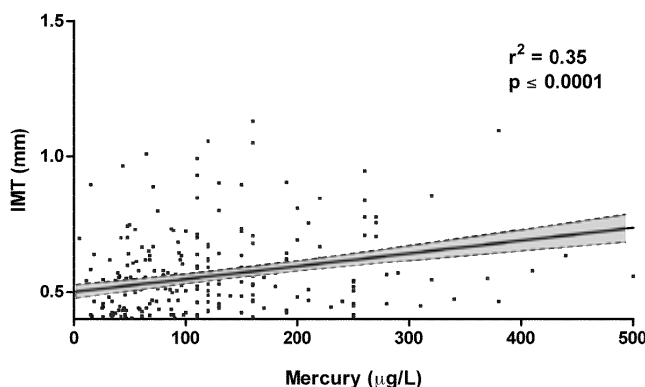


Figure 4. IMT and mercury

than their southernmost counterpart. Strikingly, cardio vascular disease risk factors such as diabetes, obesity and dyslipidemia that are known to be pro atherosclerotic did not impart IMT among Inuit. We are currently investigating possible explanations to our observations that includes a non associative cardiovascular protective effect to higher dietary intake of omega-3 from traditional marine food. What is most disturbing is the strong positive association of mercury to IMT. (Figure 4) This association disappears in multiple regressions when n-3 FA are added in the model. Perhaps higher concentration of mercury found in Inuit could outweigh the cardiovascular protective effect of omega-3 that has been otherwise well described in the literature. It is premature to foresee these hypothesis into statements since not all analysis of the carotid scans are completed and utmost statistical analysis of the data is underway.

#### g. Oxidative stress and mercury

Preliminary results show that oxidative stress as measured by LDL-ox concentration was low and was positively associated with blood mercury ( $p = 0.14$   $p < 0.0001$ ) and this correlation disappeared after adjustment for blood selenium.

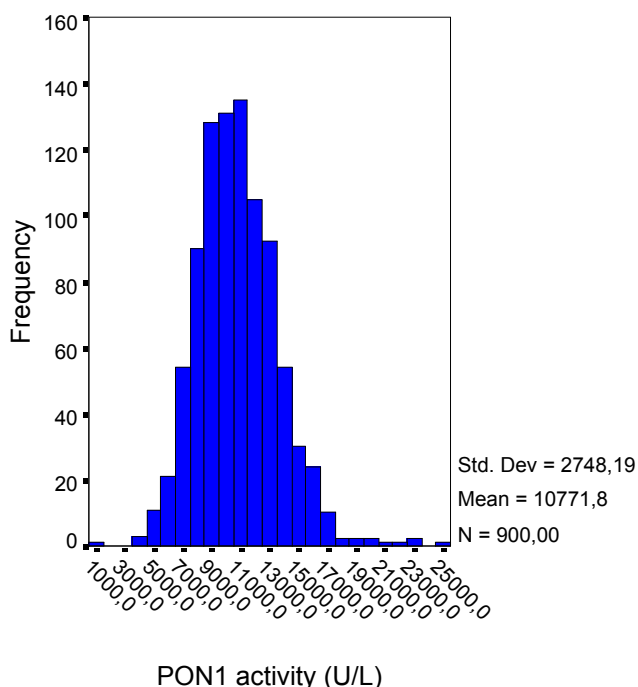


Figure 5: Distribution of PON 1 activity in Inuit

**Table 5. Multiple linear regression model of plasma PON1 activity in the Inuit population of Nunavik.**

	Unstandardized		Standardized Coefficients	t	Sig.
	B	Std. Error	Beta		
Age (years)	-44.336	8.778	-.219	-5.051	.000
Mercury (nmol/L)*	-809.584	325.239	-.135	-2.489	.013
Selenium (µmol/L)*	1634.311	605.515	.125	-2.699	.007
EPA (% total fatty acids)*	2688.968	464.874	.308	5.784	.000
HDL (nmol/L)*	5750.174	810.329	.250	7.096	.000
Alcohol consumption**	600.211	237.238	.086	2.530	.012

#### h. PON 1 activity.

Our results suggest that in the Inuit population, methylmercury exposure may have a slight inhibitory effect on plasma PON1 activity, which seems to be offset by selenium intake. PON-activity was negatively associated with blood glucose, insulin, high cholesterol/HDL-C ratio and HOMA=-IR (insulin resistance). (Table 5)

## Discussion and Conclusion

This study aimed at evaluating the hypolipidemic effect of PFOS exposure in the Inuit population, while taking account of the similar hypolipidemic effect exerted by LCPUFAs found in traditional food items. The results showed that triglyceride levels were negatively associated with PFOS plasma levels, while HDL cholesterol levels were positively associated, after adjustment for circulating EPA+DHA and interaction between gender and PFOS exposure.

For mercury, we found CVD effects mostly on HRV, and BP in adults and in children. Preliminary data also show association between mercury and atherosclerosis. Given the high prevalence of these health outcomes, the impact of mercury on health might be enormous.

Complementary analyses are ongoing to better evaluate the role of PON 1 activity, and the potential metabolic effects of POPs.

## Expected Project Completion Date

March 31<sup>st</sup> 2010 with partial funding from IPY and ArcticNet (grants on fatty acids) and Nasivvik (fellowships)

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## Presented and published materials

### a. Communications

Chateau-Degat, M.L., Dewailly, É., Noël, M., Ferland, A., Valera, B. Inflammatory markers and hypertension are associated beyond obesity? Joint Conference – 49th Cardiovascular Disease Epidemiology and Prevention and Nutrition, Physical Activity, and Metabolism, Tampa Bay, USA, 2009.



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- b. Publications**
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# Nunatsiavut Inuit Health Survey: “Kanuivit?”

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## Abstract

The Inuit Health Survey was successfully launched in 2007 with 18 coastal communities and over 1,200 participants in Nunavut participating. Nunatsiavut wished to participate and this project represents the Nunatsiavut component of the Inuit Health Survey 2008, which was also conducted in Nunavut and Inuvialuit Settlement Region. The Inuit Health Survey is compatible with similar surveys in Nunavik, Quebec and Greenland.

The Inuit Health Survey was made possible through funding by the Canadian Federal Program for International Polar Year, Canadian Institutes of Health Research, Health Canada, Indian and Northern Affairs Canada, and Government of Nunavut. The survey work in Nunatsiavut was guided through a steering committee with a memorandum of agreement for the long-term partnerships between Nunatsiavut Government and McGill University together with University of Toronto.

## Objectives

The specific objectives of the Nunatsiavut Inuit Health Survey are as follows:

- To assess dietary habits and nutrient status relevant to reproductive health and the emergence of chronic diseases;

## Résumé

L'Enquête sur la santé des Inuits (Inuit Health Survey – IHS) a été lancée avec succès en 2007. Plus de 1 200 personnes et 18 collectivités côtières y ont participé au Nunavut. Le Nunatsiavut souhaitait y participer. Le présent projet représente donc la composante du Nunatsiavut de l'Enquête de 2008, enquête qui s'est également tenue au Nunavut et dans la région désignée des Inuvialuit. L'IHS se compare à d'autres enquêtes similaires réalisées au Nunavik, au Québec et au Groenland.

L'IHS a été rendue possible grâce au financement du programme fédéral du Canada pour l'Année polaire internationale, des Instituts de recherche en santé du Canada, de Santé Canada, des Affaires indiennes et du Nord Canada et du gouvernement du Nunavut. Le travail d'enquête au Nunatsiavut a été orienté par le biais d'un comité de direction avec un protocole d'entente pour les partenariats à long terme entre le gouvernement du Nunatsiavut et l'Université McGill ainsi que l'Université de Toronto.

## Objectifs

Les objectifs précis de l'enquête de l'IHS au Nunatsiavut sont les suivants :

- Évaluer les habitudes alimentaires et l'état nutritif ayant trait à la santé reproductive et à l'émergence des maladies chroniques;

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- To determine the prevalence and risk factors for cardiovascular disease and type 2 diabetes mellitus in Inuit undergoing acculturation;
  - To evaluate social support and other determinants of resiliency and self-reported health;
  - To obtain permission to conduct a follow-up evaluation within 7 years to prospectively evaluate factors leading to progression of disease, including the emergence of pregnancy-related health complications and birth outcomes;
  - To utilize the cross-sectional health survey and the long-term follow-up data (cohort) to help prevent the emergence of chronic diseases among Inuit and to promote the reproductive health of women and their offspring.
- Déterminer la prévalence et les facteurs de risque des maladies cardiovasculaires et du diabète de type 2 chez les Inuits en cours d'acculturation;
  - Évaluer le soutien social et d'autres déterminants de la résilience et de l'état de santé déclaré par l'intéressé;
  - Obtenir la permission d'effectuer une évaluation de suivi dans les sept ans pour évaluer de façon prospective les facteurs menant à la progression de la maladie, y compris l'émergence de complications de santé liées à la grossesse et les issues des grossesses;
  - Utiliser l'enquête transversale sur la santé et les données de suivi à long terme (cohorte) pour aider à prévenir l'émergence des maladies chroniques chez les Inuits et pour promouvoir la santé reproductive des femmes et de leur descendance.
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## Introduction

The Nunatsiavut Inuit Health Survey was developed to incorporate contaminants research within a broader health research study and is a major study that will provide baseline data on the health status of Inuit in Nunatsiavut and also help explain how diets and contaminants affect the health of Inuit. Results of the study will provide constructive information to assist health professionals and policy makers at all levels of government in developing environmental health policies and empower Inuit in developing policies for their own jurisdictions as well as in making personal informed dietary choices.

## Progress to date

On October 2<sup>nd</sup> and 3<sup>rd</sup>, 2008 37 members of the Inuit Health Survey team travelled to Nain where, after a training period, they boarded the Canadian Coast Guard Ship Amundsen to conduct the Inuit Health Survey in Nain, Hopedale, Postville, Makkovik and Rigolet. They were joined by recruitment teams (nurses, community research assistants, and drivers) in each community. Participants were recruited from randomly selected

households. The study was explained to potential participants and after signing the consent form a clinic appointment on board the ship was scheduled. During the clinic appointment nurses took fasting blood samples, administered a glucose tolerance test, measured blood pressure and pulse, and took body composition measurements.

Participants also met with interviewers and answered asked questions about general health and well-being, tobacco use, mental health, alcohol and drug use, diet, physical activity and social support. The survey achieved recruitment of the target number of participants with a total of **two hundred and sixty five adults** seen on board the Amundsen. Individual result letters have been sent to participants.

## Capacity building in Nunatsiavut

Increased capacity at the community level was accomplished by employing **twenty two** community members, who received considerable training enabling them to make radio announcements, recruit participants, complete consent forms, ID charts, household questionnaires and schedule clinic appointments on board the Amundsen. In



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addition, members of the Nunatsiavut steering committee were involved in all stages of the research project.

## **Ongoing Activities**

- Completion of data entry of adult survey data collected in Nunatsiavut in 2008
- Completion of contaminant analyses of samples collected in Nunatsiavut in 2008
- Completion of analyses of various markers for participants in Nunatsiavut
- Preliminary reports for stakeholders
- Presentations at International Congress on Circumpolar Health Yellowknife 2009.

# ***Environmental Trends Related to Human Health and International Controls***





# Northern Contaminants Air Monitoring for Organic Pollutants and Data Interpretation

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## Abstract

Air monitoring of organic pollutants has been continuously conducted at Alert, Nunavut, Canada since 1992. To gain a better understanding of the transport of priority pollutants to the Arctic, the air concentrations of persistent organic pollutants (POPs), polybrominated diphenyl ethers (PBDEs) and polycyclic aromatic hydrocarbons (PAHs) measured at Alert were compared to those measured at other air monitoring stations under the Arctic Monitoring and Assessment Programme (AMAP) and the Integrated Atmospheric Deposition Network (IADN). Elevated concentrations of POPs, including *c*-chlordane, *p,p'*- and *o,p'*-DDE, measured in summer 2004 at Alert were related to biomass burning during the Yukon/Alaska forest fires followed by long-range transport to the Arctic. Relatively greater proportion of heavier PCB congeners in air at Alert may reflect the levelling off of lighter congeners as a result of environmental removal processes. The air concentrations and congener profiles of PBDEs, which are widely incorporated into industrial and commercial products to reduce fire hazards, measured between 2002 and 2004 were found to be

## Résumé

Depuis 1992, on surveille les polluants atmosphériques organiques de façon continue à Alert (Nunavut), au Canada. Afin de mieux comprendre le transport des polluants d'intérêt prioritaire vers l'Arctique, on a comparé les concentrations atmosphériques de polluants organiques persistants (POP), de polybromodiphényléther (PBDE) et d'hydrocarbures aromatiques polycycliques (HAP) mesurées à Alert à celles mesurées à d'autres stations de surveillance de la qualité de l'air dans le cadre du Programme de surveillance et d'évaluation de l'Arctique (PSEA) et dans le Réseau de mesure des dépôts atmosphériques (RIDA). Les fortes concentrations de POP, entre autres de chlordane (*c*), *p,p'*- et *o,p'*-DDE, mesurées à l'été 2004 à Alert ont été liées à la combustion de biomasse pendant les feux de forêt au Yukon et en Alaska, et étaient la conséquence d'un transport sur une grande distance, jusqu'en Arctique. Une proportion relativement plus grande de congénères de BPC lourds dans l'air à Alert pourrait être l'indication d'un plafonnement des congénères légers résultant de processus d'élimination dans l'environnement. On a constaté que les concentrations atmosphériques et les profils de

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similar at Alert and Point Petre, a rural site in the Great Lakes. Increasing BDE-209 air concentrations measured at Alert and other Great Lakes stations reflects the influence of the continued usage of the “deca-BDE” technical mixture. Significant temperature dependence in the summertime implies volatilization emissions of PBDEs at Alert. Meanwhile, episodic observations in the wintertime were likely associated with enhanced inputs through long-range transport during the Arctic Haze period. Different inter-annual variances between Point Petre and Alert further indicate that emissions from regions other than North America could also contribute to PBDEs in the Arctic. Strong seasonal variations of total PAHs (excluding naphthalene, acenaphthylene and acenaphthene) air concentrations were observed at 4 Arctic stations with higher concentrations during the winter. Principal component analysis shows that the PAH profile at Alert are slightly different from other sites; and the Arctic PAH concentrations from December to February are significantly higher than the other months. For these remote sites, differences in PAH profiles may reflect impact of local emissions.

## Key Messages

- A slight relative increase in heavier PCB congeners measured in air at Alert may indicate the levelling off of lighter congeners as a result of environmental removal processes. Increasing equivalent black carbon input to Alert in recent years may also enhance the transport of particle-bound heavier PCBs.
- Elevated concentrations of c-chlordane, *p,p'*- and *o,p'*-DDE measured in summer 2004 at Alert may be the result of biomass burning during the Yukon/Alaska forest fires followed by long-range transport to the Arctic.
- PBDE air concentrations measured at Alert and Point Petre, a rural site in the Great Lakes

congénères de PBDE, lesquels sont couramment utilisés dans des produits industriels et commerciaux visant à réduire les risques d'incendie, que l'on a mesurés entre 2002 et 2004 étaient comparables à Alert et à Point Petre, un site rural de la région des Grands Lacs. L'augmentation des concentrations atmosphériques de BDE-209 mesurées à Alert et à d'autres stations de la région des Grands Lacs reflète l'effet de l'utilisation continue du mélange technique « déca BDE ». Une forte dépendance des concentrations sur la température estivale est une indication de la volatilisation des émissions de PBDE à Alert. Les concentrations épisodiques observées l'hiver sont proprement liées à des apports accrus sous l'effet d'un transport à grande distance dans la période de brume arctique. Les différences des variances interannuelles entre Point Petre et Alert indiquent également que les émissions provenant de régions à l'extérieur de l'Amérique du Nord pourraient également contribuer à la présence de PBDE dans l'Arctique. On a observé de fortes variations saisonnières des concentrations atmosphériques de HAP totaux (à l'exclusion du naphthalène, de l'acenaphthylène et de l'acénaphthène) à quatre stations arctiques, les concentrations élevées étant observées durant l'hiver. L'analyse en composantes principales montre que le profil des HAP à Alert est légèrement différent de celui à d'autres sites, et que les concentrations de HAP dans l'Arctique de décembre à février sont nettement plus fortes que dans les autres mois. À ces sites éloignés, les différences des profils de HAP peuvent refléter l'effet des émissions locales.

## Messages clés

- Une légère augmentation relative des congénères de BPC lourds mesurée dans l'air à Alert pourrait être une indication de plafonnement des congénères légers sous l'effet de processus d'élimination dans l'environnement. L'augmentation de l'apport d'équivalent de carbone noir à Alert ces dernières années pourrait également accroître le transport de BPC lourds liés à des particules.
- Les fortes concentrations de chlordane (c), *p,p'*- et *o,p'*-DDE mesurées à l'été 2004 à Alert peuvent être le résultat de la combustion de biomasse durant les feux de forêt au Yukon et en Alaska et d'un transport sur une grande distance, jusqu'en Arctique.

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region, were found to be comparable. This indicates that Alert is affected by ongoing use of PBDEs around the world.

- Atmospheric PBDEs were found to be increasing over time at Alert. While no significant trends were observed for PBDEs measured at Point Petre, increasing BDE-209 air concentrations measured at Alert and other Great Lakes stations reflects the influence of the continued usage of the “deca-BDE” technical mixture.
- PAH air concentrations measured at 4 Arctic stations were higher in winter than in summer indicating the influence of space heating. Alert PAH air concentrations were generally lower than those measured at the other Arctic sites. Differences in PAH compound profiles reflect site-specific local emissions for these remote sites.
- Les concentrations atmosphériques de PBDE mesurées à Alert et à Point Petre, un site rural de la région des Grands Lacs, se sont révélées comparables, ce qui indique qu’Alert subit l’effet de l’utilisation continue de PBDE sur la planète.
- Les concentrations atmosphériques de PBDE ont augmenté avec le temps à Alert. Aucune tendance importante n’a été observée concernant les concentrations de PBDE mesurées à Point Petre, mais l’augmentation des concentrations atmosphériques de BDE-209 mesurées à Alert et à d’autres stations de la région des Grands Lacs sont le reflet de l’utilisation continue du mélange technique « déca BDE ».
- Les concentrations atmosphériques de HAP mesurées à quatre stations de l’Arctique étaient plus élevées l’hiver que l’été, ce qui est une indication de l’effet du chauffage des locaux. À Alert, les concentrations atmosphériques de HAP étaient généralement plus faibles que celles mesurées aux autres sites arctiques. Les différences entre les profils de composés d’HAP sont le reflet des émissions locales spécifiques à ces sites éloignés.

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## Objectives

1. To operate a major long-term trend measurement station at Alert, Nunavut (in operation since 1992), to contribute to future assessments by the Northern Contaminants Program and the Arctic Monitoring and Assessment Programme, and to advise Canadian negotiators in preparing contaminant control strategies.
2. To measure and understand the occurrence and trends of selected OCs, PAHs, and PCBs in the Arctic atmosphere and to determine whether concentrations are changing in response to national and international initiatives.
3. To develop new technologies for monitoring emerging chemicals, including current-use pesticides, perfluorinated compounds and brominated flame retardants, in air at Alert.
4. To provide insight into contaminant pathways (sources, transport, transformation, and removal processes) to the Arctic environment.
5. To enable validation of models of toxic chemicals in the Arctic environment with atmospheric observations.

## Introduction

Atmospheric measurements of organic pollutants, including polychlorinated biphenyls (PCBs), organochlorine pesticides (OCs) and polycyclic aromatic hydrocarbons (PAHs), have been conducted at Alert, Nunavut, since 1992. The atmosphere is considered the major and fastest route of transport of many priority pollutants to the remote Arctic. Comparing measurement results from Alert with those obtained from other air monitoring programs at different locations and regions may provide further insight into the atmospheric movement of pollutants. In this synopsis report, we have compared results from Alert

to those measured at other Arctic stations under the Arctic Monitoring and Assessment Programme (AMAP), as well as those measured under the Integrated Atmospheric Deposition Network (IADN) in the Great Lakes region. Three studies under this project have been summarized, namely a brief update on atmospheric persistent organic pollutants (POPs) measured at Alert and at Zeppelin Mountain (Svalbard/Norway, 78° 54' N, 11° 53' E, 1993-2006); a comparison of PBDE air concentrations measured at Alert versus that measured at a rural Great Lakes site; and a comparison of atmospheric PAHs measured at 4 AMAP stations in the Arctic. Potential factors that may influence the differences and similarities in atmospheric levels observed at the various stations are discussed.

## Activities in 2008/2009

### *Field and laboratory studies*

1. Regular atmospheric measurements of OCPs, PAHs, PCBs and PBDEs continued at Alert on a weekly basis, which includes field sampling, solvent extraction, clean-up, instrumental analysis, and data archive.
2. Since 2005, the Alert air samples have been screened for several new flame retardants, including Dechlorane Plus (DP), decabromodiphenylethane (DBDPE), tetrabromoxylene (TBX), pentabromotoluene (PBT) and bis-tribromophenoxyethane (BTBPE). PBT, BTBPE, syn- and anti-DP were frequently detected in the 2005 samples. Screening of these compounds will continue in samples collected from Alert.
3. Testing work was initiated at Alert in December 2005 for the atmospheric sampling of emerging chemicals, including currently used pesticides (CUPs) and perfluorinated compounds. Sampling generally occurred once per month from October to February and once every other week from March to September. Samples from December 2005 to August 2006 collected from Alert have shown the presence of a few CUPs in Arctic air. However, several target CUPs were found to be at relatively high levels in the field blanks. Several tests have been conducted over the last year to improve the extraction and cleanup methods.

Samples taken between Jun and Aug 2006 analyzed for perfluorinated compounds have shown similar concentrations of the fluorotelomer alcohols (FTOH) and perfluoroalkyl sulfonamido ethanols (PFASs) to those found on a cruise crossing the North Atlantic and Canadian Archipelago in July 2005 (Shoeib et al. ES&T 2006, 40, 7577-7583). Trichlorfon, trifluralin, dacthal, pentachloronitrobenzene (PCNB), chlorpyrifos and endosulfan I and II were frequently detectable in selected samples from 2006 and 2007.

4. A flowthrough passive air sampler has been deployed at Alert since October 2007 to test the use of this type of sampler under Arctic conditions. Monthly air samples have been taken with the flowthrough air sampler to compare with results obtained using the high volume air sampler. Samples are currently under analysis.

### *Related Work under IPY – the Intercontinental Atmospheric Transport of Anthropogenic Pollutants to the Arctic (INCATPA) project*

5. The Little Fox Lake station in Yukon was restarted in August 2007.
6. Two Research Affiliate Program (RAP) students, John Norman Westgate of the University of Toronto and Uwayemi Sofowote of McMaster University, have participated in two intensive air sampling excursions in August 2007 and again in September 2008 at Little Fox Lake. During these trips, 24-hour air samples were taken continuously for 14 days. Air samples were also taken simultaneously at Wudalianchi (48.6°N, 126.2°E), China, during the sampling period in August 2007. During the September 2008 sampling period, 24-hour air sampling occurred simultaneously at Fairbanks and Dillingham, Alaska. Four sampling stations in Asia, including Wudalianchi, Waliguan GAW and Xuencheng in China and Ba Vi in Vietnam, have started 24-hour continuous monitoring one week in advance and continued for 2 weeks. This potentially provides data with regard to pollutant transport across the Pacific.
7. As an in-kind contribution from Environment Canada, in July/August 2007, Prof. Frank Wania, a collaborator of IPY INCATPA, and



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RAP student, John Norman Westgate, collected soil samples along the Chilkoot Trail crossing Yukon and Alaska. Also, under an NSERC Strategic Grant, Mr. Westgate has performed passive air sampling for POPs on four mountains in B. C. at various heights on both the windward side and the leeward side to assess differences in atmospheric POPs. Results of these studies will be combined with the NCP and IPY air measurement results to assess the transport and deposition of organic pollutants to the western Canadian Arctic and sub-Arctic.

### **Data interpretation**

8. The air monitoring dataset for POPs at Alert and those collected for all satellite stations (namely Tagish, Little Fox Lake, Kinngait, Valkarkai, Amderma and Dunai) and NCP/NOAA collaborative station at Barrow, Alaska, have been included in the recent Western Europe and Other Groups (WEOG) first regional assessment report of the Global Monitoring Plan for Persistent Organic Pollutants under the Stockholm Convention Article 16 on Effectiveness Evaluation.
9. Similar to the WEOG report, all the above-mentioned data and those from the AMAP/EMEP stations of Pallas (Finland), Storhofdi (Iceland) and Zeppelin (Svalbard/Norway) have been included in a status review article for air monitoring of organic pollutants under AMAP to be published in the *Science of Total Environment*. This article will serve as a basis for the AMAP ministerial report to be presented in January 2009.
10. A paper titled "Air Concentrations of Polybrominated Diphenyl Ethers (PBDEs) in 2002-2004 at a Rural Site in the Great Lakes Region: Comparison to Measurements in the Arctic" has been submitted to *Environmental Pollution*.
11. In collaboration with Sara Becker and Crispin Halsall of Lancaster University (UK), a paper titled "Long-term trends in atmospheric concentrations of  $\alpha$ - and  $\gamma$ -HCH in the Arctic provide insight into the effects of legislation and climatic fluctuations on contaminant levels" has been published in *Atmospheric Environment* in 2008 (42: 8225-8233).
12. The effect of climate variation patterns on the transport of POPs to the Arctic continued to be investigated. Strong statistical correlations were observed between the atmospheric circulation teleconnection pattern of East Pacific-North Pacific (EP-NP) pattern and summertime air concentrations of hexachlorocyclohexanes (HCHs), chlordanes and polychlorinated biphenyls (PCBs) measured at Alert, Nunavut, Canada (1993-2001). It was found that climate conditions associated with the positive phase of EP-NP may enhance the transport of these compounds from the temperate region of eastern North America towards the Arctic. A short paper was presented at the Dioxin conference in Tokyo 2007. We are currently reanalyzing the results with the more recent dataset from Alert up to the end of 2005.

## **Results and Discussion**

### **Update on POPs air concentrations measured at Alert and Zeppelin**

For PCBs, tri-chlorinated congeners dominated the atmospheric profiles at both Alert and Zeppelin. This corresponds to the fact that tri-CBs were the most important PCB homologue produced historically (Breivik et al., 2002). At Alert, there was a slight decline in relative contribution of lower chlorinated CBs from the 1990s to early 2000s with a slight increase in contribution of higher chlorinated homologues, e.g. tetra- to octa-CBs. Since production of PCBs stopped in most industrialized countries during the 1970s and 80s, this shift in homologue profile may be considered as an indication that the air concentrations of lighter congeners are starting to level off as a result of environmental removal processes, e.g. photo- and bio-degradation. Increasing trends of equivalent black carbon (EBC) at Alert between 1998 and 2002 has been reported by Sharma et al. (2006). The increase in EBC may also enhance input of heavier PCB congeners which have higher tendencies to associate with particles. It was not possible to discern any temporal trends in homologue profile at the Zeppelin station as samples from the early 1990s were compromised by local PCB contamination.

Eckhardt et al. (2007) have attributed high air concentrations of PCBs measured at Zeppelin in

July 2004 and spring 2006 to boreal forest fires in Yukon/Alaska and agricultural fires in Eastern Europe, respectively. It was believed that biomass burning can enhance volatilization of previously deposited organic chemicals, such as PCBs, from soil. While no high air concentrations of PCBs were noted at Alert during the summer forest fire in Yukon/Alaska, elevated concentrations measured for *c*-chlordane, *p,p'*- and *o,p'*-DDE in summer 2004 at Alert and Zeppelin could possibly be the result of biomass burning. Figure 1 shows the air concentrations of black carbon (BC), *c*-chlordane and DDEs measured at Alert in 2004. Elevated air

concentrations of all three OCPs were observed during the haze period from end of April to early May. This is consistent with the fact that *c*-chlordane and *p,p'*-DDE showed elevated particle-bound concentrations during Arctic Haze seasons (December to April); with up to 80-90 % of the total concentrations found on the filter (associated with particles). During the Yukon/Alaska forest fire event from June to August 2004 (indicated on Figure 1 by blue box), EBC air concentrations were relatively low compared to the haze season. Therefore, higher concentrations of *c*-chlordane and DDEs observed during this time

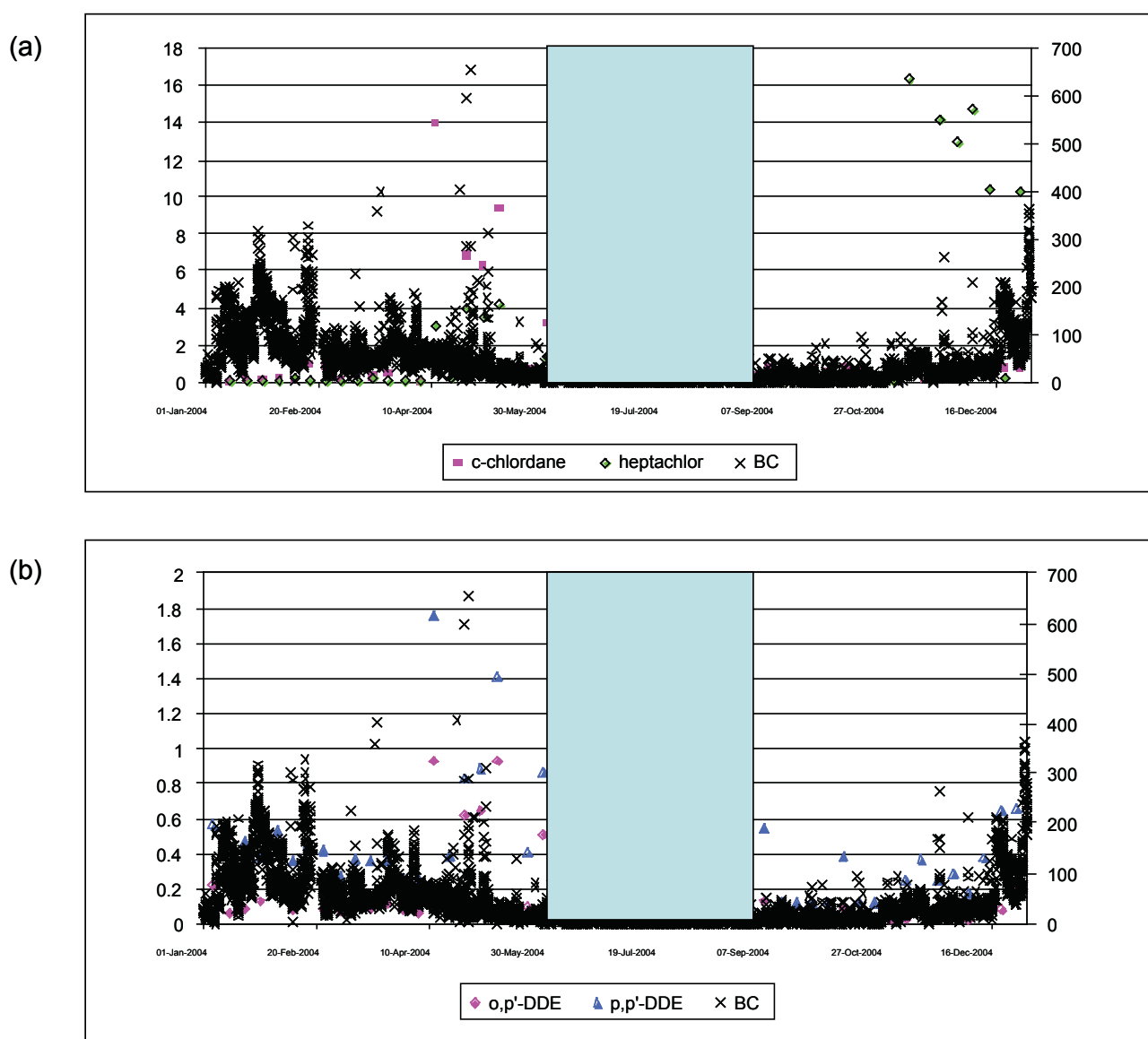


Figure 1. Air concentrations of BC (ng/m<sup>3</sup>) and (a) *c*-chlordane and heptachlor; and (b) *o,p'*- and *p,p'*-DDE in pg/m<sup>3</sup> measured at Alert in 2004. Blue rectangle indicates time period of Yukon/Alaska forest fire.

may be the result of enhanced evaporation of previously deposited pollutants from forest soil during biomass burning. Higher concentrations of other chlordane- and DDT-related compounds were not observed during this period. This could be because *c*-chlordane is relatively more stable and may remain in soil for prolonged period of time [degradation half-life of chlordanes in soil is over 20 years (Mattina et al., 1999)] and subject to evaporation when temperature increased during a forest fire event. DDTs were used in the Yukon/Alaska region before its ban. Previously deposited compounds would have been degraded to DDEs and DDDs.

***Comparison of air concentrations of PBDEs in 2002-2004 between Alert and a rural site in the Great Lakes***

The relative congener compositions of PBDEs measured at Alert and Point Petre, a rural site in the Great Lakes, were found to be similar. BDE-47, 99, and 209 were found as dominant congeners, and BDE-100, 153, 154, and 183 were commonly detected as well. These 7 PBDEs accounted for  $77 \pm 18\%$  (46-98%,  $n=144$ ) of  $\Sigma_{15}$ BDE at Alert, and  $88 \pm 9\%$  (62-99%,  $n=32$ ) of  $\Sigma_{15}$ BDE at Point Petre. Mean ratio of BDE-47/99 was  $1.3 \pm 0.66$  (0.16-3.9,  $n=97$ ) at Alert, and  $2.1 \pm 2.0$  (0.32-11,  $n=32$ ) at Point Petre. The BDE-47/99 ratio is 0.79 in DE-71 “penta-BDE” technical mixture and 0.96 in Bromkal 70-5DE mixture (La Guardia et al., 2006). The slightly higher BDE-47/99 ratios at Point Petre than at Alert were consistent with previous findings (Su et al., 2007). PBDEs are mostly emitted in mid-latitudes; therefore, the higher BDE-47/99 ratios in the High Arctic could be partially explained by faster degradation loss for BDE-47 than BDE-99 in the course of atmospheric transport (Su et al., 2007).

Concentrations of the 3 dominant congeners at Alert were fairly comparable to those at Point Petre. Long-term air monitoring under NCP and the Integrated Atmospheric Deposition Network (IADN) shows that air concentrations of PCBs were much higher in the Great Lakes than those in the Arctic (Sun et al., 2007; Hung et al., 2009). PCBs were banned from use over 3 decades ago, and their air concentrations decline continuously (Sun et al., 2007; Hung et al., 2009). In contrast, PBDEs are currently in use and primary emissions remain important. Air measurements at Point Petre

were largely influenced by primary emissions in the Great Lakes, and Alert reflects global background levels of PBDEs. A modelling study shows that PCB sources to the Arctic Circle could originate from Europe and Asia in addition to North America (Huang et al., 2007). Similar atmospheric transport pathways are applicable to PBDEs. Therefore, similar concentrations of PBDEs at Alert and Point Petre suggest that Alert is influenced by ongoing use of PBDEs around the world. At another Arctic site Nuuk in Greenland, air concentrations of BDE-47 and 99 in 2005 were about 4 times lower than those at Alert (Bossi et al. 2008), suggesting spatial variations in the Arctic. Since PBDE-impregnated consumer products have been widely used, potential influences from the nearby military base and the Global Atmospheric Watch laboratory at Alert cannot be ruled out completely (Hung et al., 2009).

Atmospheric BDE-47, 99 and 209 increased inter-annually at Alert in 2002-2005 with doubling times of 6.4, 12, and 3.5 years, respectively (Hung et al., 2009). Although usage of “penta-BDE” technical mixtures has decreased in North America, the increasing trends for BDE-47 and 99 at Alert may reflect influences of emissions from Europe and Asia. The doubling times of BDE-47 and 99 derived from 2002-2005 data were actually longer than those from 2002-2004 data at Alert, suggesting declining production/emissions of “penta-BDE” technical mixture globally (Hung et al., 2009). Venier and Hites (Venier and Hites, 2008) reported declining temporal trends for BDE-47 and 99 and an increasing trend for BDE-209 in 2003-2006 at five sites around the Great Lakes Basin. It was proposed that the declining trends of BDE-47 and 99 were related to the ban of “penta-BDE” technical mixture, whereas continuing usage of “deca-BDE” mixture contributes to the increasing trend of BDE-209 (Venier and Hites, 2008). Temporal trends were not derived at Point Petre because of the limited 32 measurements. Student’s *t*-test shows that inter-annual differences were not statistically significant for BDE-47, 99, or 209 ( $p < 0.01$ ). The increasing trend of BDE-209 at Alert and other sites in the Great Lakes reflects ongoing usage of “deca-BDE” technical mixture worldwide (Hung et al., 2009).

Significant temperature dependence of less brominated congeners was observed at Alert; indicating

volatilization of PBDEs in the summertime (Su et al., 2007). Similar seasonality of BDE-47 and 99 at Nuuk also highlights importance of volatilization emissions in the Arctic (Bossi et al., 2008). Meanwhile, episodically elevated concentrations in the wintertime suggest elevated inputs through long-range transport to Alert. Strong seasonality was apparent for atmospheric PBDEs in the Great Lakes region (Venier and Hites, 2008). On the other hand, temperature dependence of gaseous PBDEs was weak in general at Point Petre, implying that advective inputs probably played an important role at this rural area. The temperature dependence of PCBs was found to change along with time as a result of reduced primary emissions and increased secondary emissions. As primary emissions continue to decrease after the “penta-BDE” and “octa-BDE” mixtures have been banned for use, strong temperature dependence is foreseeable for less volatile congeners at Point Petre.

#### ***Atmospheric PAHs at circumpolar sites***

In urban areas, PAH concentrations are directly affected by proximity to sources and the intensity of emission, resulting in a strong urban-suburban-rural gradient (Motelay-Massei et al. 2005). Long-range atmospheric transport has been shown to deliver PAHs to the High Arctic (Patton et al. 1991).

The ultra high volume air samples taken at the four circumpolar sampling stations (Alert, Kinngait, Little Fox Lake and Barrow) on a weekly basis in 2000-2003 were analyzed for 20 PAHs (naphthalene, acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, retene, benzo(a)anthracene, chrysene, benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(e)pyrene, benzo(a)pyrene, indeno(1,2,3,cd)pyrene, dibenzo(a,h)anthracene, benzo(g,h,i)perylene, dibenzothiophene, perylene). Naphthalene, acenaphthylene and acenaphthene were excluded from the further analysis due to the breakthrough or higher method detection limits.

Figure 2 plots the monthly average total atmospheric PAH concentrations and their annual contribution at different sampling sites. It clearly shows that the total PAH levels in the Arctic are similar for most of cases. All the stations detected relatively higher total PAHs during winter time, probably as a result of space heating. However, air

concentrations also peaked in the summer between June and August at Kinngait, Little Fox Lake and Barrow, but not at Alert.

Principal component analysis was conducted for the monthly average concentrations for individual PAHs. Figure 3 shows the biplots based on the different grouping with panel A showing the centers for different sites and panel B indicating the centers for different months.

The figure clearly shows that the first two components extracted contain most of the information from the original data matrix, while PC1 most likely represents the concentration difference and PC2 distinguish the chemical fingerprint. The concentrations of dibenzothiophene, pyrene, retene, anthracene and phenanthrene show close correlations with each other, while the other chemicals also group together. Panel A indicates the variation between different sites, which shows relatively low PAH concentrations at Alert with lower percentage of anthracene (and correlating compounds). Such fingerprint difference could be due to the contribution from local emissions for these remote sites. Panel B demonstrates the seasonal variation for the whole region. In the Arctic, the PAH concentrations are significantly higher from December to February, while the heavier congeners, such as benzo(a)pyrene, also show higher percentages.

## **Conclusions**

Comparing air concentration measured at Alert with those observed in other locations of the Arctic as well as those in temperate locations, such as the Great Lakes, may provide further insight into the movement of priority pollutants. Differences and similarities in chemical profiles, spatial and temporal variations in air concentrations may reflect the sources of these pollutants. Here, we have related elevated air concentrations of legacy POPs, e.g. chlordane and DDE, in the Canadian High Arctic station of Alert to biomass burning during forest fires as a result of long-range transport. The similarity in air concentrations and congener profiles of PBDEs at Alert and at Point Petre in the Great Lakes has indicated that remote locations are still influenced by the ongoing use of PBDEs around the world. Increasing atmospheric trends of BDE-209 at Alert and other Great Lakes stations indicate the effect of continued usage

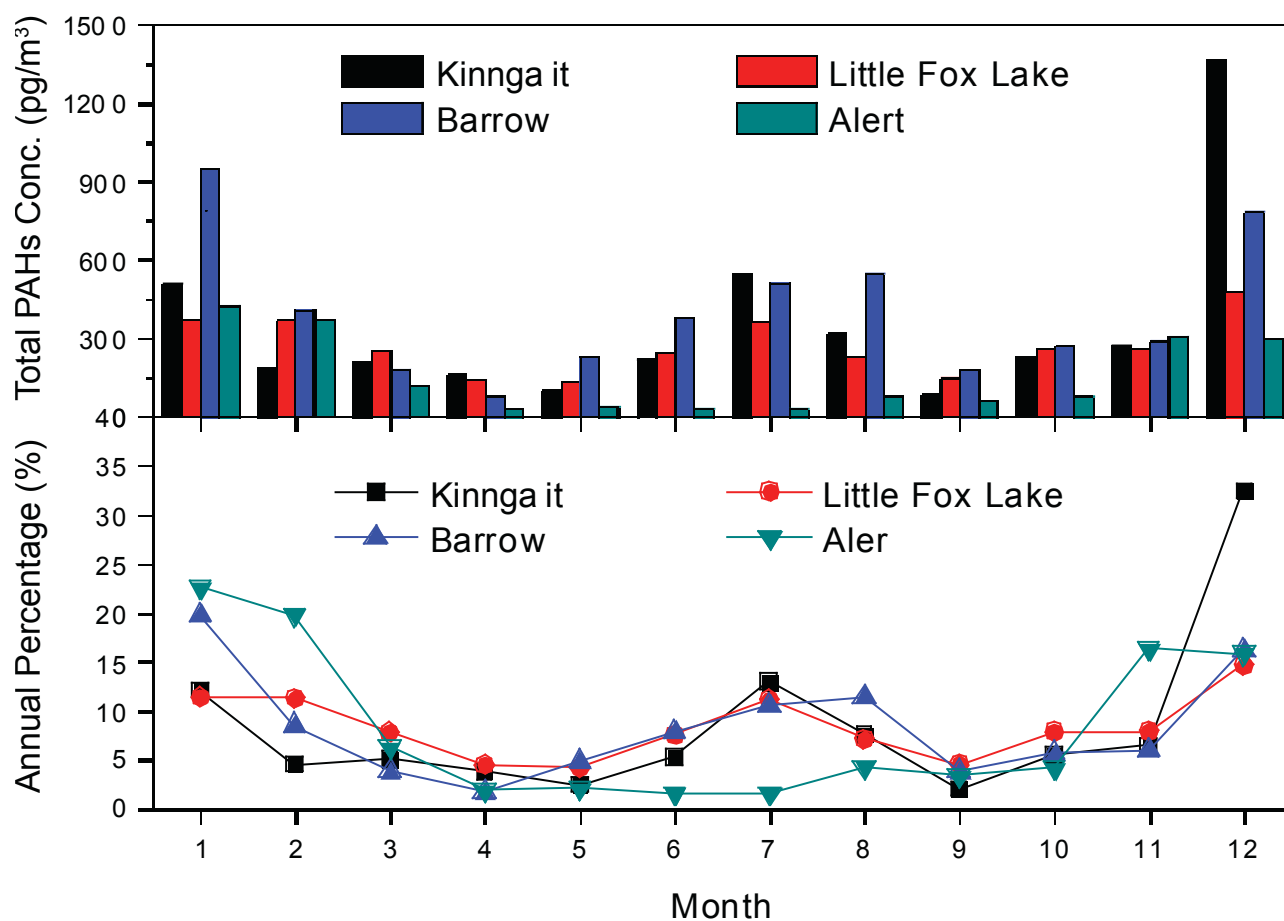


Figure 2. The monthly variation of total PAHs (17 compounds) and their annual percentage distribution at different sampling sites.

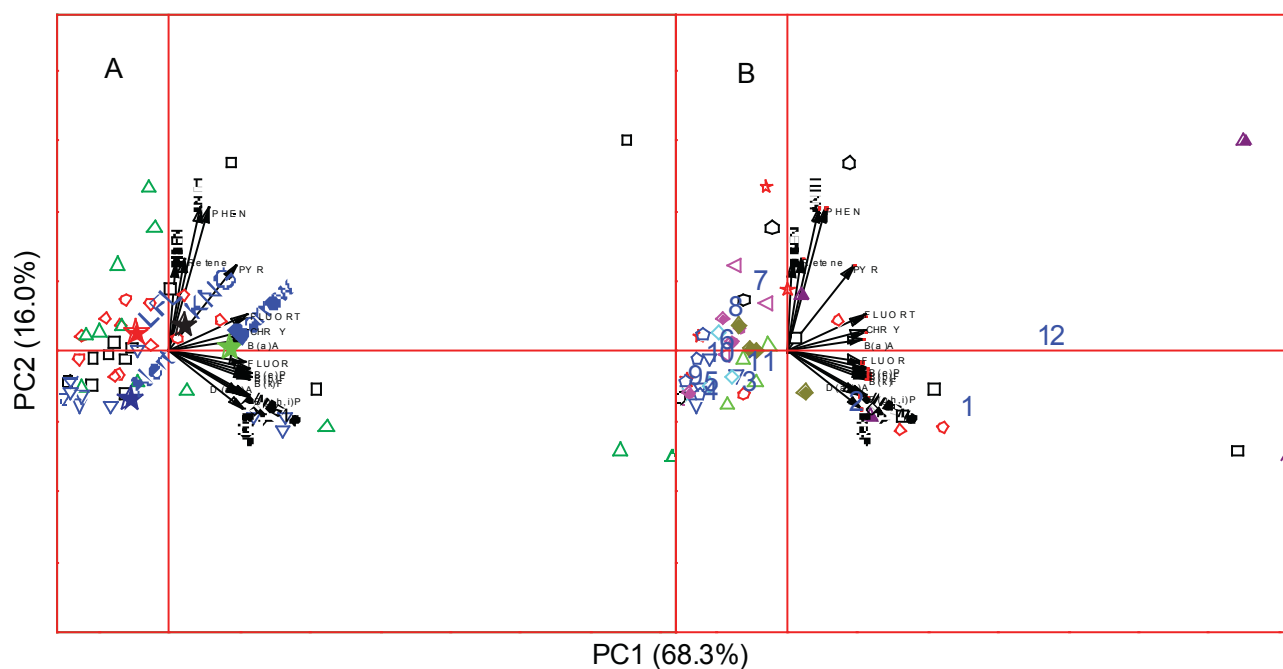


Figure 3. Principal component analysis biplots show the groups based on A), Sampling sites; B), Month.



of the “deca-BDE” technical mixture. PAH air concentrations were found to be generally higher in winter than in summer at 4 Arctic stations reflecting the overall influence of space heating in the circumpolar atmosphere, while differences in PAH compound profiles reflect site-specific local emissions.

## Acknowledgements

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## Expected Project Completion Date

On-going

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# Mercury Measurements at Alert

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## **Abstract**

Mercury (Hg) is a global priority pollutant and continues to be of concern in Arctic regions. The longest Arctic record of atmospheric mercury concentrations have been collected in the Canadian high Arctic at Alert, Nunavut. This time series shows distinct repeatable seasonal and annual patterns in the gaseous elemental mercury (GEM). Further analysis into the time trend of this data set has been undertaken. Research continued into understanding the processes, transfer and impact of atmospheric mercury depletion events (AMDEs) in the springtime to the Arctic by collecting atmospheric mercury species including Reactive Gaseous Mercury (RGM) and Particle Associated Mercury (PHg) and observing the behaviour of them over time. In the time trend analysis, the data was compared in 2 sets from 1995-2001 and from 2002-2007 on a monthly basis and has shown that all months indicate that there is a decrease in the GEM concentration at Alert, with

## **Résumé**

Le mercure (Hg) est un polluant planétaire d'intérêt prioritaire qui reste préoccupant dans les régions arctiques. Le registre arctique le plus ancien des concentrations de mercure dans l'atmosphère est celui tenu à Alert (Nunavut), dans l'Extrême Arctique canadien. Cette série chronologique révèle l'existence de régimes saisonniers et annuels répétés distincts concernant le mercure élémentaire gazeux (MEG). On a entrepris une analyse plus approfondie de la tendance temporelle de cet ensemble de données. On a poursuivi les recherches pour mieux comprendre les processus, les transferts et les effets liés aux amenuisements des concentrations atmosphériques de mercure (« AMDE ») qui surviennent au printemps dans l'Arctique en recueillant des spécimens de mercure atmosphérique, entre autres de mercure gazeux réactif et de mercure associé à des particules, et en observant comment ils se comportent avec le temps. Dans l'analyse des tendances temporelles, on a comparé sur une base

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the exception of May. This finding is interesting as it contradicts our current predictive understanding of atmospheric mercury processes in the spring. Analysis of the springtime data showed a strong relationship between air temperature and frequency of AMDEs. GEM measurements continued to be collected in the Yukon at Little Fox Lake and the progress on those measurements will be reported.

## Key Messages

- Fourteen years of atmospheric mercury measurements have been made at Alert, Nunavut. This data will be used to establish trends of atmospheric mercury in the Canadian high Arctic
- 2 years of atmospheric mercury measurements have been collected at Little Fox Lake, Yukon to establish baseline levels and the impact of long range transport from the Pacific Rim to this area.
- In collaboration with IPY efforts, intensive studies of mercury depletion/deposition episodes and their impact on the Arctic environment were undertaken
- While studies continue to further understand the processes driving mercury depletion events, the cause, effects and implications of these events are still pending.

mensuelle les données de deux ensembles couvrant les périodes 1995-2001 et 2002-2007; l'analyse montre à chaque mois une baisse de la concentration de MEG à Alert, sauf au mois de mai. Ce résultat est intéressant parce qu'il est en contradiction avec nos prévisions actuelles relativement aux processus atmosphériques printaniers du mercure. L'analyse des données de printemps montre un lien étroit entre la température de l'air et la fréquence des AMDE. On continue de mesurer les concentrations de MEG à Little Fox Lake (Yukon) et les résultats seront publiés.

## Messages clés

- On a mesuré les concentrations de mercure dans l'atmosphère durant 14 ans à Alert (Nunavut). Ces données seront utilisées pour établir les tendances du mercure atmosphérique dans l'Extrême Arctique canadien.
- On a recueilli des mesures des concentrations atmosphériques de mercure effectuées durant une période de deux ans à Little Fox (Yukon) afin d'établir des niveaux de référence et l'impact du transport à grande distance, du littoral du Pacifique jusque dans cette région.
- En collaboration avec l'API, on a entrepris des études intensives des épisodes d'amenuisement des concentrations de mercure et des dépôts de mercure ainsi que leur impact sur l'environnement arctique.
- Tandis que les travaux continuent d'éclairer les processus moteurs des épisodes d'amenuisement des concentrations de mercure, la cause, les effets et les implications de ces épisodes restent à déterminer.

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## Objectives

The objectives of this project are to establish long term baseline concentrations of mercury in the Arctic atmosphere and to study the behaviour of mercury in the Canadian high Arctic. By collecting this information on concentrations of atmospheric mercury, temporal variability, transport events and trends can be established. This information will be crucial in the development of Canadian strategies for national and international pollution control objectives. Through the NCP, the transport of

atmospheric mercury to the Arctic, the cycling of mercury in the atmosphere and the subsequent deposition of mercury from the atmosphere to the arctic environment has been studied at Alert since 1995. This long term record is advantageous to elucidate changes to and properties of the chemical and physical aspects of atmospheric mercury depletion events (AMDEs) after polar sunrise and the resulting potential link to enhanced Hg concentrations in the Arctic environment. Understanding these processes will help us to

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predict the effects that a rapidly changing Arctic climate will have on mercury deposition. More recently, the impact of mercury emissions from areas in the Pacific Rim to the Canadian western Arctic have become a concern. To address this, measurements have begun in the Yukon to measure the transport of mercury to this area.

## Introduction

Mercury (Hg) continues to be a priority pollutant of concern in Arctic regions. This project, within the NCP, provides long term data on the temporal trends and contributes to understanding the spatial variability of mercury in the High Arctic air as well as information concerning the behaviour of Hg that may impact the pristine Arctic. The change of the global atmospheric pool of Hg over time and the resulting concentration levels in particular regions are poorly defined. Thus, areas like the Arctic are a good place to assess such changes. Further, with global climate change expected to occur at a rapid pace in Arctic regions, the atmospheric dynamics and the impacts of pollutants such as Hg to this environment have to be well understood. Pollution of Hg in the Arctic has mainly occurred after industrialisation (Steffen et al., 2008). While European and North American emissions of gaseous elemental mercury (GEM) have decreased since 1995, emissions in other regions such as Asia and Africa have increased (Pacyna et al., 2006). Circulation patterns show that air masses originating in Asia can enter the Canadian Arctic (Dastoor and Larocque, 2004) and thus the increase in Asian emissions is particularly important to the Canadian north. It has been established by modellers that the Little Fox Lake site in the Yukon is an ideal location to measure such input from these sources (Dastoor and Durnford, 2008).

Annual time series of GEM have been produced and show repetitive distinct seasonal cycling of this pollutant. Through analysis of this annual cycling, it was discovered that a substantial amount of reactive mercury is present in the air and on particles during the spring time when levels of GEM in the air are very low (Schroeder et al., 1998; Steffen et al., 2003). This shift in mercury species in the air is called an atmospheric mercury depletion event (AMDE). A portion of these reactive mercury species remain in the air

while a large amount is deposited onto the snow and ice surfaces (Ariya et al., 2004). It is likely that this conversion of mercury (and subsequent deposition) after polar sunrise may provide a pathway by which these more reactive and potentially bio-available mercury species are introduced into the Arctic environment (Lu et al., 2001). These data (Canadian and other) will provide important information on the atmospheric transport, transformation and deposition processes of this priority pollutant throughout the Polar Regions.

Three Canadian IPY funded programs are directly linked with this ongoing NCP mercury research including the Intercontinental Atmospheric Transport of Anthropogenic Pollutants to the Arctic (INCATPA), Ocean Atmosphere Sea Ice and Snow pack (OASIS) -Canada and Circumpolar Flaw Leas System Study (CFL). These programs compliment and are linked to the ongoing Alert research program by contributing a historical record of atmospheric mercury as well as processes research. Both Alert and Little Fox Lake mercury measurements offer site locations for all three of these IPY programs.

## Activities in 2008/2009

Ground-based continuous atmospheric measurements of GEM, Reactive Gaseous Mercury (RGM) and Particulate Mercury (PHg) continued at Alert. RGM and PHg concentrations were collected and the data has been quality assured. The GEM data from Alert for 2008/2009 has been collected but this year found many instrumental problems which has resulted in a less complete data set than in previous years. Thus, at the time of writing, the quality control of this data has not been finalised as we are still trying to understand the impacts of the instrumental malfunction. Atmospheric measurements of Hg at Little Fox Lake continued as well but experienced similar instrumental malfunctions. At the time of writing the QCing of the data was still in process. Snow samples continued to be collected on weekly samples (ground) and on a per event basis (table). Due to ongoing lab construction, the samples collected in 2008 have not yet been analysed and thus we have no new data to report this FY. The mercury team also participated in 3 IPY funded/field intensive projects (INCATPA, OASIS-Canada and CFL) in FY 2008-2009 that directly relate to this NCP program.

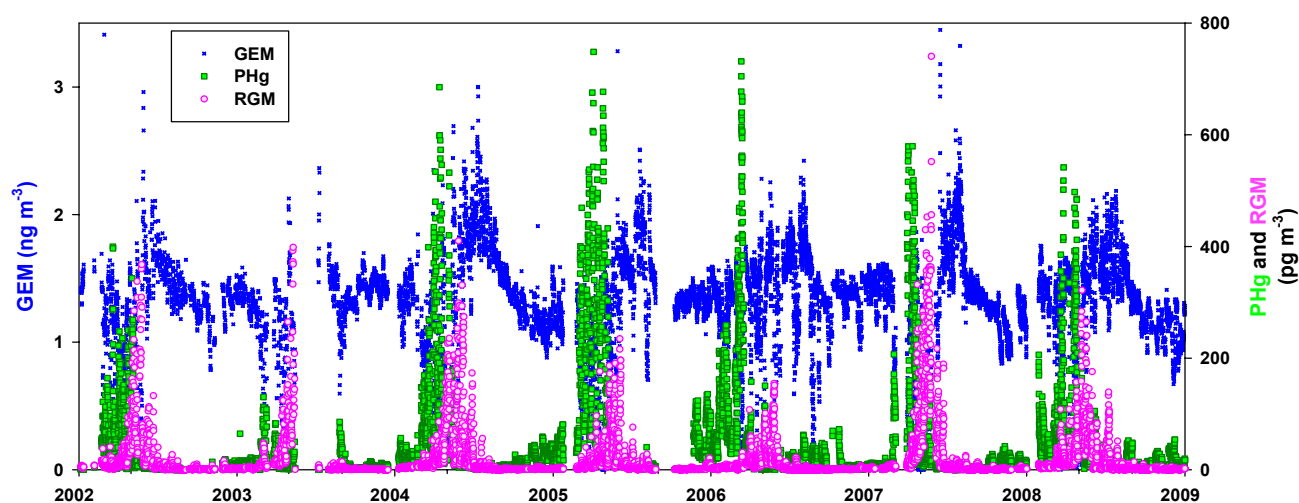


Figure 1. Atmospheric mercury speciation data from Alert 2002-2008.

## Results

Figure 1 shows the atmospheric speciation data from Alert from 2001 to 2008. Figure 2 shows the

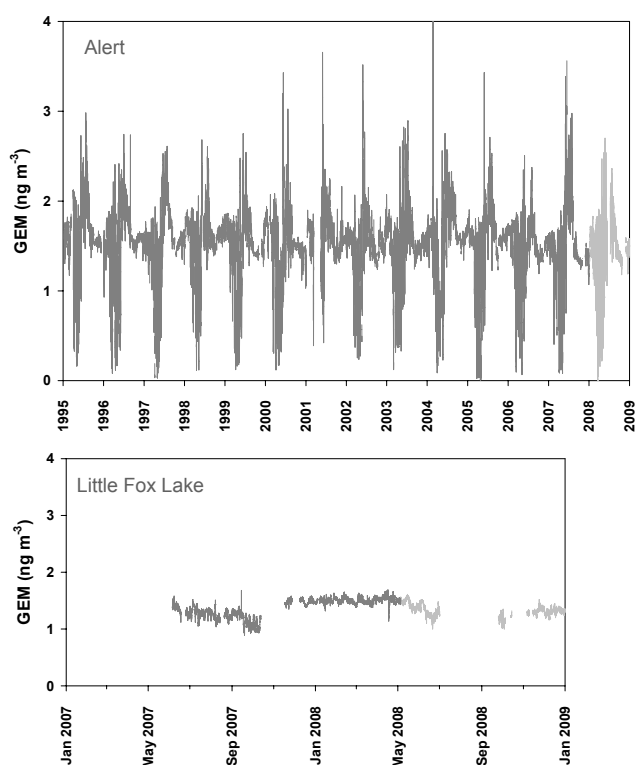


Figure 2. Atmospheric measurements of gaseous elemental mercury (GEM) at Alert, Nunavut (top) and Little Fox Lake, Yukon (bottom). Dark gray lines indicate quality assured data and light gray lines indicate non quality assured data.

preliminary GEM measurements at Alert from 1995 to 2008 and Little Fox Lake from 2007 to 2008. The data has been quality assured to the end of 2007 and submitted to the NATChem database and AMAP. Trend analysis continued on 13 years of GEM data from Alert. Some new results showing the change in monthly GEM concentrations between 1995-2001 and 2002-2007 are revealed in Figure 3. A further detailed analysis

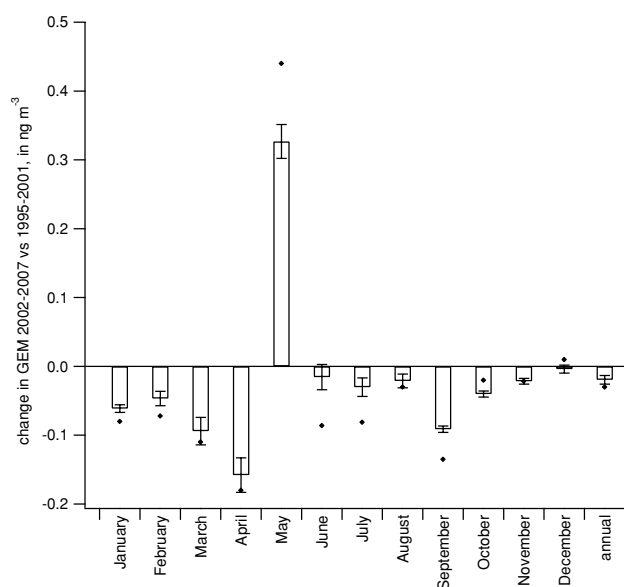


Figure 3: Change in mean (bars) and median (diamonds) gaseous elemental mercury (GEM) concentration at Alert, Nunavut from 1995-2001 to 2002-2007, pooling data from all months with >50% data completeness. Error bars show 95% confidence limits on the mean.

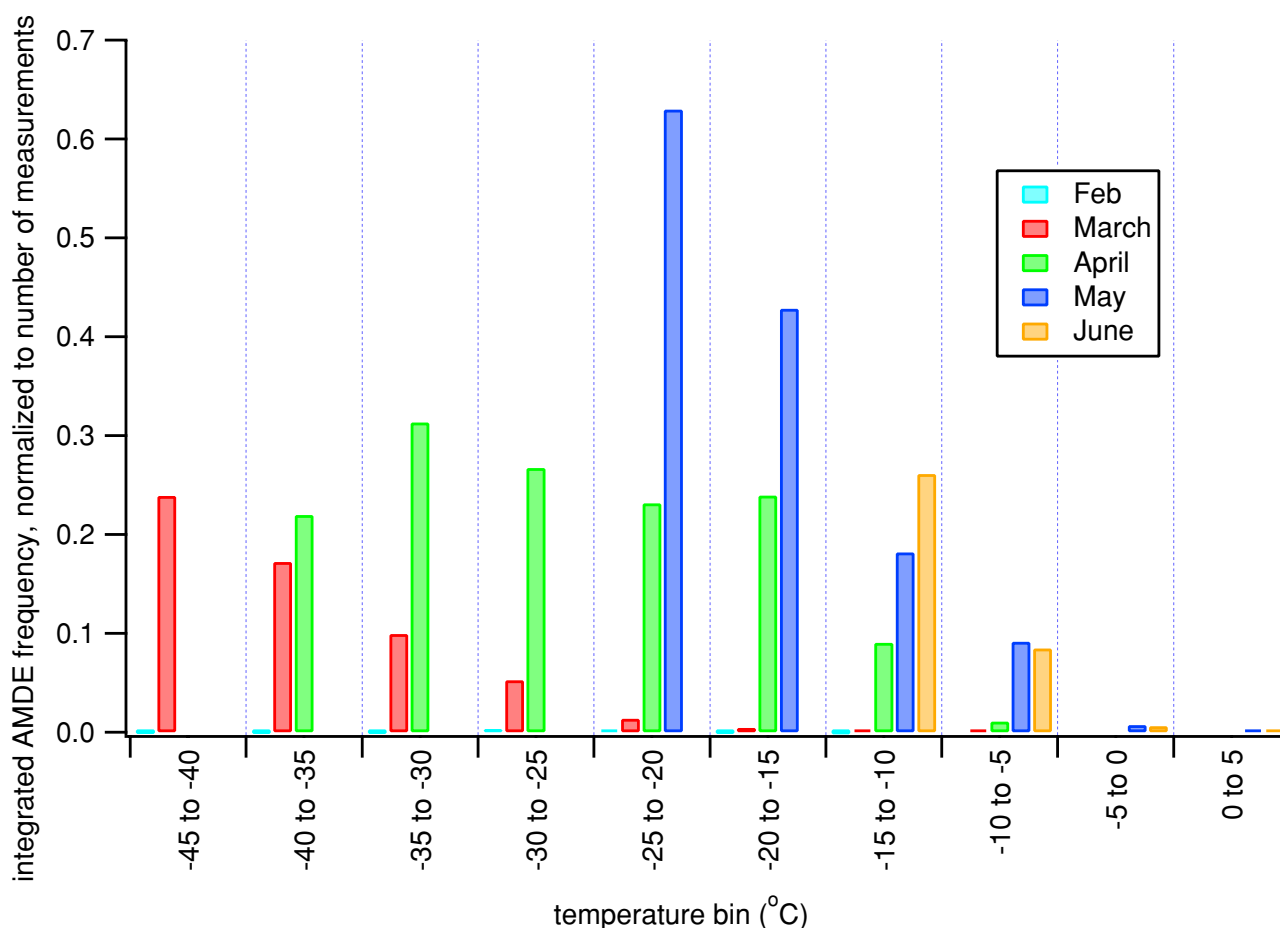


Figure 4. Temperature dependence of atmospheric mercury depletion events at Alert, Nunavut, broken down by month.

of the chemistry and dynamics of atmospheric mercury depletion events (AMDEs) was undertaken. Correlations between AMDEs and meteorological parameters are shown in Figure 4.

## Discussion and Conclusions

### *Atmospheric mercury trends:*

Fourteen years of GEM concentration data from Alert have now been collected. This is an impressive record of long term atmospheric mercury in the Arctic. Previous analyses of the data have shown no trends in the annual data set (Steffen et al., 2003; Temme et al., 2007). However, this data exhibits such seasonal fluctuations that a more robust investigation of the data needs to be undertaken in order to assess any true changes with time. Thus, trends for each month were evaluated to observe month-by-month changes. The data were split into 2 time periods: the first time period was assigned to years 1995-2001 and the

second 2002-2007. Only the months with at least 50% of the month covered by measurements (i.e. 50% completeness) were included in the multi-year calculations of the mean and median concentrations. The results, as seen in Figure 3, show a decrease in the GEM concentration for every month except May (note that the decreases observed in June and December were not significant at the 95% confidence level). In contrast, the mean GEM concentration in May increased from 0.99 to 1.29 ng m<sup>-3</sup> and the median increased from 0.89 to 1.33 ng m<sup>-3</sup>. This observation is in agreement with findings reported last year (Steffen and Cole, 2008) that show that the frequency of AMDEs has shifted to earlier in the season, thus increasing the mean/median concentrations in the month of May. These results are preliminary but demonstrate that there are significant changes in the trends of GEM at Alert.

For the first time a small, yet statistically significant, decrease in the overall annual mean GEM

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concentration was observed from 1.53 to 1.51 ng m<sup>-3</sup>, using only the years where all months had 50% data completeness. This data is reported as a change between the two time periods discussed above, i.e. 1995-2001 and 2002-2007. This result shows that the trends can be very sensitive to the conditions that are placed on the data (i.e. 50% completeness of data in a given period). We thus caution the reader to understand the conditions under which this data is reported and consider that an even more robust statistical analysis of the data must be undertaken in order to draw conclusions about the long term trends of GEM in the Canadian Arctic. In addition, it is crucial that measurements of GEM continue at Alert in order to assess long-term signals in this very noisy data set.

Measurements at Little Fox Lake were initiated in 2006 to monitor the concentration levels of atmospheric Hg in the Yukon and not at a coastal Arctic location. The data collected from this location are shown in Figure 2. These data show that the concentration level of mercury in the air falls within what is expected for a remote location and the data also shows little seasonal variability. This data set will be used to assess long range transport of mercury into the Canadian Arctic and to define background levels of GEM in the air in the Yukon. At the present time, not much analysis of this data set has been done as the focus has been collecting a quality set of data before a robust analysis is undertaken. We expect that 1 more year of data will provide enough data for us to start analysis on this set of information

#### ***Atmospheric mercury processes:***

The chemical transformation of mercury in the Arctic environment is still not fully understood and is a crucial part of predicting future inputs of mercury to the ecosystem. Thus, continuous measurements of RGM and PHg at Alert were extended through 2008/09. The time series of data from initiation to the end of 2008 has been QCd and is shown in Figure 1. No further analysis of this data set was made this year because of IPY activities but this data will be compared and used to interpret the data collected during the IPY field intensive campaigns.

However, the investigation of GEM is also a great tool to further our understanding of the chemistry and dynamics of atmospheric mercury depletion

events (AMDEs). This year a detailed analysis of correlations between AMDEs (defined as GEM < 1.066 ng m<sup>-3</sup>) and meteorological parameters was made. This analysis revealed a compelling relationship between the strength and frequency of depletion events and the local temperature at Alert. As seen in Figure 4, the frequency and/or intensity of depletion events within a given month increases with decreasing temperature. At the same time, results from specific temperature ranges show that depletion events increase as the spring progresses. The cause of this month-to-month increase in AMDEs may be due to increasing solar radiation or to changes in when new sea ice is formed. This data analysis indicates that we can see a relationship between air temperature and AMDEs that was never seen previously. Additional research is needed to truly understand how these different factors contribute to the amount of mercury deposited to the surface and predict future mercury deposition. Further, these results give us an indication that certain atmospheric processes, and potential links to surface activities, are occurring during this time period.

## **Expected Project Completion Date**

ongoing

## **Acknowledgements**

The project team would like to thank the Global Atmospheric Watch program at Alert for supplying facilities, assistance and personnel. We would like to thank Christian Wile and Adrienne Glover for collecting the 2008/2009 GEM data at Alert. Much appreciation to Greg Skelton for the ongoing work on compiling this large data set! Thanks to NCP and Environment Canada for financial support of this program.

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# Contributing to International Action on POPs and Mercury: Further Applications of Global Mass Balance models

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## **Abstract**

For a number of years, the NCP has supported a research program based at Trent University and DMER Ltd with the general aim of increasing the quantitative understanding of how contaminants are transported from a variety of global sources through the global environment to the Canadian Arctic ecosystem. This program has now reached a stage of completion in which the various findings are being compiled in a series of scientific publications. This report describes these findings and outlines the content of these publications.

Specifically, our approach has been to develop simple, transparent, computer models that quantitatively simulate how chemicals move through the global environment into the arctic ecosystem. To this end we have developed the BETR-World mass balance model and compared it with both higher and lower resolution representations of global space and processes yielding similar results. The model has been adapted to treat chemicals such as mercury that exist as inter-converting species. A major effort this year has been to publish and gain international acceptance for the “Distant Residence Time” (DRT) concept which can describe the mass of chemical accumulated in

## **Résumé**

Depuis un certain nombre d'années, le PLCN finance un programme de recherche de l'université Trent et de DMER Ltd. dont le but est d'accroître la connaissance quantitative du transport des contaminants de diverses sources à travers le monde jusqu'à l'écosystème de l'Arctique canadien en traversant l'environnement planétaire. Ce programme a maintenant atteint le stade de compilation des divers résultats dans une série de publications scientifiques. Le présent rapport décrit ces résultats et expose le contenu de ces publications.

Plus exactement, notre démarche a consisté à mettre au point des modèles informatiques simples et transparents qui simulent quantitativement comment ces substances traversent l'environnement planétaire pour pénétrer dans l'écosystème arctique. À cette fin, nous avons élaboré le modèle de bilan massique BETR World que nous avons comparé à des représentations de l'espace et des processus planétaires ayant des résolutions plus grandes et plus faibles, qui donnent des résultats comparables. Nous avons adapté le modèle afin de l'appliquer à des substances qui, comme le mercure, existent sous diverses formes interconvertibles. Cette année, nous avons beaucoup travaillé à publier des articles et à

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a remote “receptor” region that results from an emission rate in a specific “source” region. We have illustrated the use of this concept with the BETR World global model of contaminant fate and we have developed and discussed the use of DRT source-receptor relationships in the form of “Responsibility Matrices”. Work is now complete on an improved segmentation of the BETR World arctic region into six smaller regions. This will enhance our ability to reconcile NCP monitoring data from both abiotic and biotic compartments in different regions of the North with modelling results and assist in determining the significant atmospheric and oceanic pathways to the North. In total we have sought to bring the issue of long range transport (LRT) to the attention of the international community by presenting scientific findings relevant to Canada’s interests in international collaborations on LRT research, by developing improved methods of estimating and depicting LRT, by publications in the scientific literature and by suggesting an international strategy for assessing LRT.

## Key Messages

We have worked to bring issues of long-range transport (LRT) to the attention of the international community by participating in international collaborations on LRT research and publications and by freely distributing software used to assess this phenomenon to academics and researchers. A publication is nearing completion which outlines a rational strategy for assessing existing, emerging and newly proposed POPs for their potential for LRT. We hope that these activities will enhance Canada’s international negotiating position on POPs and mercury.

We assert that the message that there is a need for international controls of emission sources is best accomplished by a combination of monitoring and mass balance modelling. Consistency between these data provides a convincing argument that LRT is understood and actions are required.

obtenir l’acceptation internationale de notre concept du « temps de séjour à distance » (TSD), lequel permet de déterminer la masse d’un polluant accumulé dans une région « réceptrice » éloignée à partir de son taux d’émission dans une région « source » donnée. Nous avons illustré l’application de ce concept au moyen du modèle du devenir des contaminants BETR World; nous avons également élaboré des relations source-récepteur TSD sous la forme de « matrices de responsabilité » et en avons discuté l’utilisation. Nous avons terminé nos travaux visant à mieux segmenter en six sous-régions la région arctique visée par BETR World, ce qui accroîtra notre capacité de concilier les résultats de modélisation avec les données de surveillance du PLCN extraites de compartiments abiotiques et biotiques dans des régions nordiques différentes, et nous aidera à déterminer les voies d’entrée atmosphériques et océaniques importantes vers le Nord. En résumé, nous avons tenté de sensibiliser la communauté internationale aux enjeux du transport de polluants à grande distance en présentant des découvertes scientifiques pertinentes pour le Canada dans des collaborations internationales de recherche sur le transport à grande distance (TGD), en mettant au point de meilleures méthodes d’estimation et de représentation de ce phénomène, en publiant les résultats dans les journaux scientifiques et en proposant une stratégie internationale pour l’évaluer.

## Messages clés

Nous nous sommes efforcés de sensibiliser la communauté internationale aux enjeux du transport à grande distance (TGD) en participant à des collaborations et à des publications internationales en matière de recherche sur le TGD, et en distribuant gratuitement aux universitaires et aux chercheurs des logiciels permettant d’évaluer ce phénomène. Nous aurons bientôt terminé la rédaction d’une publication qui présentera une stratégie rationnelle pour évaluer les POP actuels, nouveaux et récemment proposés concernant leurs possibilités de TGD. Nous espérons que ces activités amélioreront la position de négociation du Canada sur la scène internationale concernant les POP et le mercure.

Nous affirmons que combiner la surveillance et la modélisation du bilan massique est la meilleure façon de transmettre le message qu’il faut contrôler les sources d’émission à l’échelle internationale. La

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To this end, we have segmented the Arctic region in the BETR-World model by treating it as six sub-regions thus enabling a better comparison with NCP sponsored monitoring. Work is now complete on obtaining air and oceanic transport data for inclusion in this revised model.

We have published two papers on the “Distant Residence Time” concept showing that it is a valuable and understandable indicator of the environmental mobility of a substance, by quantifying global-scale source-receptor relationships. A third manuscript is nearly complete.

We have adapted the BETR-World model to address chemicals such as mercury which can exist as several species, each with their individual properties and environmental mobilities.

We have extended earlier work on the use of the Equilibrium Lipid Partitioning (ELP) synoptic indicator of ecosystem contamination by developing a novel and improved “activity” criterion that has the same benefits in terms of explaining contaminant fate. It has the additional potential advantage of providing information on the proximity of calculated or observed contaminant levels to levels at which toxicity is expected. It will, we believe, be particularly valuable in the context of arctic contamination.

cohérence des données est un argument convaincant que le TGD est un phénomène compris et que des mesures sont requises.

À cette fin, dans le modèle BETR World nous avons segmenté la région arctique en six sous-régions, ce qui permet de meilleures comparaisons avec les données de surveillance du PLCN. Nous avons obtenu toutes les données sur les transports atmosphérique et océanique à inclure dans le modèle révisé.

Nous avons publié deux mémoires sur le concept du « temps de séjour à distance » pour montrer qu’il est un indicateur utile et compréhensible de la mobilité d’une substance dans l’environnement en quantifiant les relations source-récepteur à l’échelle planétaire. La rédaction d’un troisième mémoire est quasi terminée.

Nous avons adapté le modèle BETR World pour traiter des substances qui, comme le mercure, peuvent exister sous plusieurs formes, chacune avec ses propriétés individuelles et ses mobilités propres dans l’environnement.

Nous avons étendu nos travaux antérieurs sur l’utilisation du fractionnement lipidique à l’équilibre comme indicateur synoptique de la contamination d’un écosystème en élaborant un critère d’« activité » nouveau et meilleur qui a les mêmes avantages pour ce qui est d’expliquer le devenir des contaminants. De plus, ce critère a l’avantage potentiel de fournir des renseignements sur le degré de proximité des niveaux de contamination calculés ou observés et des seuils de toxicité. Nous croyons que cet indicateur sera particulièrement utile en ce qui concerne la contamination arctique.

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## Objectives

The general objective of this multi-year research program has been to generate scientific results and publications that will be helpful in Canada’s international negotiations to reduce emissions of persistent organic pollutants (POPs) and mercury as well as identifying and prioritizing emerging chemical threats to the north. We have contributed to this overall objective by achieving six more specific objectives. First, we developed the BETR-World model that segments the global environment into 25 largely oceanic or terrestrial

regions. We have encouraged the development of versions of this model in other international locations, namely the BETR-Global model in Switzerland and the European Variant of BETR (EvnBETR) in the UK (Prevedouros et al. 2004; Macleod et al. 2005). The outputs of these models have been compared with monitoring data yielding generally satisfactory results. Second, we have improved the segmentation of the arctic region to enable better reconciliation of monitoring and modelling data at various regions within the arctic, especially treating the Western Arctic Ocean the

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Archipelago, Hudson Bay and the Eastern Arctic Marine regions separately. Third, we developed and extended the Distant Residence Time (DRT) concept as a transparent and versatile method by which chemicals can be assessed for their potential for long-range transport especially in the form of source-receptor matrices using the BETR-World model regions. This has been published in 2008 in the scientific literature and a sequel has been drafted. Fourth, we are continuing to adapt the model to apply to speciating chemicals. Fifth, we continued to seek a synoptic and understandable indicator of ecosystem contamination aimed at improving communication with the public and political and regulatory agencies. A novel “activity” criterion has been developed and a publication describing it will be submitted for publication shortly. Sixth and finally, we have used our experience in the area of LRT to draft a strategy for evaluating chemical substances for LRT potential. We hope that this strategy and the models we have developed and disseminated will enhance the ability to identify and assess existing, emerging and new contaminants for their potential for LRT to the North.

## Introduction

Contamination of the Arctic ecosystem is largely a result of long-range transport of chemical substances from the temperate industrial and agricultural regions of the northern hemisphere, mainly in the atmosphere, but also by ocean currents. It is recognized that the only feasible approach for reducing levels of these contaminants in the Arctic ecosystem in general and particularly in human foods, is to reduce emissions from all global sources and allow global and Arctic levels to fall by natural dissipation and removal processes. Encouraging such reductions internationally requires that foreign governments be convinced that they are sources of at least some fraction of the mass of these substances and that they should act in concert to reduce or eliminate sources. Achieving this requires, we believe, a combination of monitoring and computer modelling data presented in an easy to understand form to those undertaking negotiations at an international level. The objective of this research program has been to generate scientific results and publications that will be helpful in this context. We have sought to cooperate with those responsible for monitoring by

conventional and passive sampling techniques in the North, thus contributing a modelling element to existing monitoring programs. This research program has been based at the Canadian Environmental Modelling Centre (CEMC) at Trent University in collaboration with DMER Ltd. The program is now essentially complete. This report describes activities to date and outlines future publications that will be submitted in 2009 as a result of this multi-year program.

## Activities in 2008/2009

The activities during this year have fallen into five categories as described in more detail below.

- Publishing the Distant Residence Time concept in two papers in the refereed scientific literature with a view to enabling International agencies to better appreciate the sources and destinations of contaminants, and thus justify and apply more effective regulations.
- Improving and parameterising the model segmentation of the BETR-World arctic region in order to help explain differences in contamination levels between locations in the North.
- Applying our model of speciating substances to mercury to elucidate features of global contamination, especially the need for global-scale controls on emissions.
- Devising a novel synoptic indicator of contamination level, namely the “activity” concept that can give a simple measure of contamination level in multiple media.
- Drafting a paper suggesting an international strategy for assessing LRT potential.

## Results, Discussion and Conclusions

### *1. Distant residence time concept*

In 2008 we published two papers in Environmental Pollution deriving and illustrating the DRT concept and discussing possible application. The basis of the concept was described in the 2007 Synopsis of Research and only a very brief outline is given here. The global environment is divided into N regions and a mass balance model is used N times to estimate the fate of a specific chemical emitted in each of the N regions as it is transported to all N regions. The steady state quantities of the



chemical in each of the  $N$  receptor regions are then calculated for unit emission (e.g.  $1 \text{ kg h}^{-1}$ ) in a specific source region. This is repeated for each region as a source. The results are compiled as a  $N \times N$  unit emission matrix source-receptor matrix. In principle any model can be used, however, it is necessary to consolidate several regions if the segmentation of the model is into more than  $N$  regions. If emission estimates are available for each source region they can be assembled as a scalar factor or vector and applied to each source to give estimates of the steady state quantities in all receptor regions for all sources. The total quantities in each receptor region can be obtained as well as the global total for all  $N$  regions. This is termed the  $N \times N$  responsibility matrix.

It is thus possible to assign responsibility for the quantity of chemical in each receptor to each source. For example of the total quantity in the

Arctic,  $x\%$  is from Asia,  $y\%$  from N. America and  $z\%$  from Europe.

Another important piece of information is an estimate of the global residence time or persistence, namely the global quantity (kg) divided by the total emission rate ( $\text{kg h}^{-1}$ ). This persistence depends on the distribution of emission locations.

For a given receptor region the masses and hence residence times of the chemical in each compartment (air, water, soil, biota etc.) can be obtained. The sum of these individual compartment residence times is the regional residence time. This can reveal the potential to bioaccumulate (as is the case for PCBs) as distinct from remaining in air (e.g. HCB or  $\text{CCL}_4$ ).

It may not always be desirable to use steady state quantities, and quantities after 5 or 10 years may be preferable. Inferences can also be made

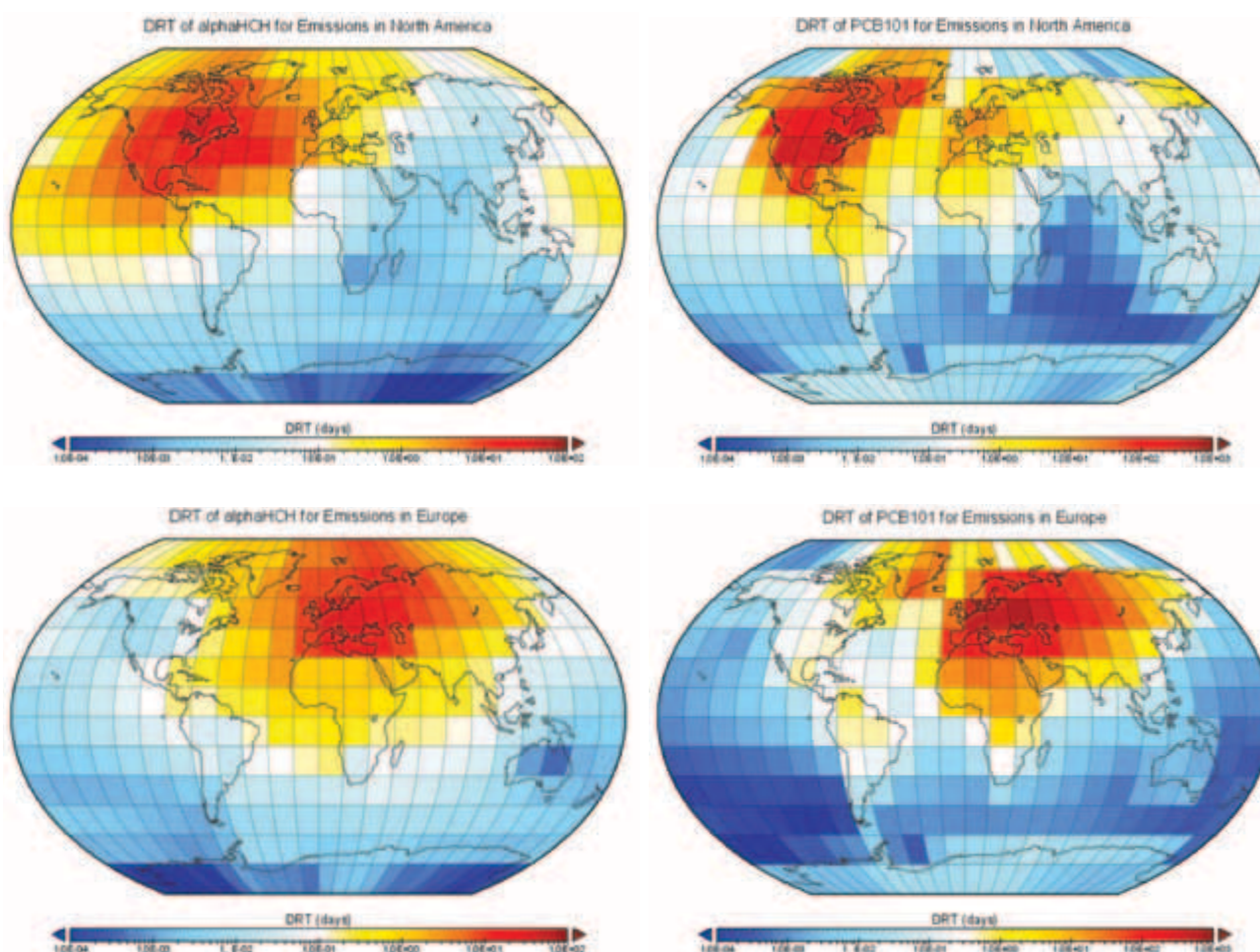


Figure 1. A) DRT of  $\alpha$ -HCH for emissions in North America. B) DRT of  $\alpha$ -HCH for emissions in Europe. C) DRT of PCB 101 for emissions in North America. D) DRT of PCB 101 for emissions in Europe.



regarding the time dependence of chemical concentrations if the emissions occur as “pulses” lasting several years rather than steady and continuous in nature. The development and application of the DRT concept was published in *Environmental Pollution* in December 2008 (Mackay et al. 2008; Reid et al. 2008)

In 2008-2009 we sought to apply three models (BETR-World, BETR-Global and GloboPOP) to a set of chemicals using identical properties and emission characteristics to ascertain if they give comparable results. This proved to be more challenging than expected because the segmentations in the three models are very different (25, 288 and 10 regions). It transpired that the two BETR models can be more easily compared. Dr. MacLeod, the author of the BETR-Global model suggested that valuable insights into global scale chemical fate could be presented pictorially if the distribution was presented separately for each source region as illustrated in Figures 1A to 1D. This series of figures comprise colour-coded maps of the globe showing the relative DRTs in various regions for emissions of  $\alpha$ -HCH and PCB101 in North America (Figures 1A and 1C) and Europe (Figures 1B and 1D). These results show that there is potential for intercontinental transport of PCB101 from North America to Europe as well as from either source region to Greenland. This clearly shows the receptor regions that are most susceptible to contamination from a given source region, e.g. emissions in Europe are more likely to reach the Arctic than emissions from North America. It also provides insights into the relative roles of atmospheric and oceanic transport.

A paper is being prepared for submission later in 2009 presenting and discussing these results. It is hoped that authors of other models will subject their findings to comparison using this DRT methodology.

## 2. Segmentation of the BETR-World arctic region

The development of the BETR-World model began with an NCP contract in the year 2001. The original 25-region global model was parameterized and tested using  $\alpha$ -HCH, a well-studied chemical that has been demonstrated to undergo long range transport (Toose et al. 2004). The model was also used to determine the global fate of toxaphene (Toose et al. 2002) and to date, has been cited in

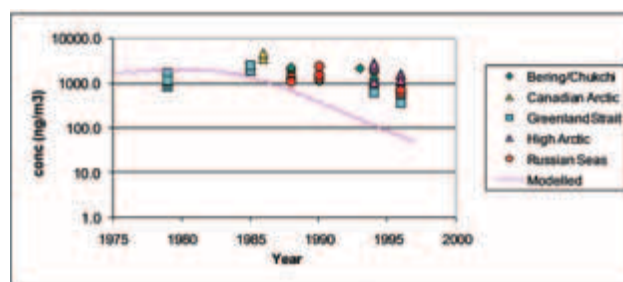


Figure 2. Modelled results compared with measured concentrations in Arctic seawater as grouped by generalized area (after Toose (2005)).

19 publications from around the world. Since the original development of the model other work has cited various pathways to and variations in arctic contamination. Becker et al. (2008) compare atmospheric levels and trends of HCH at Alert and at Zeppelin stations; they conclude that there is little spatial variation in atmospheric concentrations. Other studies show that there is spatial variation of concentrations in marine water (Figure 2, (Toose-Reid 2005); (Bidleman et al. 2007) and biota ((Li et al. 2005); (Braune et al. 2005; Gamberg et al. 2005; Rush et al. 2008)). To better understand these variations the single arctic region of the BETR-World model has been segmented into six separate, connected, regions. Figure 3:



Figure 3: The new BETR-World arctic segmentation. All other regions have remained unchanged.

shows the new segmentation which is based on oceanic circulation patterns, including the new arctic regions, namely:

- A. Arctic Ocean
- B. Canadian Archipelago
- C. Hudson's Bay
- D. Davis Strait
- E. Greenland
- F. Barents/Greenland Seas

The model has been run for  $\alpha$ -HCH to compare to steady-state results from the single arctic region. Analysis of a dynamic emission scenario, as described in Toose et al. (2004) is underway. Steady-state emissions were set to 1980 levels and compared to the BETR-World original model run for the same emission scenario and to concentration data spanning 1987 to 2005. Figures 4 and 5 show that the segmented model describes the atmospheric concentrations of  $\alpha$ -HCH with greater fidelity than the single-region arctic, which over-predicted levels by a factor of 2 to 3. Concentrations in arctic marine water environments are under-predicted by both models. This is likely because the arctic marine environment is either slow to reach, or may not reach steady-state due to slow circulation rates and annual sea-ice fluctuations. This issue is being analysed in more detail and may require changes in flow parameters. An important finding from these exploratory studies is that gaining an understanding of the fate characteristics of a specific chemical is best done using steady-state model results since these results are easier to interpret. Reconciling monitoring data with model estimates requires, however, that dynamic results be used since the “ages” of the contaminants differ considerably in different locations due to relatively slow oceanic transport. This issue is particularly important in the Canadian Arctic because of the complex oceanic circulations in the Beaufort Sea and through the Archipelago. These results are being analyzed in more depth and will be discussed in more detail in upcoming publications.

The segmentation of the Arctic region has increased the ability of the simple BETR-World model to estimate concentrations of POPs across the Arctic, without impacting the transparency

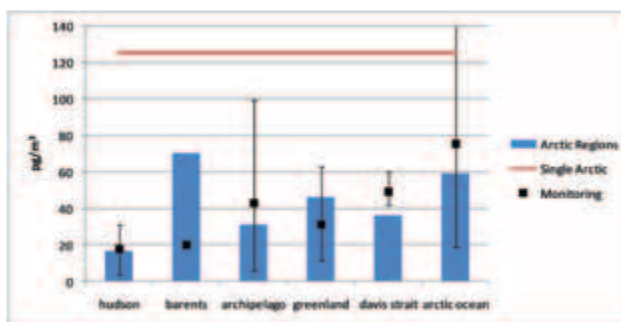


Figure 4: Steady state atmospheric concentration results for the original single arctic region, the new segmented arctic and average monitored levels. Error bars represent minimum and maximum reported concentrations.

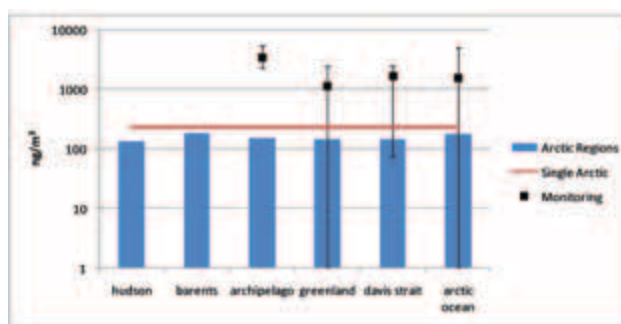


Figure 5: Steady-state marine water concentration results for the original single arctic region, segmented arctic and average monitored levels. Error bars represent minimum and maximum reported concentrations. Note the logarithmic scale of the y-axis.

or function of the BETR-World model. A manuscript fully describing the changes to the model and its application to  $\alpha$ -HCH and other POPs is in preparation.

### 3. Mercury

The mass balance models that we have developed apply primarily to single species organic chemicals such as DDT,  $\alpha$ -HCH or lindane.

It is clear that a major concern in the North is human exposure to mercury which can adopt one of several species, each of which has unique properties and species-to-species conversion is possible. We sought to modify our existing models of chemical fate, such as BETR World, to address multi-species substances. This was eventually

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accomplished and published by developing a “multiplier” method which is applicable provided that the ratios of the various species are fairly constant in each phase. This method has been successfully applied by others to metals in general.

Preliminary results from the BETR-World mercury model were encouraging and in the last year we have sought to apply it to recently published global emission data. We have also sought to apply the Distant Residence Time (DRT) concept to mercury. Further we have collaborated with Dr. Matthew MacLeod’s group in Zurich to assess mercury using elements of the LRT strategy described earlier in this report.

The results from these efforts have confirmed that mercury is truly a global contaminant that is, to a first approximation, evenly mixed in the global environment. As a result, it is not possible to argue that mercury contamination in the North is mostly attributable to a specific geographic source. The DRT results confirm this conclusion. This has, of course, been long appreciated by the mercury science community but until quite recently it has not resulted in a coordinated international effort to reduce all emissions analogous to that inherent in the Stockholm Convention and in the LRTRAP convention.

In some respects, while mercury obviously deserves high priority attention we have been reluctant to devote high priority to it in our NCP research program because the conclusions from our modelling are fairly obvious, there has been little international commitment to address the problem (but this may have changed recently) and there is an excellent highly segmented global model of mercury that has been (and continues to be) developed by Dr. Dastoor of Environment Canada.

We have plans to publish the work we have done, if only to add weight to the consensus that it is essential to seek a reduction in global anthropogenic emissions.

#### ***4. Activity as a synoptic indicator of contaminant levels in the arctic***

We believe that there is a compelling incentive to develop a synoptic indicator of contamination that can bring together the diverse concentration data in a single indicator. This will clearly show

the relationships between abiotic and biotic concentrations and it can help to establish time trends. In an earlier paper we advocated the use of the Equilibrium Lipid Partitioning criterion for this purpose (Webster et al. 1999) but it has failed to gain wide acceptance, although it has been used by others to analyse trend data on hexachlorobenzene (HCB) (Bailey 2001).

In 2008, as a result of a joint paper with Dr. Gilman Veith, formerly with the USEPA, we revisited this issue and determined that the criterion of chemical activity was a feasible and preferable criterion. It has the advantage that for chemicals inducing only narcotic effects, the activity is relatively constant at a value of approximately 0.05. A paper on this topic has been accepted for publication in SAR and QSAR in Environmental Research (Mackay et al. in press). A second paper is in preparation in which we apply this criterion to two chemicals globally, namely HCB and pentachlorobenzene (PeCB). This is being done in collaboration with Dr. Robert E. Bailey, a private consultant who has recently published a comprehensive study of the sources and prevalence of PeCB (Bailey et al. 2009). We are hopeful that if this work is accepted we will be able to apply the criterion to other substances including PCBs, DDT, brominated flame retardants and possibly polyfluorinated substances.

Our goal in this project is to develop a scientifically credible tool for assessing contaminant levels in the complex multimedia environment and especially for persistent substances that are susceptible to LRT.

#### ***5. Towards an international strategy for assessing LRT***

As a result of the work done in this NCP research program and an assessment of the published studies by others, we believe that it is opportune to suggest an international strategy for assessing LRT on a global scale. This would represent a “road map” that could be followed for new and emerging chemical of concern. It is planned to publish it in the refereed scientific literature in a journal that is read by the international regulatory community and is written in a relatively easily understood, non-technical language. A draft has been prepared and its present content is outlined below.

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In an Introduction, existing approaches for assessing LRT are first described. We suggest that for candidate chemicals of concern there should be a systematic or multi-tier process in which the chemical is assessed with increasing detail and rigour using physical-chemical and monitoring data and models.

### **Tier 1 Preliminary chemical-specific data**

First is acquisition of physical-chemical property data for the substance including the three partition coefficients and enthalpies of phase change transitions between air, water and octanol (i.e.  $K_{AW}$ ,  $K_{OW}$  and  $K_{OA}$ ), melting point, solubility in water, vapour pressure and pKa (if applicable).

Also required are estimates of degradation characteristics of the substance in media such as air, water and soils. This requires insight into likely reaction mechanisms such as reaction with hydroxyl radicals and enzymatic conversion. A conservative assumption may be to assume infinite half lives, i.e. no degrading reactions. An obvious approach is to use both estimation methods and empirical measurements and take into account available data on structurally similar substances.

Second is compiling an inventory of likely discharge rates to the environment based on known production and uses. At an early stage such estimates are likely to contain significant uncertainties but it should be possible to suggest several feasible order-of-magnitude emission scenarios.

Third is application of a simple screening level multimedia model using the physicochemical data and estimates of the magnitude of the emission rates to obtain a first “picture” of the likely environmental fate of the substance, the media of likely accumulation and the potential for bioaccumulation and biomagnification. An existing model such as Fugacity Level III or EQC or RAIDAR is suitable for this task.

### **Tier 2 Monitoring data.**

In this tier, monitoring data is acquired to the extent possible on the presence of the substance in various media including air, oceanic and fresh water, soils and biota such as fish, birds and marine and terrestrial mammals. To assist in this regard there is, as described above, an advance model-based estimate of how the substance is likely to partition and bioaccumulate. These estimates are

likely to be in considerable error but they do provide a first hypothesis against which monitoring data can be obtained and tested. Obviously, this empirical work may be complicated by issues such as availability of standards (especially for complex mixtures), the requirement to develop appropriate analytical methodologies and the possibility of speciation or dissociation.

### **Tier 3 LRT modelling**

In this tier, available LRT models are applied as outlined below in three categories.

First are evaluative models that view the potential for LRT as an intensive property of the substance and its environment, i.e. the resulting metric is independent of the quantity of chemical discharged to the environment. The metric does not apply to any specific geographic area. An example is the Characteristic Travel Distance as calculated variously by the TaPL3 model (developed with NCP support) and the OECD “Tool”.

Second are multi-regional multimedia models which segment the global environment into a relatively small number of evaluative regions, i.e. 5 to 30 that may have different properties (including temperature and precipitation). The aim is to explore how mobile chemicals are in these systems. Examples are Wania’s GloboPOP and Scheringer’s Chemrange and CliMo Chem models. Wania’s Arctic Contamination Potential is one form of presenting output from these models. A valuable feature of these models is that their simplicity enables them to be run for a large number of chemicals using, for example, chemical space diagrams. Useful insights from these chemical space diagrams is the categorisation of chemicals into groups such as “flyers”, “swimmers” and “multi-hoppers” that convey directly the nature of chemical behaviour globally. This also helps to identify chemicals that are expected to behave similarly.

Third are geographically explicit models that purport to describe chemical fate and transport in real environmental compartments of the global system. These models vary greatly in segmentation from 25 segments for the BETR-World to 288 segments in BETR-Global, to models such as CanPOP and GRAHM. These models can use averaged or real atmospheric and oceanic transport data. They are inevitably dynamic in nature but they may



also yield steady state results. Actual chemical use or emission data can be used yielding results that are capable of being compared to real monitoring data. Results from these models can be presented using the Distant Residence Time concept discussed elsewhere in this report.

Most of these models are applicable only to single species substances, thus they can not treat dissociating substances such as acids or bases or mercury. The exceptions are BETR World and GRAHM that can treat mercury.

#### Tier 4 Reconciliation of monitoring and modelling data.

In this final tier, the data obtained in earlier tiers is consolidated to test if the model predictions are in reasonable accord with the monitoring data, given the many likely uncertainties about chemical properties fate and emission estimates.

### Expected Completion Date

This project is now complete, no further funds are being sought and the remaining tasks are to complete and submit publications as described above.

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# Temporal trends of persistent organic pollutants and metals in ringed seals from the Canadian Arctic

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## **Abstract**

Trends in concentrations of polybrominated diphenyl ethers (PBDEs), endosulfan, toxaphene, PCBs, DDT, and other persistent organic pollutants (POPs) were studied using blubber of female ringed seals from up to 12 Canadian Arctic communities based on samples collected by local hunters in 2008 at 5 communities combined with results from previous years. Archived samples from Grise Fiord, collected in 1972, were also analysed in 2008. Total PBDE concentrations in blubber collected in 2008 were generally low averaging 4 ng/g (Resolute Bay) to 10 ng/g (Gjoa Haven). Maximum PBDE concentrations were found in samples from 2005 and 2006 and lowest concen-

## **Résumé**

Les tendances des concentrations d'éthers diphenyliques polybromés (PBDE), d'endosulfan, de toxaphène, de BPC, de DDT et d'autres polluants organiques persistants ont été étudiées dans le lard de phoques annelés femelles dans jusqu'à 12 collectivités de l'Arctique canadien, à l'aide d'échantillons recueillis par des chasseurs locaux en 2008 dans 5 collectivités et combinés avec les résultats des années précédentes. Les échantillons archivés provenant du fjord Grise, recueillis en 1972, ont également été analysés en 2008. Le total des concentrations de PBDE mesurées dans le lard recueilli en 2008 était généralement faible, en moyenne de 4 ng/g (baie Resolute) à 10 ng/g (Gjoa Haven). Les

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trations in archived samples from 1972 (Grise Fiord, Resolute and Sachs Harbour). Endosulfan was detectable in all seal blubber samples but at very low concentrations (means of 1.0-1.3 ng/g). Mercury was measured in seal muscle from 5 communities (Arviat, Grise Fiord, Resolute Bay, Kangiqsualujjuaq, and Gjoa Haven) and average concentrations ranged from 0.027 ug/g wet wt (Gjoa Haven) to 0.45 ug/g (Resolute).

## Key Project Messages

- Temporal trends of POPs and mercury were examined in ringed seals from up to 12 Canadian Arctic communities by combining new results from samples collected in 2008 with previous results.
- Brominated diphenyl ether flame retardants were detected at low concentrations in all seals with highest in seals from southern and eastern locations e.g. Nain, Arviat, Inukjuaq, Pangnirtung.
- Mercury concentrations in seal muscle were very low relative to liver. Average concentrations of mercury in muscle varied widely among communities.

concentrations maximales de PBDE ont été trouvées dans les échantillons datant de 2005 et de 2006, tandis que les plus faibles concentrations ont été relevées dans les échantillons archivés à partir de 1972 (fjord Grise, baie Resolute et Sachs Harbour). L'endosulfan était détectable dans tous les échantillons de lard de phoque mais à de très faibles concentrations (moyennes de 1,0-1,3 ng/g). Le mercure a été mesuré dans les muscles des phoques de cinq collectivités (Arviat, fjord Grise, baie Resolute, Kangiqsualujjuaq et Gjoa Haven) et les concentrations moyennes variaient entre 0,027 ug/g poids frais (Gjoa Haven) et 0,45 ug/g (baie Resolute).

## Messages clés

- Les tendances temporelles des POP et du mercure ont été examinées chez les phoques annelés dans jusqu'à 12 collectivités de l'Arctique canadien en combinant les nouveaux résultats des échantillons prélevés en 2008 avec les résultats précédents.
- Les ignifugeants (éther diphenylique polybromé) ont été détectés à de très faibles concentrations chez tous les phoques, ceux du Sud et de l'Est présentant les concentrations les plus élevées (p. ex. Nain, Arviat, Inukjuaq, Pangnirtung).
- Les concentrations de mercure dans les muscles des phoques étaient relativement faibles par rapport aux concentrations dans le foie. Les concentrations moyennes de mercure dans les muscles variaient considérablement d'une collectivité à l'autre.

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## Objectives

1. Determine temporal trends of persistent organic pollutants (POPs) and new organic chemicals of potential concern, as well as mercury and other metals in ringed seals using annual collections at 3 communities.
2. Determine temporal trends of POPs and mercury at 10 other locations on a 5 year cycle using previous data from the 1970s, 1980s and 1990s as well as archived samples if available.
3. Identify and prioritize other new contaminants that are entering the Arctic environment and

contribute information to Canadian and International assessments of new candidate POPs.

4. Provide the information on levels and temporal trends of these contaminants to each participating community and to the Territorial contaminants committees.

## Introduction

The ringed seal is the most abundant Arctic pinniped with a circumpolar distribution and has been a key biomonitoring animal for examining spatial and temporal trends of persistent organic

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pollutants (POPs) and mercury in the Arctic since the 1970s. This project began in April 2004 under NCP Phase III and follows up earlier projects on ringed seals (Muir and Lockhart 1994; Muir 1996, 1997; Muir et al. 1999; 2001; 2003). Results for POPs and heavy metals including mercury in the form of the original raw data are available going back to the 1980s, and earlier in some cases.

Archived ringed seal samples are available mainly from the Environment Canada tissue bank (National Wildlife Research Centre, Ottawa), from the 1970s and 1990s. In our 2007-08 report we focused on the temporal trends of mercury, perfluorinated compounds and toxaphene in selected communities (Muir et al. 2008). This report represents a continuation of that work.

Because ringed seals are an important species harvested by hunters each year in almost all communities in Nunavut, Nunavik, Nunatsiavut, and the Inuvialuit Settlement Region, this project provides an opportunity to involve the communities in the scientific program of the NCP.

Participation of hunters in each community has been consistent and the quality of the hunter based collection has generally been high. For example, requested information on gender, girth, length, blubber thickness has been provided for about 90% of the animals sampled which is good considering the logistical challenges the hunters face in having to harvest and dissect the animals.

## Activities in 2008/2009

### Sample collection:

In 2008-09 ringed seal samples were successfully collected with the help of hunters in the communities of Arviat (N=25), Grise Fiord (N=20), Resolute Bay (N=25), Sachs Harbour (N=20) and Gjoa Haven (20). At Kangiqsualujjuaq we were able to get only 6 samples collected. Samples from Gjoa Haven were destroyed by the failure of the freezer in the HTA office. However samples from 8 seals collected by Adam Morris with the help of local hunters in June 2008 were available for analysis. Samples from Sachs Harbour were shipped by air freight but did not arrive at the Nunavik Research Centre and are presumed lost.

Collections consisted of blubber, liver, muscle, kidney, tooth/lower jaw (for aging). Essential data on length, girth, blubber thickness at the sternum,

and sex was provided for almost all animals for all locations. Samples were stored at -20°C and then shipped frozen to NvRC in Kuujjuaq for processing and metals analysis. Large subsamples of all tissues were archived in walk-in freezers at -20 to -35°C in sealed plastic bags (double bagged). For tooth aging, samples from Resolute and Arviat were sent to Matson Labs (Milltown, MT) while those from Arviat were aged by DFO (Winnipeg).

Short reports (in English and Inuktitut) on the results of the study to date were faxed to the Hunters and Trappers committee offices of each community in March/April 2009 as part of communication and consultations. In Jan/Feb 2009, project summaries were also sent to the Chairs and Inuit Research Advisors of the Nunavut Niqiit Avatitinni Committee, the NWT Environmental Contaminants Committee and the Nunavik Nutrition and Health Committee.

### Chemical analyses:

Organochlorine pesticides (OCPs), PCBs and polybrominated diphenyl ethers (PBDEs) in seal blubber were determined as described by Muir et al. (2005) with minor modifications. Samples were extracted using pressuring fluid extraction (ASE 300, Dionex Instruments) and all extractions and solid phase fractionation on silica gel columns were conducted in a "clean room" (HEPA and carbon filtered air) at Canada Centre for Inland Waters (Burlington ON). Only samples from females or juvenile males were analysed.

All liver and kidney samples were analysed for heavy metals (cadmium (Cd), mercury (Hg), lead, selenium and arsenic) at NvRC (Kuujjuaq) using atomic absorption spectrometry (AAS) (Muir et al. 2005). Seal muscle was analysed for total mercury using a Direct Mercury Analyser (DMA; Milestone Instruments). Seal liver samples were also analysed for perfluorinated alkyl acids as described by Butt et al. (2008). Instrumental analysis was performed by LC-MS/MS following previously described conditions (Butt et al. 2008).

Short and medium chain chlorinated paraffins (SCCP-MCCPs) were quantified in the same fractions analysed for PBDEs by GC-high resolution mass spectrometry (HR MS) in negative chemical ionization mode. Toxaphene and endosulfan ( $\alpha$ ,  $\beta$  isomers and sulfate metabolite) were determined

by GC-negative ion low resolution MS (GC-NIMS). Toxaphene was determined as “total” toxaphene using a technical toxaphene standard and also by quantification of individual chlorobornanes (see Muir et al. (2004) for further details on methods).

#### Quality assurance and statistical analysis:

QA steps included the analysis of reference materials for heavy metals and organochlorines and reagent blanks. All results were blank subtracted. Prior to use of clean room conditions for seal blubber analysis (1998-2006 samples) blank results were typically <1% of individual PCBs except for CB31, 28 and 52 where they were 5-15% of values in seal blubber. Similarly PBDEs congeners BDE 47, 99 and 100 were 30-70% of the values of these congeners in seal blubber. Under clean room conditions used for 2007 and 2008 sample extractions blank values for PCBs were <1% of typical values and <10% of BDE47, 99 and 100 values.

Seven seal muscle samples were analysed for mercury in triplicate using the DMA with average relative standard deviation of 18%. Mercury results for reference materials DORM-1 and NIST oyster muscle were within 10% and 5% of certified values, respectively. The NvRC metals lab and the NLET organics lab have participated in the NCP Quality Assurance Program.

Basic statistics, correlations and frequency distributions were determined using Systat Version 12 (Systat Software Inc, Chicago IL). Organohalogen concentrations in ringed seals were normalized to 100% lipid. For temporal trend comparisons results for both mercury and organohalogens were first tested for normality using the Shapiro-Wilk test. All contaminants data were log10 transformed to give coefficients of skewness and kurtosis <2 and geometric means (back transformed log data) were calculated.

## Results and Discussion

### Spatial trends of mercury in seal muscle:

Geometric mean concentrations of mercury in ringed seal muscle from 5 study locations in 2008 are shown in Table 1. Concentrations ranged widely (0.006 to 1.3 ug/g ww) and were highly skewed i.e. about 50% of concentrations were <0.2 ug/g ww, therefore only the geometric mean and minimum and maximum are shown. Geomean concentrations ranged from 0.027 ug/g wet wt (Gjoa Haven) to 0.45 ug/g (Resolute). The frequency distribution and correlations of length, blubber thickness and mercury concentrations in muscle for 68 ringed seals from the 5 communities are shown in Figure 1. A significant correlation of

**Table 1. Concentrations (ug/g or parts/million) of mercury in ringed seal muscle**

ID		N	Length (cm)	Girth (cm)	Blubber thickness (cm)	Mercury ug/g ww
Arviat	Mean	25	106	92	5	0.261
	min		77	49	2	0.126
	max		130	119	7	0.959
Gjoa Haven	Mean	6	146	100	4	0.027
	min		117	84	3	0.006
	max		181	133	5	0.095
Grise Fiord	Mean	22	118	97	4	0.097
	min		86	64	3	0.015
	max		183	145	6	0.375
Kangiqsualujjuaq	Mean	5	66	40	3	0.130
	min		64	38	3	0.082
	max		69	41	3	0.209
Resolute	Mean	20	127	111	5	0.454
	min		97	84	3	0.067
	max		152	137	8	1.374

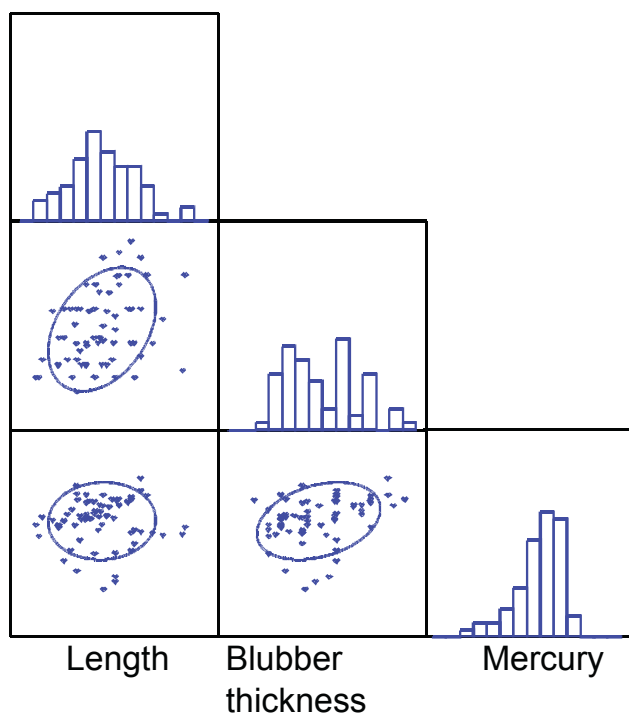


Figure 1. Frequency distribution and correlations of length, blubber thickness and mercury concentrations in muscle for 68 ringed seals from 5 communities (see Table 1 for locations). Significant correlations of mercury (log transformed) and blubber thickness, as well as blubber thickness with length (at  $P=0.05$ ) were found.

(log transformed) mercury concentrations was found with blubber thickness ( $P=0.02$ ) but not with length. Blubber thickness and length were also correlated ( $P=0.007$ ). Results for both males and female ringed seals were combined because there was no influence of gender of the animals on mercury muscle concentrations.

#### Polybrominated diphenyl ethers and other brominated flame retardants:

PBDEs have been determined in ringed seal blubber since 2004 and also retrospectively in archived samples from the 1970s and 1990s. Concentrations of total PBDEs, and major congeners BDE47 and BDE 99, in female (or juvenile male) blubber for the most recent sampling year from 12 communities are shown in Table 2. Highest total PBDE concentrations were generally found in seals from more southern and eastern locations e.g. Nain, Arviat, Inukjuaq, Pangnirtung, Kangiq-sualujuaq. Concentrations were generally low compared to total PCBs in the same samples ranging from

averages 7 ng/g lipid wt (Resolute Bay) to 29 ng/g (Nain) for total PBDEs. Kelly et al. (2008) reported a geometric mean concentration of 10 ng/g lipid wt for total PBDEs in female ringed seal blubber from Nunavik (samples were from Salluit and Quaqtaq provided to Kelly et al by the present study). Results for total PBDEs, BDE47 and 99 are in good agreement with results reported by Kelly et al.

PBDE concentrations have changed over the past 5 years and this may complicate spatial comparisons. Temporal trends for 6 locations are shown in Figure 2. Maximum PBDE concentrations were found in samples from 2005 and 2006 and lowest concentrations in archived samples from 1972 (Grise Fiord, Resolute and Sachs Harbour). Based on the most recent analyses concentrations appear to be declining at Arviat, Inukjuaq, Gjoa Haven, Resolute and Sachs Harbour.

Blubber samples were also analysed for penta-bromoethylbenzene, hexabromocyclododecane, bis(tribromophenoxy)ethane and decabromodiphenylethane. However none of these brominated compounds were detected consistently in seals and, where occasionally detected, results were close to instrument detection limits.

#### Chlorinated and fluorinated organic compounds

In 2008-09 analyses continued for legacy POPs (PCBs, DDT, chlordanes, toxaphene, HCH), chlorinated byproducts (tetra-, penta and hexachlorobenzene, hexachlorobutadiene, octachlorostyrene) and chlorinated pesticides (methoxychlor, lindane, endosulfan). Temporal trends for PCBs, DDT, HCH and chlordanes related compounds have been reported in previous years (Muir et al. 2006; Muir et al. 2007) and remain basically unchanged.

Results for perfluorinated carboxylates (PFCAs) and perfluoroalkylsulfonates (PFSA) in ringed seal liver also remain unchanged from 2007. Time trends of the PFCA and PFSA will be reported in future years.

This year we focus on results for endosulfan because far less has been published on this contaminant in ringed seals and results have been problematic due to use of GC-electron capture detection for some analyses (Weber et al. 2009). For this study all analyses were by GC-NIMS

**Table 2. Concentrations<sup>1</sup> of total PBDEs and major congeners in ringed seal blubber (ng/g lipid weight) from 12 communities. Results for the most recent sampling year.**

Location	Year	N		BDE47	BDE99	Total PBDEs <sup>2</sup>
Arviat	2008	9	Mean	3.2	1.0	5.8
			Range	1.7-8.4	0.4-2.3	3.2-13
Gjoa Haven	2008	6	Mean	5.6	1.4	9.7
			Range	3.6-9.7	0.6-2.1	6.5-15
Resolute	2008	10	Mean	4.0	0.07	7.3
			Range	1.2-22	0.0-5.7	2.9-36
Grise Fiord	2008	7	Mean	2.3	0.08	4.2
			Range	1.3-3.5	<0.01-0.3	2.0-7.5
Kangiqsualujjuaq	2008	3	Mean	13	1.4	17
			Range	5.0-23	0.37-3.3	6.5-32
Sachs Harbour	2007	12	Mean	3.8	0.16	4.94
			Range	0.71-7.9	<0.01-0.96	0.99-10
Inukjuaq	2007	10	Mean	9.9	2.6	16
			Range	2.4-27	0.69-78	4.0-38
Holman	2006	10	Mean	1.6	0.04	5.0
			Range	<0.01-6.9	0.01-5.3	0.4-18
Pangnirtung	2006	10	Mean	6.0	0.8	9.4
			Range	4.0-7.5	<0.01-3.2	6.9-13
Nain	2005	8	Mean	23	2.2	29
			Range	11-46	1.0-4.5	15-55
Qikiqtarjuaq	2005	7	Mean	3.0	0.17	5.7
			Range	1.4-8.3	<0.01-4.2	2.6-18
Arctic Bay	2004	9	Mean	5.2	0.43	6.6
			Range	0.80-12	0.10-1.3	1.0-16

<sup>1</sup> Geometric means and ranges of concentrations

<sup>2</sup> Sum of BDE congeners in order of their elution on a GC column: BDE 17, 28, 49, 47, 66, 100, 99, 85, 154, 153, 138, 183, 190, 209)

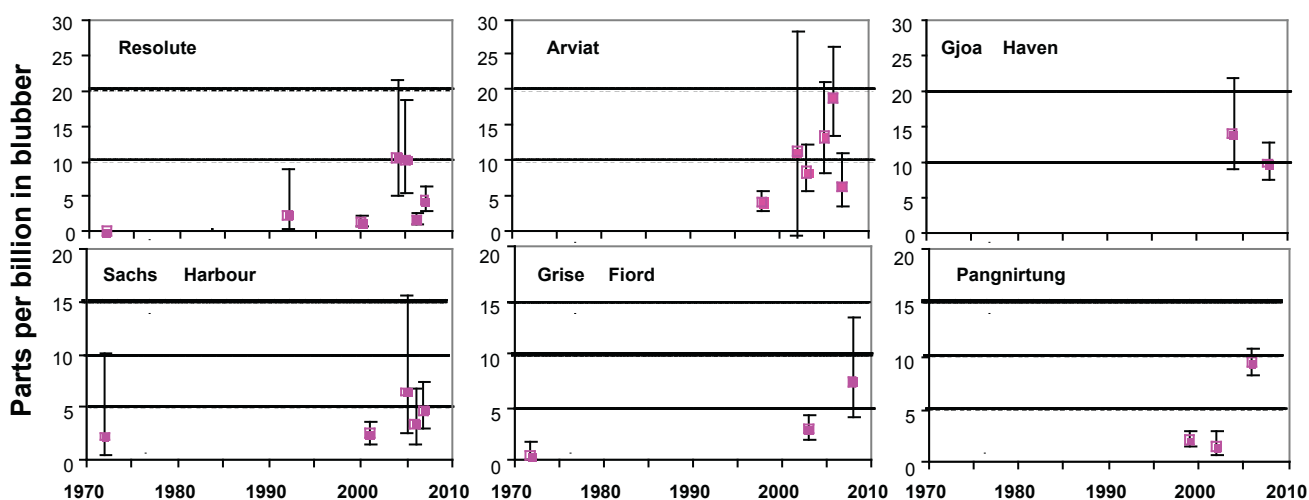


Figure 2. Temporal trends of polybrominated diphenyl ethers in ringed seal blubber (females and juvenile males only) from 6 communities. Symbols represent geometric mean concentrations and vertical lines are 95% confidence intervals.



**Table 3. Concentrations of endosulfan isomers and the degradation product endosulfan sulfate in ringed seal blubber collected in 2008 (ng/g lipid wt).**

Location	N		$\alpha$ -endosulfan	$\beta$ -endosulfan	Endosulfan sulfate	Total Endo
Arviat	9	Mean	0.12	0.85	0.09	1.1
		Range	0.03-0.25	0.51-2.0	0.06-0.20	0.73-2.3
Gjoa Haven	6	Mean	0.08	0.78	0.14	1.0
		Range	0.04-0.11	0.58-1.2	0.06-0.21	0.78-1.3
Grise Fiord	10	Mean	0.12	0.79	0.03	0.98
		Range	0.06-0.31	0.49-2.0	0.01-0.14	0.61-2.4
Kangiqsualujuaq	3	Mean	0.29	1.0	0.03	1.3
		Range	0.20-0.39	0.50-1.5	0.02-0.05	0.71-1.9
Resolute	7	Mean	0.22	0.87	0.02	1.1
		Range	0.18-0.28	0.51-1.5	0.01-0.05	0.76-1.8

using ions  $m/z$  406 and 372 for  $\alpha$ - and  $\beta$ -endosulfan and  $m/z$  386 and 388 for endosulfan sulphate. Beta-endosulfan was the predominant component of endosulfan representing 77-80% of total endosulfan (Table 3). The predominance of the beta- isomer is surprising given its low proportions in seawater (Weber et al. 2009). However, Kelly et al.(2007) also found  $\beta$ -endosulfan was the major endosulfan isomer in ringed seal blubber from Nunavik using high resolution MS analysis. They reported mean concentrations of 3.02 ng/g lipid wt in female ringed seals which is 1.5 to 3-fold higher than found in this study.

## Discussion and Conclusions

This study has provided new information on the spatial and temporal trends of brominated diphenyl ethers, and on levels of endosulfan, in ringed seals. This is the first temporal trend information on PBDEs in Canadian arctic ringed seals with the exception of the long term study at Ulukhaktok (Ikonomou et al. 2002) which included data to 2000. Results for endosulfan confirm that it is present at low concentration compared to other POPs. The reason for the predominance of the beta-endosulfan isomer in seals is unknown and deserves further investigation. The increased number of sampling times at Sachs Harbour, Arviat and Resolute is permitting more accurate estimates of the annual changes in concentrations and this accuracy is likely to improve in the future as annual collections are made at these locations.

This study also has provided more information on mercury in seal muscle. Relatively little information was available previously due to the focus on liver samples by early investigators which continued in NCP Phase III blueprints. However, Gaden et al. (2009) have shown that muscle mercury is less variable temporally than liver. The data from 5 communities show that mercury concentrations in seal muscle vary widely across the arctic. Further study is needed to investigate the reasons for this variation.

## Expected Project Completion Date

March 31, 2010 will represent the end of a 5 year annual sampling campaign at 3 locations and the time to fully evaluate results.

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# Temporal and Spatial Trends of Organic and Metal/ Elemental Contaminants in Canadian Polar Bears: 2008-2009 NCP Project Summary Report

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Taloyoak HTA, Box 6, Taloyoak, X0B 1B0

Coral Harbour HTA, Box 108, Coral Harbour X0C 0C0

Western Hudson Bay (WHB):

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Rankin Inlet HTA, Bag 002, Rankin Inlet, X0C 0A0

Southern Hudson Bay (SHB)

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Lancaster Sound/Jones Sound (LS/JS):

Arctic Bay HTA, Box 99, Arctic Bay, X0A 0A0

Grise Fiord HTA, Box 71, Grise Fiord, X0A 0J0

Resolute Bay HTA, Box 217, Resolute Bay, X0A 0V0

Baffin Bay (BB):

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Clyde River HTA, Box 90, Clyde River, X0A 0E0

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## Abstract

The purpose of this core monitoring polar bear project has two facets. First is to examine recent time-related changes in persistent organic pollutants (POPs) and metals/elements in bears from the seven territorial management zones in the Canadian arctic (Nunavut and the NWT). That is, to make two-time point comparisons (2007-2008 compared to 2001-2002) of levels and patterns of legacy (e.g., PCBs and organochlorine (OC) pesticides such as chlordanes and DDTs) and newly identified and/or emerging contaminants (e.g., polybrominated diphenyl ethers (PBDEs), hexabromocyclododecanes (HBCDs) and perfluorinated compounds (PFCs)), and metals/elements (e.g., mercury (Hg)) in tissues of bears. The seven territorial management zones are the Gulf of Boothia/Foxe Basin (GB/FB), Western Hudson Bay (WHB), Southern Hudson Bay (SHB), Lancaster Sound/Jones Sound (LS/JS), Baffin Bay (BB), Davis Strait (DS) and Amundsen Gulf (AG). For all bear samples obtained in 2007 and 2008, the determination of stable carbon and nitrogen isotopes (SIs) and fatty acids (FAs) are being used as indicators of trophic as well as dietary differences among bear populations.

All tissue sample (fat, muscle and liver) collections for polar bears were completed in 2008. With the exception of the determination of metals/elements (e.g., Hg) and isomer-specific perfluorooctane sulfonates (PFOSs), all contaminant analyses were completed as of 2008-2009 as well as all stable isotope (SI), fatty acid (FA) and age determinations. For the 2007-2008 samples, some of the results to date include the detection of numerous established BFRs (e.g., PBDEs and HBCD), environmentally emerging BFRs (e.g., decabromodiphenyl ether (BDE209) and decabromodiphenyl ethane (DBDPE)), and PFCs and precursors (e.g. (PFOS) and its precursor perfluorooctane sulfonamide (PFOSA)). Among

## Résumé

Le but de ce projet stratégique de surveillance de l'ours polaire est double. Il s'agit tout d'abord d'étudier les changements temporels récents quant aux polluants organiques persistants (POP) et aux métaux et éléments chez les ours de sept zones de gestion territoriale de l'Arctique canadien (Nunavut et T. N.-O.). Autrement dit, il s'agit de faire des comparaisons entre deux périodes (2007-2008 par rapport à 2001-2002) des niveaux et des modèles d'anciens pesticides (BPC et composés organochlorés (OC) comme les chlordane et le DDT) et les contaminants émergents ou nouvellement identifiés (éthers diphenyliques polybromés (PBDE), hexabromocyclododécane (HBCD) et composés perfluorés (PFC)), et les métaux et éléments (mercure (Hg)) dans les tissus des ours. Les sept zones de gestion territoriale sont celles du golfe de Boothia/bassin Foxe (GB/FB), de l'Ouest de la baie d'Hudson (WHB), du Sud de la baie d'Hudson (SHB), du détroit de Lancaster et du détroit de Jones (LS/JS), de la baie de Baffin (BB), du détroit de Davis (DS) et du golfe d'Amundsen (AG). Pour tous les prélèvements effectués chez les ours en 2007 et 2008, la détermination du carbone stable et des isotopes d'azote (SI) et des acides gras (FA) était utilisée comme indicateur des différences trophiques et alimentaires chez les populations d'ours.

Tous les échantillons de tissus (graisse, muscle et foie) prélevés chez les ours polaire l'ont été en 2008. À l'exception de la détermination des métaux et des éléments (comme le Hg) et des perfluorooctane sulfonates spécifiques aux isomères (PFOS), toutes les analyses des contaminants ont été faites en 2008-2009, ainsi que les déterminations des isotopes stables (SI), des acides gras (FA) et de l'âge. Pour les échantillons de 2007-2008, certains des résultats obtenus jusqu'ici incluent la détection de nombreux BFR établis (PBDE et HBCD), de BFR émergents de l'environnement (éther décabromodiphényle (BDE209) et éthane décabromodiphényle (DBDPE)),

the preliminary assessments, and with respect to PFCs,  $\Sigma$ -perfluorinated sulfonates (PFSA; mainly perfluorooctane sulfonate (PFOS)),  $\Sigma$ -perfluorinated carboxylates (PFCA), perfluorooctane sulfonamide (PFOSA) and  $\Sigma$ PBDE flame retardant levels were highest in bears from SHB, and lower but comparable among the AG, GB/LS and DS and BB bears. SI and FA tracers indicated that in addition to geographical factors, dietary differences existed for some bear populations. Based on deviations from a ringed seal blubber diet, dietary differences partially explained the geography-dependent differences of the levels and patterns of several POPs in polar bears.

For 2007-2008 samples, PFC levels in bears appear to unchanged in western and increasing in eastern populations compared to levels from previous 2001-2002 assessments. PBDE levels are somewhat lower now than they were in 2001-2002, which is the only earlier time point where PBDEs were reported in Canadian polar bears. Longer term temporal (1991-2007) trends of legacy and emerging POPs in WHB bears are also being assessed using available and archived fat biopsies. Sea ice-mediated diet change (as a function of climate change) is being examined in WHB polar bears and impacts on levels of PCBs, OC pesticides, the flame retardants PBDEs and HBCD. This work is of significant value to Northerners, whose cultural lifestyle depends on subsistence foods (seals) that polar bears prey on, as well as polar bears being a part of the Inuit diet.

## Key Messages

- Several emerging and newly detected, non-PBDE BFRs (e.g., DBDPE, BDE-209 and HBCD) were detected infrequently in the fat of Canadian polar bears, but at substantially lower levels than other POPs and including PBDEs.
- Among Canadian bear populations, the highest concentration of sum-PBDEs was in animals from SHB.

et de PFC et leurs précurseurs ((PFOS) et de son précurseur le perfluorooctane sulfonamide (PFOSA)). Parmi les évaluations préliminaires, en ce qui concerne les PFC, les niveaux de sulfonates  $\Sigma$ -perfluorés (PFSA; principalement le perfluorooctane sulfonate (PFOS)), des carboxylates  $\Sigma$ -perfluorés (PFCA), de perfluorooctane sulfonamide (PFOSA) et d'ignifugeant  $\Sigma$ PBDE étaient plus élevés chez les ours de SHB, et plus faibles mais comparables chez les ours de AG, GB/LS et DS et BB. Les traceurs de SI et FA indiquaient qu'en plus des facteurs géographiques, il existait des différences alimentaires chez certaines populations d'ours. En fonction d'écarts de la diète de graisse de phoque annelé, les différences alimentaires expliquaient en partie les différences liées à la géographie des niveaux et des modèles de plusieurs POP chez l'ours polaire.

Pour ce qui est des échantillons de 2007-2008, les niveaux de PFC chez les ours semblaient ne pas avoir changé dans les populations de l'Ouest et avoir augmenté dans celles de l'Est, comparativement aux niveaux des évaluations antérieures de 2001-2002. Les niveaux de PBDE semblent en quelque sorte moins élevés qu'ils ne l'étaient en 2001-2002, soit la seule année pour laquelle les PBDE ont été analysés chez l'ours polaire canadien. Les tendances temporelles à plus long terme (1991-2007) des anciens et des nouveaux POP chez les ours de WHB ont aussi été évaluées à partir des biopsies de graisse disponibles et archivées. Le changement de régime atténué par la glace marine (fonction du changement climatique) est étudié chez les ours de WHB et les impacts sur les niveaux de PCB, de pesticides OC, d'ignifugeants PBDE et des HBCD. Ces travaux sont importants pour les habitants du Grand Nord, car leur mode de vie culturel dépend de la nourriture de subsistance (phoques) des ours et du fait que les ours polaire font partie de l'alimentation des Inuits.

## Messages clés

- Plusieurs BFR non PBDE émergents et nouvellement détectés (DBDPE, BDE-209 et HBCD) ont été détectés peu fréquemment dans la graisse des ours polaire canadiens, mais à des niveaux substantiellement moindres que les autres POP, y compris les PBDE.
- Chez les populations d'ours canadiens, la plus forte concentration du total des PBDE se retrouvait chez les animaux de SHB.



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- ΣPFSA (PFOS), ΣPFCA and PFOSA were highest in bears from SHB, and lower but comparable among the AG, GB/LS, BB and DS.
  - Temporal data for WHB bear shows that SI and FA values and patterns varied from 1991 to 2007, which is consistent with increased consumption of open-water seal species in years of earlier sea ice break up.
  - The diet shift in WHB bears over the period of 1991-2007 led to greater rates of contaminant level increases over time (e.g., PBDEs and some OC pesticides), and an apparent decreasing trend to increasing trend for PCBs, compared to trends assuming a constant diet.
  - ΣPFSA (PFOS), ΣPFCA et PFOSA étaient les plus élevés chez les ours de SHB, et moins élevés mais comparables chez ceux de AG, GB/LS, BB et DS.
  - Les données temporelles de l'ours de WHB montrent que les valeurs et modèles SI et FA variaient entre 1991 et 2007, ce qui correspond à une consommation accrue d'espèces de phoques en mer ouverte dans les années de débâcle précoce de la glace marine.
  - Le changement de régime des ours de WHB entre 1991 et 2007 a entraîné des taux plus élevés d'augmentation des niveaux de contaminants dans le temps (PBDE et certains pesticides OC), et une apparente tendance à la baisse vers une tendance à la hausse des BPC, comparativement aux tendances si l'on suppose un régime constant.
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## Project Objectives

### Long term:

To determine and monitor the spatial and temporal trends (e.g., concentrations and congener patterns), bioavailability, fate, and toxicokinetics (e.g., bio-transformation and tissue distribution) of legacy and “new” POPs (chlorinated, brominated and fluorinated), their persistent degradation products, precursors and isomers, as well metal and other elements, in polar bears from seven Canadian arctic management zones.

### Short term

1. To determine the spatial (2007-2008) and temporal (two-time point comparison, 2001-2002 vs 2007-2008) trends of legacy and emerging POPs (e.g., PBDEs, HBCD and PFCs) and degradation products, precursors and/or isomers, and metals/elements (e.g., Hg) in polar bears using the appropriate tissues collected in communities within seven management zones in the Canadian Arctic, using appropriate tissues (fat or liver) collected over the 2007-2008 period.
  2. To use carbon and nitrogen stable isotopes (Sis) and fatty acids (FAs) as ecological tracers of trophic levels and diet, and determined in polar bear and ringed seal tissues (muscle or fat) from all possible management locations (assessed in (1)) to examine the influence of diet and trophic level as confounding factors on POP spatial and temporal trends, in addition other influential factors such as sex, age, time of collection, and lipid content.
  3. To identify and determine emerging chlorinated, brominated and fluorinated POPs that may persist in the tissues of polar bears, are not necessarily listed as a NCP, LRTAP or Stockholm Convention priority POP, and can be monitored for as part of or with little deviation from analytical methods applied for determination of the NCP priority POPs.
  4. To provide information in a timely manner to each Inuit community participating in the study, on the levels and trends of POPs in polar bears. This would include translation of documentation and deliverables into Inuktitut.
  5. To archive the remaining polar bear tissue samples that were collected as part of this project, in Environment Canada's Wildlife Specimen Bank located at the NWRC, Carleton University, Ottawa.
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## Introduction

The polar bear (*Ursus maritimus*) is the apex predator of the Arctic marine ecosystem. Due to its position at the top of the marine food web, levels of persistent organic pollutant (POP) and metal/element contaminants in polar bears are among the highest observed in the Arctic. Like humans, polar bears are circumpolarly distributed, and thus are an ideal sentinel/monitoring species for contaminants in the Arctic. Through the process of biomagnification the polar bear achieves some of the highest contaminant concentrations of any arctic species or any species on the planet (Braune et al. 2005; de March et al. 1998; Letcher et al. 2009a; Muir et al. 1998, 2006, 2007; Norstrom 2001; Smithwick et al. 2005; Verreault et al. 2005a), with potential effects ramifications in exposed bears and the human consumers who eat them or their prey items such as ringed seals (Letcher et al. 2009a; Fisk et al. 2005).

Although the Arctic is far from large urban and industrial areas, it has relatively high levels of previously used, or “legacy” chemical contaminants including PCBs and organochlorine pesticides, such as DDT and chlordane. Recently and currently used chemicals include BFR materials such as PBDEs, which are used in furniture and electronics, have also found their way into polar bears and their Arctic food chain. Also there are other new, or “emerging” chemicals being found in polar bears and other Arctic wildlife including PFCs and their precursors, which are used in water-repellent coatings such as on carpets and furniture coverings as well as in non-stick coating for pots and pans.

Ecological tracers such as fatty acids (FAs) and stable nitrogen and carbon isotopes (SIs) are stable chemical or biochemical compounds that can be used to trace the movement of energy and contaminants through food webs, or between predator (polar bears) and prey (ringed seals and possibly other prey) (Iverson et al. 2004, 2006). Some FAs cannot be synthesized with high efficiency in higher trophic level organisms. Instead, these “essential” FAs are formed by primary producers and are passed up the food chain. Nitrogen and carbon SIs are routinely used in ecotoxicological studies. They can define organism trophic position, food source, and the chemical characteristics of habitats used by wildlife. Through the

integrated application of FA and SI tracers, we can gain insights into possible dietary and trophic level variations in polar bears and ringed seals from different populations.

Previous research has shown that polar bears have higher levels of legacy, and very recently emerging POPs, in their tissues than other animals such as fish because they are at the top of the Arctic food chain and they feed mainly on fat tissue of marine mammals (the tissue containing the largest amount of these chemicals). So, it is important 1) for polar bear health and 2) as polar bears are occasionally consumed in the diet of local people, to monitor the temporal and spatial levels and patterns of these chemicals. Regardless, there remains exceedingly little study understanding on the potential changes in the patterns and levels of e.g., BFRs and PFCs over time and on these time changes as a function of the Arctic location of the bears in question.

The emergence of PFCs as ubiquitous environmental contaminants is recent (Houde et al. 2006). We showed in the last monitoring cycle that several emerging POPs such as PFCs (e.g., PFOS and perfluorooctanoic acid (PFOA)) accumulated in the liver of bears spanning the Canadian arctic (Smithwick et al. 2005a, 2005b). There is only one known study on the temporal trends of PFCs in polar bears for any North American bear population. In liver samples of bears collected in 6 years between 1972 and 2002 from one Canadian location (Northern Baffin Island) and Barrow, Alaska, concentrations of PFOS and PFCAs with carbon chain lengths from C<sub>9</sub> to C<sub>11</sub> were reported to be exponentially increasing at both locations with doubling times ranging from  $3.6 \pm 0.9$  years for perfluorononanoic acid in the eastern group to  $13.1 \pm 4.0$  years for PFOS in the western group. PFOSA showed decreasing concentrations over time at both locations, while the remaining PFCs showed no significant trends or were not detected in any sample (Smithwick et al. 2006). Very recently a temporal trend study (1972-2006) on PFCs was completed for East Greenland polar bears (Dietz et al. 2008). Given the PFC changes reported in Canadian arctic ringed seals (Butt et al. 2007), that there has been a pan-Canadian spatial assessment for one time point for polar bears, and that there has been a temporal assessment (up

until 2002) for only one Canadian population, further spatial/temporal assessments of PFCs in polar bears are highly warranted.

PBDEs have also been reported in Arctic biota with increasing frequency (Braune et al. 2005; Letcher et al. 2009a) including in circumpolar polar bears (Muir et al. 2006; Sørmo et al. 2006); however the focus has generally been on Br<sub>4</sub> to Br<sub>8</sub> PBDE congeners, which largely comprise the PentaBDE and OctaBDE technical mixtures, and continue to be phased out from commercial use. Much less is known about in Arctic biota about higher brominated PBDEs, and especially BDE-209, which is the major constituent of the unregulated DecaBDE technical mixture (Braune et al. 2005). Verreault et al. (2005b) reported on PBDEs in plasma of Svalbard polar bear although BDE-209 was not detectable. Up until the present study, other important and bioaccumulative BFRs such as HBCD (total and not isomer-specific) have yet to be determined in Canadian polar bears (Letcher et al. 2009a). In fat samples collected in 2001-2002 we did report on total-HBCD in Alaskan, East Greenland and Svalbard bears, and levels were high and comparable to ΣPBDE concentrations (Muir et al. 2006). Clearly more spatial and temporal studies on HBCDs are necessary for Canadian bears. Furthermore, there are other current-use, non-PBDE BFRs that may also be of environmental relevance to the Arctic and present in polar bears.

The last study that examined spatial trends of metals and elements in Canadian polar bears was carried out in 2001-2002 (Rush et al. 2008). These studies showed that the levels of many metals, and especially the major metal contaminants Cd, Hg and Se varied between different regions of the Canadian arctic. For example, polar bears collected from the western Canadian arctic had lower concentrations of Cd but higher concentrations of Hg and Se than those collected from other regions. In light of the evidence of increasing Hg concentration in Arctic biota and the lack of temporal trend metal data for polar bears there is a need to assess the current levels of metals.

## Activities in 2008/2009

In 2008, and in collaboration in Nunavut and Northwest Territories (NWT) agencies and governments, remaining polar bear samples (fat, liver and muscle) for the present study were collected

by hunters in several of the fourteen participating communities via interaction with local Hunters and Trappers Associations (HTAs) and conservation officers. We have now obtained fat, liver and/or muscle samples from a total of 120 polar bears collected in the late winter/early spring 2007 and 2008 harvests of participating communities, HTAs, hunters, and Nunavut and NWT collaborators encompassed within the seven Canadian polar bear management zones. The samples collected were from 7 to 46 bears per zone, and comprised of adults and some sub-adults and males and females. Although adult females were desired, some males were collected and those POP levels were converted to females. The blubber of ringed seals (2004-2006) from Canadian populations complementary to the present polar bears were obtained via Dr. D.C.G. Muir for profiling of FAs (as well as SIs) as ecological tracers of diet in comparison to that of polar bears.

We have made substantial progress in 2008-2009 having completed all POP analyses (PCBs OCs, BFRs and PFCs, as well as identified precursor (of PFCs) and degradation (e.g., methyl sulfone-PCBs (MeSO<sub>2</sub>-PCBs)) products, of all polar bear tissue samples collected, as well as completing the determination of stable (nitrogen and carbon) isotopes and fatty acids for the same samples (i.e., ecological tracers). Age determinations of all (Canadian) bears from which samples were obtained were also completed in early 2009. The sample set sizes (per zone) and resulting data sets are now large enough to satisfy the numerical requirements of individual bears to allow for robust and reasonable power in statistical assessments of the two-time point comparisons and contrasts in PCB, OC, PFC and BFR patterns and levels. For longer term temporal (1991-2007) trends of legacy and emerging POPs in WHB bears we have completed all PCB, OC, PBDE/BFR and HBCD determinations as well as stable carbon isotope determinations. The years examined, and for which we exhausted available and archived fat biopsies, were 1991, 1992, 1994, 1995, 2001, 2003 and 2007), where we obtained fat samples for 9 to 15 individual bears (total of n=92 samples). Of the total number of samples, 90% were adults (> 5 years of age) and 10% were sub-adults (3 to 4 years of age). All samples for all years were collected from bears on the fall/winter.

We completed an exhaustive assessment for new and current-use BFRs that have been reported in the scientific literature in the environment, produced in high commercial/industrial volumes, described as replacements of PBDE technical formulations, and/or the commercial availability of analytical standards. As a result of this literature search, and in consultation with the NCP core monitoring study in ringed seals being led by D.C.G. Letcher, other BFR analytes monitored in the present polar bear fat included total-HBCD, PBEB, 1,4-bis(pentabromophenoxy)-tetrabromobenzene, BTBPE, DBDPE, HBB, pentabromobenzyl acrylate (PBBA), pentabromobenzyl bromide (PBBB) and PBT.

In 2008-2009, results from this NCP project and other polar bear projects were disseminated via papers in peer-reviewed scientific journals (Basu et al. 2009; Bechshøft et al. 2008, 2009; Gebbink et al. 2008a, 2008b; Letcher et al. 2009b; McKinney et al. 2009; Rush et al. 2008; Verreault et al. 2008) and reports (Letcher et al. 2008; Peacock et al. 2009). Results were also presented at relevant scientific conferences, Arctic workshops, meetings with northern/Arctic stakeholders and communities, and other venues which reached interested individuals and organizations including northerners in participating communities (see following list).

#### **Conference and Workshop Presentations in 2008-2009:**

McKinney, M.A., E. Peacock, M. Branigan, T.J. Evans, G.W. Gabrielsen, R. Dietz, C. Sonne and R.J. Letcher. 2009. (ORAL) Regional Differences and Temporal Patterns of Brominated Flame Retardants in Polar Bears from Alaska, Canada, Greenland and Svalbard. 11<sup>th</sup> Workshop on Brominated Flame Retardants (BFR'2009), May 19-20, Ottawa, ON, Canada.

McKinney, M.A., E. Peacock, M. Branigan, T.J. Evans, G.W. Gabrielsen, R. Dietz, C. Sonne, F.F. Rigét and R.J. Letcher. 2009. (POSTER) Chemical contaminants in polar bears can be influenced by location, time and the diet they eat. Nunavut Wildlife Symposium, March 19-20, Rankin Inlet, Nunavut.

Dietz, R., F.F. Rigét, C. Sonne, R.J. Letcher, M.A. McKinney, D. Muir, R. Bossi, K. Vorkamp and E. Born. 2008. (ORAL) Contaminant

linkages to climate parameters in polar bears (*Ursus maritimus*) from Greenland and Svalbard. Arctic Change 2008 Conference, Dec. 9-12, Québec City, Canada.

McKinney, M.A., E. Peacock and R.J. Letcher. 2008. (ORAL) Does sea ice-associated variation in diet influence the temporal trends of organohalogen concentrations in Western Hudson Bay polar bears? Arctic Change 2008 Conference, Dec. 9-12, Québec City, Canada.

McKinney, M.A., E. Peacock and R.J. Letcher. 2008. (ORAL) Climate change, sea ice-associated variation in diet and the influence the temporal trends of organohalogen concentrations in Western Hudson Bay polar bears. NWRC Seminar Series, Nov. 20, Carleton University, Ottawa, Canada.

Butt, C.M., S.A. Mabury, D.C.G. Muir, S. Sturman, A.D. Morris, R.J. Letcher, S.-G. Chu, M.A. McKinney and B.M. Braune. 2008. (ORAL) Trophic magnification and food web dynamics of perfluorinated organic compounds in the Arctic. 29<sup>th</sup> Annual Society of Environmental Toxicology and Chemistry (SETAC) Meeting, Nov. 16-20, Tampa, FL, U.S.A.

Letcher, R.J., M.A. McKinney, E. Peacock, R. Dietz, C. Sonne, G.W. Gabrielsen, S.-G. Chu, M. Branigan, M. Dyck, F.F. Rigét, D.C.G. Muir and T.J. Evans. 2008. (ORAL) The Current State of Contaminants Monitoring and Research in Polar bears from Canadian and Other Circumpolar Populations. 16<sup>th</sup> Annual Results Workshop of the Northern Contaminant Program (NCP), Canadian Arctic Contaminants Assessment Symposium, Sept. 22-25, Yellowknife, NWT, Canada.

Dietz, R., F.F. Rigét, C. Sonne, R.J. Letcher, M.A. McKinney, D. Muir, R. Bossi, K. Vorkamp and E.W. Born. 2008. (ORAL) Temporal trends and bioaccumulation of selected contaminants in East Greenland ringed seal and polar bears. 5<sup>th</sup> Society of Environmental Toxicology and Chemistry (SETAC) World Congress, Aug. 3-7, Sydney, Australia.

Letcher, R.J., S.G. Chu, E. Peacock, R. Dietz, C. Sonne, G.W. Gabrielsen, M.A. McKinney, M. Branigan, M. Dyck, D. Muir and T.J. Evans. 2008. (ORAL) Spatial distribution and temporal



comparisons of bioaccumulative polyfluoroalkyl contaminants in polar bears from circumpolar populations. 5<sup>th</sup> Society of Environmental Toxicology and Chemistry (SETAC) World Congress, Aug. 3-7, Sydney, Australia.

Letcher, R.J., S.G. Chu, E. Peacock, R. Dietz, C. Sonne, G.W. Gabrielsen, M.A. McKinney, M. Branigan and T.J. Evans. 2008. (POSTER) Associations between perfluorinated carboxylate and sulfonate contaminants and precursors in polar bears from circumpolar populations. 5<sup>th</sup> Society of Environmental Toxicology and Chemistry (SETAC) World Congress, Aug. 3-7, Sydney, Australia.

McKinney, M.A., E. Peacock, M. Dyck, M. Branigan, T.J. Evans, G.W. Gabrielsen, R. Dietz, C. Sonne, F.F. Rigét, D. Muir and R.J. Letcher. 2008. (ORAL) Use of dietary tracers in the interpretation of current geographical organohalogen contaminant spatial trends in polar bears. 5<sup>th</sup> Society of Environmental Toxicology and Chemistry (SETAC) World Congress, Aug. 3-7, Sydney, Australia.

McKinney, M.A., E. Peacock, M. Branigan, T.J. Evans, G.W. Gabrielsen, R. Dietz, F.F. Rigét, C. Sonne, and R.J. Letcher. 2008. (POSTER) Temporal patterns of chlorinated and brominated contaminants in polar bears from western Hudson Bay, Canada and other circumpolar populations. 5<sup>th</sup> Society of Environmental Toxicology and Chemistry (SETAC) World Congress, Aug. 3-7, Sydney, Australia.

Letcher, R.J. 2008. (ORAL) Comparative toxicokinetics of chlorinated and brominated contaminants in captive Greenland sled dogs and potential as a surrogate for wild polar bears. Integrated Toxicology & Environmental Health Program Seminar Series, April 18, Duke University, Durham, NC, U.S.A.

## Results

For those polar bear samples collected in 2007, preliminary data had been previously reported in Letcher et al. (2008). For all 2007 and 2008 collected samples, it was found that the effect of sex on log(e)-transformed concentrations of  $\Sigma$ CHL, *p,p'*-DDE, and  $\Sigma$ PCB concentrations was independent of region and age, therefore the data for the solitary females and females with cubs were first

standardized to adult males by multiplying the untransformed data by a standardization coefficient. For the present 2007-2008 spatial/regional and temporal assessments, we initially did apply the same male-to-female conversion factors (standardization coefficients) as reported in Norstrom et al. (1998) for the legacy organochlorine data. However, for e.g., PBDEs, HBCD, PFSA and PFCAs, where we found no significant difference between males and females, no male-to-females conversion was applied. This is consistent with the lack of male/females differences for PBDEs, HBCD, PFSA and PFCAs as previously reported in circumpolar polar bears (Dietz et al. 2007, 2008; Smithwick et al. 2005). With all contaminant data sets now completed, and with the attainment in 2009 of all ages for, especially NWT and Nunavut bears, we are presently re-visiting and assessing any effects of age and sex on the variability of contaminants in polar bears (2007-2008).

Across the seven management zones,  $\Sigma$ ClBz,  $\Sigma$ CHL and  $\Sigma$ MeSO<sub>2</sub>-PCB concentrations were not significantly different.  $\Sigma$ PCB concentrations were also spatially similar with only SHB concentrations being significantly ( $p < 0.05$ ) higher than for LS/JS bears.  $\Sigma$ PBDE concentrations were more geographically variable (Figure 1).  $\Sigma$ PBDE concentration was highest in bears from SHB followed by WHB and DS. The major PBDE congeners detected in polar bears from all regions were BDE47 and 153 (combined >80% of the  $\Sigma$ PBDE concentrations), with lesser amounts of BDE99 and BDE100. Pentabromobiphenyl (BB) 153 was the major single contaminant in all populations and at levels comparable to  $\Sigma$ PBDE concentrations. Total- $\alpha$ -HBCD concentrations were at low ppb levels for bears from all locations except the GB/FB (<0.2 ng/g lipid weight (lw)). The highest total- $\alpha$ -HBCD levels were found in SHB polar bears. BDE209, BTBPE and DBDPE were infrequently detected (23%, 18% and 9% of samples, respectively) at levels close to the quantitative detection limits, which prevented accurate quantification.

$\Sigma$ PFSA concentrations were comprised of >99% PFOS. The  $\Sigma$ PFCA concentrations were comprised largely of C<sub>9</sub> (PFNA), C<sub>10</sub> (PFDA) and C<sub>11</sub> (PFUdA), with much lesser amounts of C<sub>8</sub> (PFOA), C<sub>12</sub> (PFDoA) and C<sub>13</sub> (PFTrA), and very low or non-detectable C<sub>6</sub> (PFHxA), C<sub>7</sub> (PFHpA), C<sub>14</sub> (PFTeDA) and C<sub>15</sub> (PFPA). Of all the Nunavut

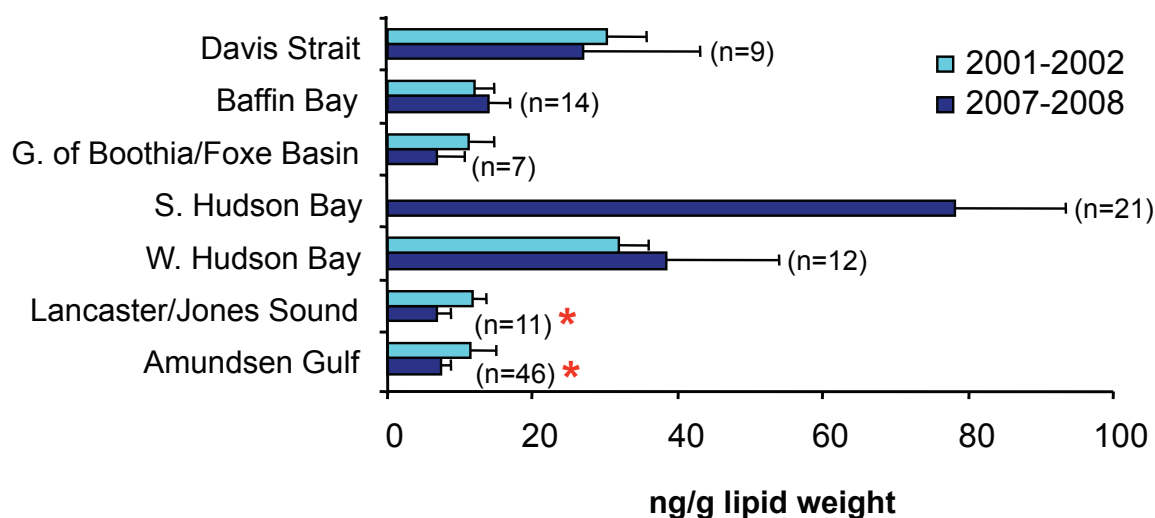


Figure 1. Two point temporal comparisons of  $\Sigma$ PBDE concentrations in Canadian polar bear populations between 2001-2002 (Muir et al. 2006; Southern Hudson Bay data not available) and the present 2007-2008 levels. Asterisks mark significant ( $p < 0.05$ ) decreases from 2001-2002 to 2007-2008.  $\Sigma$ PBDE = BDE47, 99, 100, 153 as these were the only congeners consistently detected in all samples.

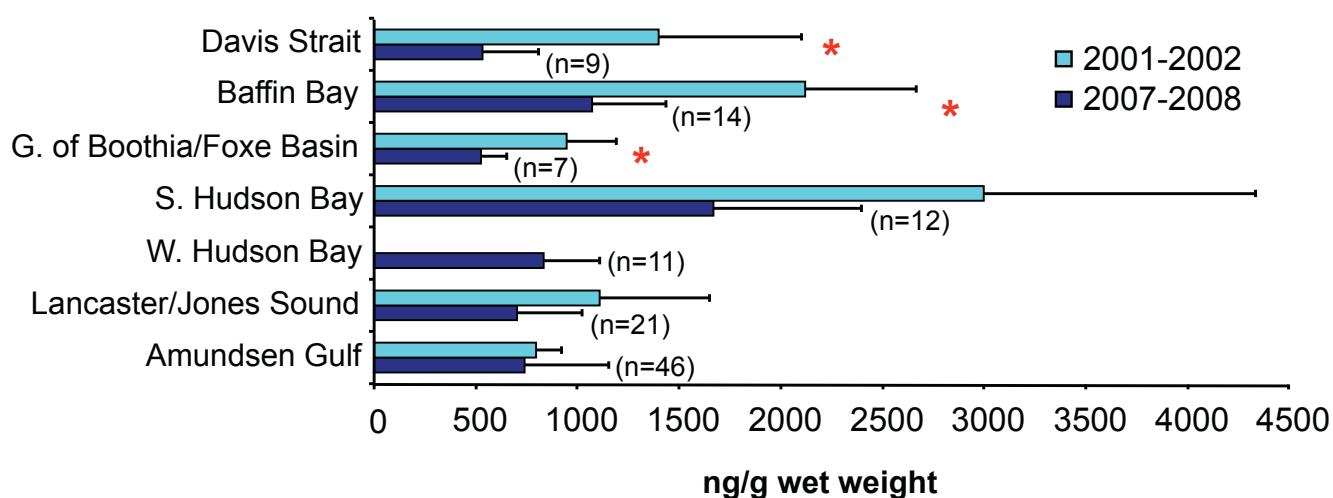


Figure 2. Two point temporal comparisons of perfluorooctane sulfonate (PFOS) concentrations in Canadian polar bear populations between 2001-2002 (Smithwick et al. 2005; Western Hudson Bay data not available) and the present 2007-2008 levels. Asterisks denote significant ( $p < 0.05$ ) decreases from 2001-2002 to 2007-2008.

and NWT samples, the 10:2 FTUCA (FDUEA) was barely quantifiable in all but one NWT bear. FTOHs were not detectable in any bear sample, and levels of PFOSA were in the low ng/g (wet weight) range but quantifiable with very high frequency in samples. Spatial trends for the 2007-2008 samples showed that  $\Sigma$ PFSA (mainly PFOS; Figure 2),  $\Sigma$ PFCA and PFOSA levels were highest in bears from SHB, and lower but

comparable bears from the other six Canadian population management zones.

Two time-point comparisons of POP concentrations are currently being assessed for data from the NCP II assessment from 2001-2002 (Letcher et al. 2008; Muir et al. 2006; Smithwick et al. 2005; Verreault et al. 2005a) and the present NCP III assessment. For example, in comparing  $\Sigma$ PBDE concentrations for 2001-2002 (Muir et al. 2006)



and the present 2007-2008 data for bears from the seven Canadian management zones, concentrations are similar with the exception of SHB due to the lack of data for this population from 2001-2002 and LS/JS and AG where concentrations are significantly ( $p < 0.05$ ) lower in 2007-2008 (Figure 1). For PFOS, the trend was also for lower levels in 2007-2008 as compared to 2001-2002 (Figure 2). PFOS concentrations were significantly lower for BB, DS and GB/FB.

Dietary tracer analyses were also measured for both polar bear and selected ringed seals from the Canadian populations. Measurements of  $\delta^{15}\text{N}$ ,  $\delta^{13}\text{C}$ , and FA profiles showed distinct patterns between the different Canadian polar bear populations and thus trophic as well as dietary differences from some bear populations. Based on deviations from a ringed seal blubber diet, trophic level and diet factors were shown to influence the spatial differences of the levels of several POPs. For example, the apparent west to east increasing trend among population for both  $\Sigma\text{PCB}$  and  $\Sigma\text{PBDE}$  concentrations is changed when adjusting for diet and trophic level as measured by variations among populations of FA and SI values in polar bears relative to ringed seals.

For bear samples from WHB spanning 1991-2007, as described in McKinney et al. (2009) carbon SIs were used to discern changes in the proportions of benthic versus pelagic prey species. Since FA signatures in mammalian predators reflect the FA composition of their diet, we used changes in polar bear FA signatures to evaluate alterations in the consumed proportions of prey species. We examined the relationships of  $\delta^{13}\text{C}$  and FA signatures to ice breakup date in WHB. We then determined the impact of this dietary variation on the temporal trends of persistent organic pollutants in WHB bears.

When assessing the annual percent change, concentrations of  $\beta\text{-HCH}$  and  $\Sigma\text{PBDE}$  increased over time (Figure 3, black bars), whereas  $\Sigma\text{DDT}$  decreased.  $\Sigma\text{PCB}$  and  $\Sigma\text{CHL}$  showed non-significant increases. However, when we controlled for diet, the rate of concentration change over time was altered (Figure 3, white bars). Diet changes over time resulted in a more rapid increase of  $\beta\text{-HCH}$  (by 20%) and  $\Sigma\text{PBDE}$  (by 28%) and reversed the direction of  $\Sigma\text{PCB}$  and  $\Sigma\text{CHL}$  temporal change from decreasing to increasing trends.

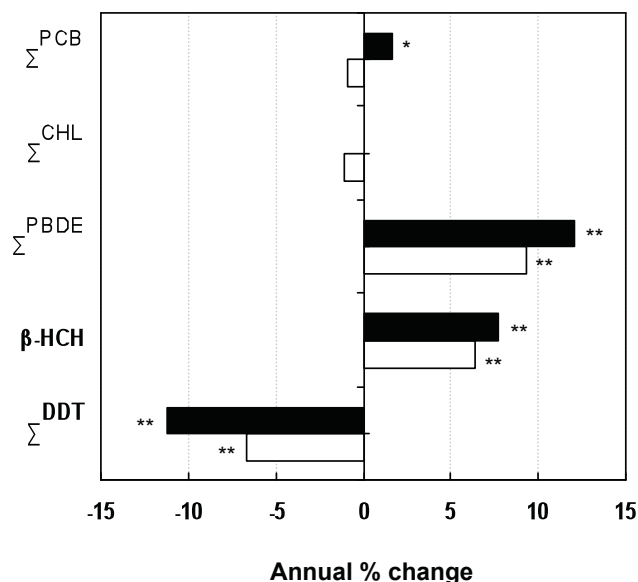


Figure 3. Annual percent (%) change in persistent organic contaminant concentrations. Temporal trends controlled for diet (white bars) were calculated as  $(1-10^\beta) \times 100\%$ , where  $\beta$  was the slope of the correlation between the contaminant concentration and the year, from a multiple regression of the log-transformed contaminant concentration (lipid weight basis) versus the dependent variables,  $\delta^{13}\text{C}$ , FA Index 1, FA Index 2 and year. Temporal trends with the influence of diet included (i.e. the actual trends) (black bars) were calculated as  $(1-10^\beta) \times 100\%$ , where  $\beta$  was the slope of the simple regression of the log-transformed contaminant concentration versus year. All statistically significant correlations are indicated by: \*  $p < 0.10$ , \*\*  $p < 0.05$ .

This is consistent with studies demonstrating relatively lower contaminant levels in bearded seals and our observed diet shift over time. In contrast, diet change resulted in a more rapid decline of  $\Sigma\text{DDT}$  (by 64%).

## Discussion and Conclusions

This assessment is providing new data on the current geographical patterns of legacy OCs and PCBs, as well as for newer contaminants found at quantifiable levels and with high frequency in polar bear samples such as PBDEs and total- $\alpha$ -HBCD and several PFCs. The present study is the first on the spatial trends of HBCD in Canadian polar bears, and indicated low ppb levels (0.68 to 5.1 ng/g lw) across the Canadian regions. Bears from SHB had the highest burdens of both PBDEs and total- $\alpha$ -HBCD, followed by WHB and DS. Other less-studied BFRs, including BDE209, BTBPE and

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DBDPE were infrequently detected and near the limit of quantification. These analytes are not reported on further here, but should be monitored in future assessments.

There appears to be no clear temporal differences in PBDE concentrations for each of the Canadian bear populations comparing 2001-2002 to 2007-2008 concentrations, with the exception of lower recent levels for LS/JS and AG bears. However, an observed shift to a greater proportion of BDE153 relative to BDE47 may be due to a greater arctic lag-time for BDE153, or the decrease in BDE47 emissions from phase-out of the PentaBDE product, possibly in combination with more rapid environmental or biological degradation of BDE47. We are currently examining the hepatic *in vitro* depletion (metabolism) capacity of polar bears to metabolically degrade various BFRs including PBDE congeners (e.g., BDE47 and BDE209), HBCDs and DBDPE.

The present PFOS results for the 2007-2008 collected samples are highly consistent with results reported previously for Canadian bears collected in 2001-2002 (Smithwick et al. 2005b). That is, PFCs were highly dominated by PFOS, and with lesser but substantial levels of PFNA, PFDA and PFUdA), and with much lesser amounts of PFOA, PFDoA, PFTrA and PFOSA. Spatial trends for the 2007-2008 samples indicate that ΣPFSA (PFOS), ΣPFCA and PFOSA levels are highest in bears from SHB, and lower but comparable among the AG, GB/LS, DS and BB collected samples. We are currently examining the hepatic *in vitro* depletion (metabolism) capacity of polar bears to metabolically degrade PFOSA to PFOS.

We have focused more in-depth on temporal trends of various POPs in WHB bears spanning the years 1991-2007 where archived samples were available. Recent FA research has demonstrated that the WHB bear diet consists of two ice-associated prey, ringed and bearded seal (*Erignathus barbatus*), and two open water-associated prey, harbour (*Phoca vitulina*) and harp seal (*Phoca groenlandica*). Here, we found that  $\delta^{13}\text{C}$  values decreased in WHB bears with earlier ice breakup date. This is consistent with a lower proportion in the diet of benthic-foraging bearded seals and consequently a relative increase in one or more of the other prey in years with a shortened period of ice cover. FA indices increased with earlier ice breakup date. The FAs contributing most to the variation were  $\alpha$ -linolenic acid (ALA),

docosapentaenoic acid (DPA), docosahexaenoic acid (DHA), eicosapentaenoic acid (EPA) and gamma-linoleic acid. Proportions of ALA, DPA and DHA are higher in harbour and harp seals than in bearded seals, whereas EPA and gamma-linoleic acid are lower. Thus, both the  $\delta^{13}\text{C}$  and FA signatures suggest decreases in bearded and increases in harbour and harp seal consumption in years with a shorter ice season. This is consistent with an known and observed diet shift for WHB bears over the period of 1994 to 2004, but may also reflect more extensive food web changes in the Hudson Bay ecosystem.

To our knowledge, this is the first examination for any Arctic species that has shown a link in the variation of sea ice and diet to altered POP exposure (McKinney et al. 2009). An increasing ratio of open-water seals to ice-associated seals in the polar bear diet is in the direction predicted by climate change. The observed change in diet resulted in greater contaminant levels in WHB bears over time (except ΣDDT) relative to levels if diet had remained constant. These results suggest that although some of the effects of climate change will be direct, others may also involve interaction with different stressors such as anthropogenic contaminants, which may exacerbate the decline in health and survivability of already sensitive populations and species.

This project is ongoing. For 2007-2008 bears from all Canadian management zones, we are currently examining branched and linear chain PFOS isomers and spatial trends of isomer levels and profiles. We are also currently determining various metals (e.g., Hg) and elements in livers of bears from Canadian management zones to assess spatial and temporal trends.

## Expected Project Completion Date

All aspects of the project (e.g., sample collection, data generation, data compilation and interpretation, data interpretation and writing of papers) are expected to be completed by December 2010. The exception may be the full publication of some papers in peer reviewed scientific journals, which may occur in 2011.

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## Acknowledgments

The project leaders and team members thank all individuals (e.g. northern peoples, field biologists and students) and agencies that participated in this project. The collection of polar bear tissues in Nunavut for contaminant analysis was initiated and carried out by the Department of Sustainable Development (Government of Nunavut), which was included as part of the normal sampling of all polar bear that are collected as part of the traditional hunt by northern peoples. Many thanks to the hunters and trappers organizations and conservation officers for coordinating sample collection: Allen Niptanatiak, Dustin Fredlund, Joe Ashevak, Joe Savikataaq, Jeffrey Qaunaq, Alden Williams, Paul Prefontaine, Kimball Nogier, Andrew Nakashuk, Louisa Kudluk, Noah Nakoolik, Ema Qaggutaq, Teema Palluq, Nancy Amarualik, Tabitha Mullin, Bruce Jerry Hainnu, George Koonoo, Teema Palluq and Johanne Coutu-Autut.

All appropriate permits were obtained (Nunavut Wildlife Research Permit). The collection of polar bear tissues in Northwest Territories was carried out by the Department of Resources, Wildlife and Economic Development, Government of the Northwest Territories. Appropriate permits and community approval were approved and granted by the Inuvialuit Game Council and appropriate Hunters and Trappers Committees. Collection of samples was carried out exclusively by hunters in the NWT and Nunavut communities. The project therefore relies heavily on the knowledge and experience of these hunters for its samples and for the ecological information on behavior, condition and population numbers they provide to wildlife officers and biologists.

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# Temporal Trends of Halogenated Chemicals of Emerging Concern in Beluga Whales (*Delphinapterus leucas*) from Hendrickson Island and Pangnirtung

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## Abstract

This long-term study is examining the temporal trends of halogenated chemicals of emerging concern in beluga whales from two sites: Hendrickson Island (HI) and Pangnirtung. Our time-series dates back to the early 1980s and the resolution or frequency of our time-series is particularly strong for samples collected post-2000: in the HI animals, for example, our data-set includes collections from 1984, 1993 and every year from 2000 to 2008. Similarly, for Pangnirtung, our data-set includes animals sampled in 1982, 1986, 1992, 1995 and every year from 2000 to 2008 with the exception of 2003 and 2004. The chemicals of interest include the suite of fluorinated surfactants, brominated flame retardants and short-chain chlorinated paraffins (SCCPs). For the brominated diphenyl ethers (BDEs) measured in blubber, the profiles of the animals from both locations were dominated by BDE-47 (~ 50%) followed by BDE-99 and -100. For beluga from HI, there was a significant ( $p < 0.05$ ) but small increase in total ( $\Sigma_6$ ) BDEs (sum of congeners: -47, -85, -99, -100, -153 and -154) over our study time period. The rate of

## Résumé

Cette étude à long terme se penche sur les tendances temporelles des produits chimiques halogénés qui posent de nouvelles préoccupations chez les bélugas de deux sites : l'île Hendrickson (IH) et Pangnirtung. Notre série chronologique remonte au début des années 1980 et la résolution ou fréquence de cette série est particulièrement solide pour les échantillons recueillis après 2000 : chez les animaux de l'IH, par exemple, notre ensemble de données comprend des prélèvements effectués en 1984, 1993 et tous les ans entre 2000 et 2008. Pour ce qui est de Pangnirtung, notre ensemble de données comprend des prélèvements effectués en 1982, 1986, 1992, 1995 et tous les ans entre 2000 et 2008, à l'exception de 2003 et 2004. Les produits chimiques d'intérêt incluent un ensemble d'agents de surface fluorés, des ignifugeants bromés et des paraffines chlorées à chaîne courte (PCCC). En ce qui concerne les éthers de diphenyle bromés (EDB) mesurés dans la graisse, le profil des animaux des deux sites était dominé par l'EDB-47 (~ 50 %), suivi de l'EDB-99 et -100. Dans le cas du béluga de l'IH, on a observé une augmentation ( $p < 0,05$ ) du total ( $\Sigma_6$ ) des EDB (somme des



increase of  $\Sigma_7$ BDEs in animals from HI was  $0.36 \pm 0.1$  [slope  $\pm$  standard error (SE)] ng/g per year.  $\Sigma_6$ BDE concentrations in Pangnirtung beluga increased in an exponential manner with a calculated doubling time of 12 years. These results suggest that although the flame retardant industry have phased out the production of two formulations, *penta*- and *octa*-BDE, the *deca*BDE formulation, which is still being used in North America, is likely contributing to the continued increase in BDE concentrations observed in beluga from our two sampling locations. The concentration of  $\Sigma_2$ HBCD (sum of  $\alpha$ - and  $\beta$ -isomers) in animals from HI also showed a small but insignificant increase ( $p=0.07$ ) over our study time period. Average concentrations of  $\Sigma_2$ HBCD in the blubber were generally  $10\times$  smaller than  $\Sigma_6$ BDEs and ranged from  $0.34 \pm 0.57$  ng/g (lw: geometric mean  $\pm$  SE) in 1993 to a peak in 2008 of  $2.53 \pm 0.26$  ng/g (lw). For animals from Pangnirtung,  $\Sigma_2$ HBCD concentrations increased linearly over our study period ( $p<0.01$ ) with a rate of increase of  $0.04 \pm 0.01$  ng/g per year. Conflicting results were observed for liver-based concentrations of  $C_8$ - $C_{12}$  perfluorocarboxylic acids ( $\Sigma_5$ PFCA) in animals from our two study sites: Pangnirtung beluga showed a small but significant increase ( $r^2=0.1703$ ,  $p<0.001$ ) in  $\Sigma_5$ PFCA concentrations with an annual rate of increase of  $1.8 \pm 0.5$  ng/g (slope  $\pm$  SE) while there was a strong negative relationship between liver based concentrations and sampling year in animals from HI ( $\Sigma_5$ PFCA =  $-7.4 \times \text{Year} + 14892$ ,  $r^2=0.4738$ ,  $p<0.001$ ). Interestingly, from 1980s to 2000, liver based concentrations of PFOS increased linearly in animals from both study sites ( $p<0.01$ ); the annual rate of increase was *ca.*  $2\times$  greater in animals from Pangnirtung ( $0.88 \pm 0.26$  ng/g) relative to HI animals ( $0.53 \pm 0.10$  ng/g). Liver-based concentrations of PFOS in beluga from both sites showed a significant rate of decrease post-2000. For example, in Pangnirtung, a rate of decrease of *ca.*  $2$  ng/g per year from 2000 to 2008 ( $r^2=0.1439$ ,  $p<0.05$ ) was observed while at HI an annual rate of decrease of  $0.93 \pm 0.40$  ng/g ( $r^2=0.0859$ ,  $p=0.02$ ) from 2003 to 2008 was observed. Taken together, this suggests that these populations are responding positively to the global reduction in emission of PFOS. Animals from Pangnirtung also showed a decrease in PFOSA, the metabolic

substances organoleptiques : -47, -85, -99, -100, -153 et -154) pendant la période de notre étude. Le taux d'augmentation des  $\Sigma_7$ EDB chez les animaux de l'IH a été de  $0,36 \pm 0,1$  [pente  $\pm$  erreur standard (ES)] ng/g par an. Les concentrations de  $\Sigma^6$ EDB chez le béluga de Pangnirtung ont augmenté de façon exponentielle avec un temps de doublement calculé de 12 ans. Ces résultats donnent à penser que, bien que le secteur des ignifugeants ait supprimé graduellement la production de deux formules, *penta*- et *octa*-EDB, la formule *déca*EDB, toujours utilisée en Amérique du Nord, contribue probablement à l'augmentation continue des concentrations d'EDB observée chez le béluga de nos deux sites d'étude. Les concentrations de  $\Sigma_2$ HBCD (somme des isomères  $\alpha$ - et  $\beta$ ) chez les animaux de l'IH ont aussi légèrement augmenté ( $p=0,07$ ) pendant la période de notre étude. Les concentrations moyennes de  $\Sigma_2$ HBCD dans la graisse étaient en général 10 fois plus petites que les  $\Sigma_6$ EDB et variaient de  $0,34 \pm 0,57$  ng/g (lw : moyenne géométrique  $\pm$  ES) en 1993 à une crête, en 2008, de  $2,53 \pm 0,26$  ng/g (lw). Pour ce qui est des animaux de Pangnirtung, les concentrations de  $\Sigma_2$ HBCD ont augmenté de façon linéaire pendant la période de notre étude ( $p<0,01$ ) avec un taux d'augmentation de  $0,04 \pm 0,01$  ng/g par an. Des résultats contradictoires ont été observés au niveau des concentrations dans le foie des acides perfluoro carboxyliques  $C_8$ - $C_{12}$  ( $\Sigma_5$ PFCA) chez les animaux des deux sites étudiés : le béluga de Pangnirtung montrait une petite augmentation significative ( $r^2=0,1703$ ,  $p<0,001$ ) des concentrations de  $\Sigma_5$ PFCA, avec un taux d'augmentation annuel de  $1,8 \pm 0,5$  ng/g (pente  $\pm$  ES) alors qu'il y avait une très forte relation négative entre les concentrations dans le foie et l'année d'échantillonnage chez les animaux de l'IH ( $\Sigma_5$ PFCA =  $-7,4 \text{ années} + 14892$ ,  $r^2=0,4738$ ,  $p<0,001$ ). Il est intéressant de noter qu'entre les années 1980 et 2000, les concentrations dans le foie de PFOS ont augmenté de façon linéaire chez les animaux des deux sites étudiés : ( $p<0,01$ ); le taux d'augmentation annuel était de *ca.*  $2$  fois plus élevé chez les animaux de Pangnirtung ( $0,88 \pm 0,26$  ng/g) que chez les animaux de l'IH ( $0,53 \pm 0,10$  ng/g). Les concentrations dans le foie de PFOS chez les bélugas des deux sites ont montré un taux important de diminution après 2000. Par exemple, à Pangnirtung, un taux de diminution de *ca.*  $2$  ng/g par an entre 2000 et 2008 ( $r^2=0,1439$ ,  $p<0,05$ ) a été observé alors qu'à l'IH, un taux de

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PFOS precursor of  $9.89 \pm 2.26$  ng/g per year from 2002 to 2008 ( $r^2=0.4054$ ,  $p<0.01$ ).

Concentrations of SCCPs exceeded those of any other chemical study especially in animals collected prior to 2000. From both sites,  $\Sigma$ SCCP concentrations decreased linearly at rates of *ca.* 30 and 8 ng/g per year in animals from HI and Pangnirtung, respectively.

diminution annuel de  $0,93 \pm 0,40$  ng/g ( $r^2=0,0859$ ,  $p=0,02$ ) a été observé entre 2003 et 2008. Ensemble, ces résultats donnent à penser que ces populations réagissent positivement à la réduction générale des émissions de PFOS. Les animaux de Pangnirtung faisaient aussi état d'une baisse de PFOSA, le précurseur métabolique des PFOS, de  $9,89 \pm 2,26$  ng/g par an entre 2002 et 2008 ( $r^2=0,4054$ ,  $p<0,01$ ).

Les concentrations de PCCC dépassaient celles des autres produits chimiques étudiés, notamment dans les prélèvements effectués avant 2000. Sur les deux sites, les concentrations de  $\Sigma$ SCCP ont baissé de façon linéaire aux taux de *ca.* 30 et 8 ng/g par an chez les animaux de l'IH et de Pangnirtung, respectivement.

## Key Messages

- Beluga from HI showed a small linear increase in total BDE concentrations in blubber *ca.* rate of increase = 0.36 ng/g per annum. Conversely, BDE concentrations in Pangnirtung beluga blubber increased in an exponential manner with a calculated doubling time of 12 years;
- Average HBCD concentrations in the beluga blubber from both study sites were generally 10× smaller than BDEs;
- Dissimilar trends were observed for liver-based concentrations of the PFCAs: beluga from Pangnirtung showed a small but significant increase in  $\Sigma$ PFCAs concentrations while animals from HI showed a strong decrease at a rate of *ca.* 7.4 ng/g per year;
- From the 1980s to 2000, liver based concentrations of PFOS increased linearly in animals from both study sites;
- Liver-based concentrations of PFOS in beluga from both sites showed a significant rate of decrease post-2000: for Pangnirtung, we observed a rate of decrease of *ca.* 2 ng/g per year from 2000 to 2008 while at HI an annual rate of decrease of *ca.* 1 ng/g was observed for 2003 to 2008.

## Messages clés

- On a noté chez le béluga de l'IH une légère augmentation linéaire des concentrations totales d'EDB dans la graisse *ca.* taux d'augmentation = 0,36 ng/g par an. Réciproquement, les concentrations d'EDB dans la graisse des bélugas de Pangnirtung ont augmenté de façon exponentielle, avec un temps de doublement calculé de 12 ans.
- Les concentrations moyennes d'HBCD dans la graisse des bélugas des deux sites étudiés étaient en général de 10 fois moindres que celles des EDB.
- Des tendances différentes ont été observées quant aux concentrations de PFCA dans le foie : les bélugas de Pangnirtung montraient une augmentation légère, mais significative des concentrations de  $\Sigma$ PFCA alors que les animaux de l'IH montraient une forte baisse du taux *ca.* 7,4 ng/g par an.
- Entre les années 1980 et 2000, les concentrations de PFOS dans le foie ont augmenté de façon linéaire chez les animaux des deux sites étudiés.
- Les concentrations de PFOS dans le foie des bélugas des deux sites ont affiché un taux de diminution significatif après 2000 : à Pangnirtung, nous avons observé un taux de diminution de *ca.* 2 ng/g par an entre 2000 et 2008 alors que sur l'IH, on a observé un taux de diminution annuel de *ca.* 1 ng/g entre 2003 et 2008.

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## Objectives

- To continue to build on our temporal trend data set for a suite of halogenated chemicals of emerging concern in beluga whales from Hendrickson Island (HI) and Pangnirtung;
- To identify any potential 'new' chemicals of emerging concern.

## Introduction

Prevailing conditions such as reduced levels of solar radiation, long periods of permanent sea-ice, and relatively low annual temperatures make the Circumpolar Arctic an ideal environment for persistent chemicals to remain chemically unchanged for long-periods. Persistent organic chemicals produced in regions far removed from the Arctic can be delivered there *via* the atmosphere either in the gaseous phase and/or bound to particulates or, if non-volatile, by ocean currents. That many Arctic marine food chains are long facilitates the bioaccumulation of many persistent organic pollutants. The end result is that animals at upper trophic levels (TLs) of a food chain can accumulate high concentrations of many of these chemicals.

Muir and Howard and Brown and Wania have compiled a list of potential chemicals of emerging concern (CECs) (1,2). Considering that there are over 20 000 chemicals in commerce today, it is perhaps not too surprising that the list of CECs has the potential to be quite extensive. Unfortunately, our knowledge of the contemporary and/or historical environmental emissions of many CECs is quite sparse and the only means of constructing an emission profile is to examine concentrations in a well defined environmental compartment. Because of their abundance, ecological significance and that they are top TL animals, beluga whales make an ideal bio-indicator species. The overriding assumption in using beluga whales as an indicator species to track emissions of chemicals is that changes in the inputs of any persistent chemical into the environment will be reflected by a similar concentration change in these animals. While it is thought that changes in climate could confound interpretations of temporal trend studies there is currently no method to control or correct for this variable.

This current study builds on our on-going annual sampling campaign and chemical analyses of

beluga whales for CECs from two locations in the Canadian Arctic: Hendrickson Island and Pangnirtung. In addition to the inclusion of a contemporary data point (2008), we have also enhanced the resolution of our time series by filling in data gaps in the post-2000 dataset. The CECs in our study are the *poly*- and *per*fluorinated compounds (PFCs), brominated flame retardants (BFRs) and short chain chlorinated paraffins (SCCPs).

## Activities in 2008/2009

**Samples investigated:** The animals selected for study were stored in our archived repository at Fisheries & Oceans Canada. Beluga from Hendrickson Island (HI: 1984, 1993, 1995, 2001, 2002, 2003, 2004, 2005, 2006, 2007 and 2008) and Pangnirtung (1982, 1986, 1992, 1995, 2002, 2005, 2006, 2007 and 2008) were the basis of our time-trend study. Table 1 shows the number of animals analyzed from each of the collection years; when available, ten male animals were selected for study.

**Chemical analysis:** Analytical methods adopted for our study have already been published in the peer-review literature. All analyses were performed in the toxic aquatic chemical laboratory at Fisheries & Oceans Canada, Winnipeg. Hydrophobic chemicals like BDEs, HBCD and SCCPs were analyzed in the blubber of animals as described previously(3,4). BDEs were analyzed by gas chromatography with electron capture negative ion (ECNI) low resolution mass spectrometry. Quantification of BDE congeners were done using external standard solutions. Total BDE congeners are based on the sum of -47, -85, -99, -100, -153 and -154. Isomers of HBCD were analyzed by liquid chromatography tandem mass spectrometry in the negative electrospray ionization mode. Total HBCD is based on the sum of the  $\alpha$ - and  $\gamma$ -diastereoisomers. SCCPs were analyzed by high resolution mass spectrometry. Total SCCP concentrations are based on the sum of C<sub>10</sub>-Cl<sub>3</sub> chain lengths. Fluorinated compounds were extracted and analyzed in the liver of the animals as described in Tomy *et al.*(5,6). Compounds investigated include perfluorooctanoate (C<sub>8</sub>: PFOA), perfluorononanoate (C<sub>9</sub>: PFNA), perfluorodecanoate (C<sub>10</sub>: PFDA), perfluoroundecanoate (C<sub>11</sub>: PFUA), perfluorododecanoate (C<sub>12</sub>: PFDoDA), perfluorooctane sulfonate (PFOS) and perfluorooctane

sulfonamide (PFOSA). Total perfluorinated carboxylate concentrations are based on the sum of C<sub>8</sub>-C<sub>12</sub>.

**Quality assurance:** Certified reference materials for BDEs in beluga blubber from the National Institute of Standards and Technology (SRM 1945) were used for each batch of 20 samples. The agreement between our measured BDE concentrations and the accepted SRM-1945 values were excellent. No reference materials are yet available for HBCD, SCCPs or for the fluorinated compounds. Our laboratory also participates in the NCP Quality Assurance Program.

**Statistical analyses:** For calculating arithmetic and geometric means, non-detect concentrations were replaced with half the method detection limit. Statistical treatment of the data was done using SigmaStat. The Q-test was used to remove outliers in the data-set(7).

## Results

Liver based concentrations of  $\Sigma_5$ PFCAs, PFOS, PFOSA and lipid based concentration of  $\Sigma_6$ BDEs,  $\Sigma_2$ HBCDs and  $\Sigma$ SCCPs in beluga whales from our two study sites are shown in Table 1 and 2.

### (a) Beluga from Hendrickson Island

Total SCCP concentrations were greater than any other chemical measured in the blubber in animals collected before 2000. For example, in 1993, geometric mean concentrations of  $\Sigma$ SCCPs in beluga blubber were *ca.* 65 and 1000 $\times$  greater than  $\Sigma_6$ BDE and  $\Sigma_2$ HBCD concentrations, respectively. The profiles of the SCCPs were dominated by the C<sub>10</sub> and C<sub>11</sub> homologs. Total BDE concentrations were greater than those of  $\Sigma_2$ HBCD for all sampling years and likely reflects the greater usage volumes of BDEs relative to HBCD. BDE concentrations were dominated by congener 47 which accounted for *ca.* 58% of the total BDE burden in the animals (profile analysis based on animals collected in 2000 and after). The overall rank order of BDE congeners were -47 > -99 > -100 > -85 > 153 $\approx$ 154. For the HBCD, the  $\alpha$ -isomer was consistently greater than that of  $\gamma$ -isomer. For the fluorinated compounds studied,  $\Sigma_5$ PFCA concentrations were generally greater than those of PFOS and its metabolic precursor, PFOSA, in animals collected before 2000.

Time trend analysis using the geometric mean data shows that for  $\Sigma_6$ BDEs there was a small but significant ( $p < 0.01$ ) increase in concentrations over our study period. While this increase was subtle relative to the increase observed for animals from Pangnirtung (*see below*) it does suggest that *deca*BDE, which is the only formulation still produced by industry and is known to degrade to lower BDE congeners in biota, is still contributing to the overall burden of BDEs in animals from the Canadian Arctic(8-10).

Similarly, the temporal trend profile for  $\Sigma_2$ HBCD shows a small but insignificant increase ( $p = 0.07$ ) in concentrations over our study period.

There was a strong inverse relationship between wet weight liver  $\Sigma_5$ PFCA concentrations and sampling year for the animals from Hendrickson Island (*see Figure 1*). Based on the regression analysis, the annual rate of decrease of total PFCAs was  $7.41 \pm 0.71$  ng/g ( $\pm$  SE in slope). For PFOS, there was linear increase ( $0.53 \pm 0.10$  ng/g, per year) in measured concentrations between 1984 to 2000 ( $r^2 = 0.4319$ ,  $p < 0.01$ ). After that time, we observed a small decrease in PFOS concentrations for 2001 and 2002 followed by a small but insignificant increase (Student *t*-test,  $p > 0.05$ ) in 2003. Concentrations declined in a linear manner after that time at a rate of *ca.* 1 ng/g per year ( $r^2 = 0.0859$ ,  $p = 0.02$ ).

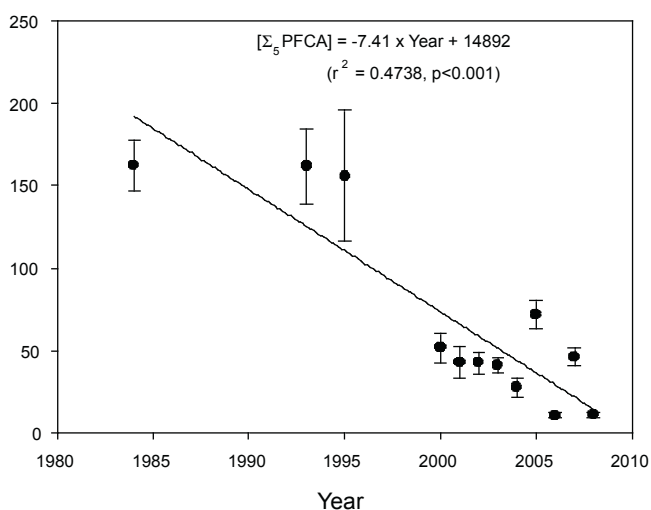


Figure 1. Temporal trend of  $\Sigma_5$ PFCAs in beluga whales from Hendrickson Island. Each data point represents the geometric mean  $\pm 1 \times$  standard error. Results of the regression analysis inserted as text in plot. See Table 1 for number of animals per collection year.

**Table 1. Geometric mean (GM), maximum (max) and minimum (min) concentrations (ng/g) of chemicals of emerging concern in beluga whales from Hendrickson Island<sup>a</sup>.**

Year	N		$\Sigma_6$ BDEs	$\Sigma_2$ HBCD	$\Sigma$ SCCPs	$\Sigma_5$ PFCAs	PFOS	PFOSA
1984 <sup>b</sup>	10	GM				156.83	4.85	48.09
		Min				98.35	2.23	8.47
		max				235.32	10.40	111.67
1993	10	GM	5.21	0.34	340.01	147.35	10.46	102.79
		min	3.61	0.086	114.88	71.71	4.37	65.72
		max	7.57	4.59	493.85	313.10	15.89	154.94
1995	10	GM	6.31	1.39	254.12	116.08	10.04	97.67
		min	3.76	0.88	44.02	35.01	5.0	44.19
		max	10.49	5.88	399.87	282.37	14.8	216.07
2000	10	GM	9.18	1.20	NC <sup>c</sup>	45.32	13.63	7.32
		min	4.27	0.095		22.92	8.58	3.87
		max	17.62	2.77		113.82	20.27	16.33
2001	11	GM	8.89	2.22	204.64	30.73	9.87	65.61
		min	1.54	0.75	30.38	6.02	5.22	31.90
		max	17.14	5.48	192.91	88.61	16.12	146.52
2002	10	GM	8.31	0.82	NC	37.17	8.08	22.26
		min	3.51	0.13		15.47	1.67	5.40
		max	18.50	1.72		78.63	24.26	51.74
2003	11	GM	11.63	2.15	NC	38.67	12.01	7.81
		min	7.02	1.28		21.68	5.71	4.56
		max	24.32	3.36		76.21	19.91	16.54
2004	10	GM	18.39	2.95	NC	21.57	11.21	6.96
		min	14.56	2.26		8.72	6.88	4.45
		max	25.94	3.68		50.32	24.12	12.05
2005	10	GM	4.77	1.70	248.98	67.16	11.91	127.01
		min	1.29	0.85	3.06	35.64	6.01	48.23
		max	15.69	3.32	430.26	122.69	23.04	228.40
2006	10	GM	8.63	1.17	3.96	9.72	7.98	22.51
		min	4.43	0.77	0.82	4.87	1.67	5.41
		max	17.10	2.98	13.90	16.03	24.26	51.74
2007	10	GM	8.91	1.37	NC	43.64	11.17	16.09
		min	1.86	0.62		29.77	4.25	7.76
		max	15.03	3.47		82.81	20.32	24.47
2008	10	GM	8.66	2.53	NC	9.88	6.71	8.59
		min	1.55	1.36		5.19	4.17	3.77
		max	22.76	3.97		21.31	11.67	14.03

<sup>a</sup> concentrations of  $\Sigma_6$ BDEs,  $\Sigma_2$ HBCD and  $\Sigma$ SCCPs were determined in the beluga blubber and are expressed on a lipid weight basis.  $\Sigma_5$ PFCAs, PFOS and PFOSA were measured in the liver and are expressed on a wet weight basis;

<sup>b</sup> blubber samples were not available for this year;

<sup>c</sup> NC = not complete.

**Table 2. Geometric mean (GM), maximum (max) and minimum (min) concentrations (ng/g) of chemicals of emerging concern in beluga whales from Pangnirtung<sup>a</sup>.**

Year	N		$\Sigma_6$ BDEs	$\Sigma_2$ HBCD	$\Sigma$ SCCPs	$\Sigma_5$ PFCAs	PFOS	PFOSA
1982	8	GM	3.50	0.17	199.04	9.56	10.67	21.27
		min	2.79	0.08	108.28	6.06	7.25	13.83
		max	6.23	0.97	320.12	27.19	19.30	31.24
1986	7	GM	3.61	0.09	161.49	5.07	14.24	42.26
		min	3.00	0.03	83.33	1.46	9.28	33.06
		max	5.01	1.72	513.90	12.79	20.19	47.41
1992	8	GM	6.24	1.17	330.28	9.49	20.61	48.12
		min	4.72	0.54	101.64	4.03	14.83	29.70
		max	9.63	2.01	724.19	22.30	31.40	74.03
1995	10	GM	5.60	0.04	116.90	14.52	14.58	44.64
		min	3.08	0.03	98.58	7.02	5.39	24.46
		max	15.68	0.61	282.48	24.65	43.43	120.43
2000	4	GM	17.14	1.51	NC <sup>b</sup>	95.14	29.09	16.71
		min	14.73	0.93		52.04	12.26	9.94
		max	20.89	2.63	NC	163.13	56.24	22.59
2001	3	GM	15.96	1.06		73.38	25.45	19.93
		min	14.46	0.99		64.50	18.30	18.11
		max	16.82	1.15		80.90	33.57	21.81
2002	8	GM	6.14	0.05	116.71	23.18	22.61	61.16
		min	3.68	0.02	92.48	11.79	14.64	23.76
		max	22.93	0.15	201.87	54.97	35.71	160.57
2005	10	GM	9.98	1.26	16.48	17.22	16.34	26.62
		min	3.49	0.88	3.29	2.40	3.81	12.59
		max	17.47	1.77	82.49	46.57	51.68	82.89
2006	4	GM	14.53	0.97	2.95	16.23	21.98	29.02
		min	10.55	0.65	1.10	6.69	12.71	20.16
		max	17.09	1.35	8.72	49.37	44.52	34.24
2007	5	GM	9.20	0.93	NC	43.08	9.43	26.03
		min	4.86	0.42		38.07	7.92	18.21
		max	18.18	1.92		47.60	11.40	40.87
2008	4	GM	21.46	2.57	NC	103.82	17.06	6.91
		min	15.90	1.54		53.95	9.02	4.80
		max	31.55	4.64		171.05	35.16	18.14

<sup>a</sup> concentrations of  $\Sigma_6$ BDEs,  $\Sigma_2$ HBCD and  $\Sigma$ SCCPs were determined in the beluga blubber and are expressed on a lipid weight basis.  $\Sigma_5$ PFCAs, PFOS and PFOSA were measured in the liver and are expressed on a wet weight basis;

<sup>b</sup> NC = not complete.



### (b) Beluga from Pangnirtung

Similar to what was observed in HI,  $\Sigma$ SCCPs was the dominant lipophilic CEC measured in beluga blubber from Pangnirtung and  $\Sigma_6$ BDE concentrations consistently exceeded those of  $\Sigma_2$ HBCD. The rank order of BDE congeners measured in blubber were: -47 > -99 > -100 > -153 > -85 > -154; the -47 congener accounted for *ca.* 70% of the total BDE burden (profile analysis based on animals collected in 2000 and after). For the fluorinated compounds, PFOS concentrations were generally smaller than those of  $\Sigma_5$ PFCA and PFOSA.

Time trend analysis of  $\Sigma_6$ BDEs in the blubber of animals from Pangnirtung shows a non-linear increase in concentrations over our study period (see Figure 2). Based on the coefficient of the exponentially fitted-curve a doubling time of *ca.* 12 years was calculated. This result corroborates the  $\Sigma_6$ BDEs time-trend we observed in animals from Hendrickson Island and suggests that BDEs concentrations are still increasing the beluga whales in the Canadian Arctic.

The temporal trend of  $\Sigma_2$ HBCD shows a significant increase ( $p < 0.05$ ) in concentrations over our study period. Regression analysis suggested a small annual rate of increase of  $0.04 \pm 0.01$  ng/g per year.

The liver based concentrations of  $\Sigma_5$ PFCA in beluga whales from Pangnirtung beluga show a

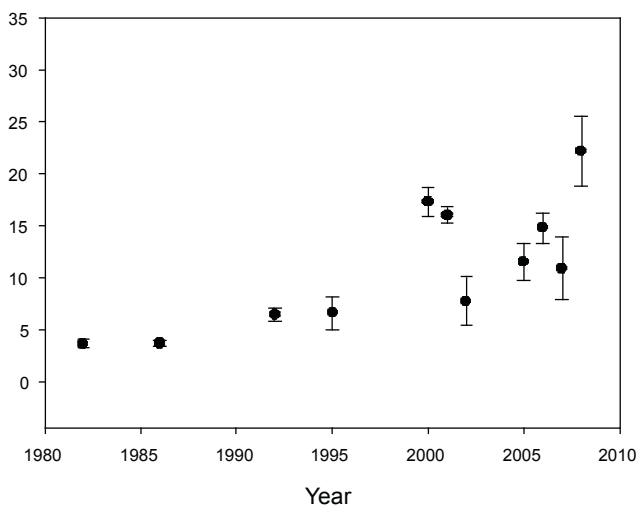


Figure 2. Temporal trend of  $\Sigma_6$ BDEs in beluga whales from Pangnirtung. Each data point represents the geometric mean  $\pm 1 \times$  standard error. See Table 2 for number of animals per collection year.

linear decrease ( $r^2 = 0.1703$ ,  $p < 0.01$ ) over our time period. While the measured rate of increase in liver was small *ca.* 1.8 ng/g per year, it does conflict with what we observed in beluga from Hendrickson Island. Reasons for this difference remain unclear. Conversely, we observed fairly good agreement with the PFOS temporal trend in beluga at both study sites: from 1980s to 2000 from Pangnirtung, liver based concentrations of PFOS increased linearly at an annual rate of  $0.88 \pm 0.26$  ng/g. This rate was *ca.*  $2\times$  greater than what we observed in animals from Hendrickson Island. After that time, liver-based concentrations of PFOS showed a significant linear decrease: the calculated annual rate of decreased was  $1.78 \pm 0.73$  ng/g ( $r^2 = 0.1439$ ,  $p < 0.05$ ). A similar observation was made for PFOSA, which also showed a linear decrease from 2003. The rate of decrease for PFOSA in the animals was more dramatic, *ca.* 10 ng/g per year ( $r^2 = 0.4054$ ,  $p < 0.001$ ).

## Activities That Are Ongoing

There are still gaps in the SCCP dataset which we hope to fill over the next few months. Progress has been made with the analysis of beluga blubber for polychlorinated naphthalenes (PCNs). Twenty animals from Hendrickson Island (10 each from 2000 and 2008) have been extracted and submitted to the Ministry of Ontario Environment (MOE, Toronto, ON) for PCN analysis. Work is ongoing to screen samples for previously unreported CECs.

## Acknowledgments

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# Mercury in Beluga, Narwhal and Walrus from the Canadian Arctic: Status in 2009

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## Abstract

Mercury levels in organs of beluga and walrus have been determined and added to a growing database on concentrations of these elements in arctic animals. The data on these animals offer opportunities to test for differences over time and geographic ranges as well as to compare among species and organs. Opportunities to test for temporal changes at locations of interest will grow as future collections add additional data. Mercury content varies from organ to organ and animal to animal from a given site and year. Of the organs analyzed in this study, usually the liver has the highest concentrations of mercury, followed by kidney, muscle and muktuk. Estimation of temporal change is complicated by the fact that mercury accumulates with age so that older animals usually have higher levels than younger ones from the same location. The role of age was complicated for beluga when it was learned that these whales form one growth layer group per year in their teeth, not two as had been assumed previously (Stewart et al, 2006). Previously reported ages have been doubled. Since the basis of comparison of mercury levels among different groups of beluga

## Résumé

Les niveaux de mercure dans les organes des bélugas et des morses ont été déterminés et versés dans une base de données en construction sur les concentrations de ces éléments dans les mammifères arctiques. Les données sur ces animaux offrent la possibilité de vérifier les différences au fil des ans et en fonction des régions géographiques, et de les comparer entre espèces et organes. Les occasions de vérifier les changements temporels aux points d'intérêt augmenteront alors que d'autres collectes ajouteront de nouvelles données. La teneur en mercure varie d'un organe à l'autre et d'un animal à l'autre dans un lieu donné, au cours d'une année donnée. Parmi les organes analysés dans le cadre de cette étude, le foie est celui qui, en général, présente les plus fortes concentrations de mercure, suivi des reins, des muscles et du muktuk. L'estimation du changement temporel se complique du fait que le mercure s'accumule avec l'âge, ce qui fait que les animaux plus âgés présentent habituellement des niveaux plus élevés que les jeunes d'un même lieu. Le rôle de l'âge s'est compliqué chez le béluga lorsque nous avons appris que ces baleines forment un groupe de couches de croissance par an dans les dents et non

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includes an adjustment for differing ages, age data are critical. Usually the chemical analyses are completed prior to the age determinations and so there is a lag in the interpretations. This year, a simplified analysis has been used as further age data are anticipated. Mercury and selenium in liver are related statistically with consistent indications that slopes of mercury on selenium are about 0.7 indicating a molar surplus of selenium over mercury. This surplus appears to be exaggerated in walrus, notably in kidney.

## Key Messages

- New collections of beluga and walrus became available during 2008/09. However, ages for some animals are not available yet.
- Regardless of ages, the levels of total mercury in beluga liver and kidney remain higher than those used to regulate the sale of commercial fish (0.5 ug/g). The average level in liver for beluga from Pangnirtung in 2008 was 9.79 ug/g and in kidney 5.12 ug/g. Mean levels in beluga from Sanikiluaq in 2008 were 5.88 and 1.64 ug/g respectively and levels in muscle averaged 0.67 ug/g. Similarly, levels in beluga from Arviat in 2008 were 11.0 ug/g in liver, 3.23 ug/g in kidney and 0.87 ug/g in muscle. Levels in beluga from Hendrickson Island remained high in 2008 with concentrations of 22.7 ug/g in liver, 5.32 ug/g and 1.14 ug/g in muscle. The only new samples of muktuk obtained were from Hendrickson Island and these averaged 0.37 ug/g.
- Levels of mercury in walrus from Hall Beach in 2008 averaged 3.08 ug/g, another relatively high value for this species and location.
- The question of temporal change in levels of mercury is of great interest but it is complicated by a relationship between mercury in liver and age of beluga and walrus. We lack age data for some collections reported here. Year-to-year variation in levels can be quite

pas deux comme on l'avait cru jusqu'ici (Stewart et coll., 2006). Les âges qui avaient été indiqués jusqu'alors ont été doublés. Étant donné que la base de comparaison des niveaux de mercure chez les différents groupes de bélugas inclut un rajustement du fait des différences d'âge, les données sur l'âge sont importantes. Habituellement, les analyses chimiques se font avant la détermination de l'âge, ce qui entraîne un délai dans l'interprétation. Cette année, une analyse simplifiée a été utilisée du fait que l'on attend d'autres données sur les âges. Le mercure et le sélénium dans le foie sont liés, sur le plan statistique, avec des indications constantes que les pentes du mercure par rapport au sélénium sont d'environ 0,7 ce qui indique un surplus molaire de sélénium par rapport au mercure. Ce surplus semble exagéré chez le morse, surtout dans les reins.

## Messages clés

- De nouvelles données sur les bélugas et les morses ont été publiées en 2008-2009. Toutefois, l'âge de ces animaux n'est pas encore disponible.
- Peu importe l'âge, les niveaux de mercure total dans le foie et le rein des bélugas sont plus élevés que ceux utilisés pour le contrôle de la vente commerciale du poisson (0,5 ug/g). Le niveau moyen dans le foie du béluga de Pangnirtung en 2008 était de 9,79 ug/g et dans les reins, de 5,12 ug/g. Les niveaux moyens dans le béluga de Sanikiluaq en 2008 étaient de 5,88 et de 1,64 ug/g respectivement et les niveaux moyens dans les muscles étaient de 0,67 ug/g. De la même façon, les niveaux dans le béluga d'Arviat en 2008 étaient de 11,0 ug/g dans le foie, de 3,23 ug/g dans les reins et de 0,87 ug/g dans les muscles. Les niveaux dans le béluga de l'île Hendrickson sont demeurés élevés en 2008, avec des concentrations de 22,7 ug/g dans le foie, de 5,32 ug/g et de 1,14 ug/g dans les muscles. Les seuls nouveaux échantillons de muktuk ont été prélevés dans l'île Hendrickson et leur moyenne était de 0,37 ug/g.
- Les niveaux de mercure chez le morse de Hall Beach, en 2008, étaient en moyenne de 3,08 ug/g, une autre valeur relativement élevée pour cette espèce et cet emplacement.
- La question du changement temporel des niveaux de mercure est très intéressante, mais elle se complique du fait de la relation entre le

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large and this makes it difficult statistically to discriminate between temporal variation and temporal trend.

- Strong statistical linkages exist between mercury and selenium in liver of both beluga and walrus (and narwhal based on the report in 2008). The slopes of regressions of micromolar concentrations of mercury on selenium differ among species with walrus from Igloodik having strikingly lower slopes than beluga or narwhal from other collections. The walrus have a larger excess of selenium over mercury and the biological basis for this is unknown.

mercure dans le foie et l'âge du béluga et du morse. Il nous manque les données sur l'âge de certains échantillons dont il est question ici. La variation annuelle des niveaux peut être relativement importante ce qui fait qu'il est difficile, sur le plan statistique, de faire la différence entre la variation et la tendance temporelle.

- Il existe une très étroite relation statistique entre le mercure et le sélénium dans le foie du béluga et du morse (et du narval, selon le rapport 2008). Les pentes des droites de régression des concentrations micromolaires du mercure par rapport au sélénium varient entre les espèces, le morse d'Igloodik ayant des pentes nettement plus basses que le béluga ou le narval des autres échantillons. Le morse a un excès plus important de sélénium par rapport au mercure et la raison biologique de cette différence est inconnue.

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## Objectives

To continue to assess long term trends and to maintain the current data-base on levels of bioaccumulating substances such as mercury, cadmium, selenium and halogenated organic compounds (e.g. PCBs, DDT, toxaphene) in beluga, narwhal and walrus from selected locations in the Canadian Arctic.

## Introduction

The levels of mercury in organs of northern marine mammals generally exceed the two guidelines used to regulate the sale of fish ( $0.5 \text{ ug g}^{-1}$  for sale of commercial fish). There is also a published recommendation that fish consumed in a subsistence fishery not exceed  $0.2 \text{ ug g}^{-1}$  (Health and Welfare Canada, 1979) although this recommendation has no legal status. The extent to which the accumulation of mercury results from natural processes acting on the northern geological settings, or from climate change or from the import of industrial mercury with air and water movements is not clear. Studies of sediment cores suggest that about half the mercury coming into northern lakes is of anthropogenic origin (Lockhart et al., 1998). Studies of mercury in beluga teeth suggest that most of the mercury in contemporary beluga from

the Beaufort Sea has been derived from recent anthropogenic activity (Outridge et al., 2002). However, parallel studies of mercury in teeth of walrus from Igloodik were not elevated over archaeological samples suggesting little anthropogenic mercury in those animals (Outridge et al., 2002). Since previous studies by Outridge et al. (2000) has shown that mercury levels in teeth are correlated with those in liver, kidney, muscle and muktuk, it seems likely that the trends reported in teeth occurred similarly in other organs. Mercury has increased in air over the North Atlantic (Slemr and Langer, 1992) and mercury has been measured in air and in snow in the Arctic (Lu et al., 2001). While there is little doubt that industrial mercury is deposited in the Arctic, the ultimate fate of that mercury is still under investigation. Processes within the snow suggest that much of the mercury deposited to Arctic snow may be volatilized back into the air without actually reaching arctic animals. The question of greatest interest is whether the mercury resulting from human activities is sufficient to cause changes in the levels of mercury in arctic animals. Stern and Macdonald (2005) postulated that the observed increases of mercury in western Arctic beluga since the early 1990's may be attributed to recent changes in ice cover and distribution in the western Arctic Ocean.

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A previous study of mercury in northern animals pooled data from different locations in the eastern and western Arctic and reported a trend to higher levels in both regions (Wagemann et al., 1996). With growing recognition that arctic populations of marine mammals are composed of multiple stocks that are hunted separately in different communities, the need is for trend data on a stock-specific basis. A more recent analysis considering locations separately was provided by Lockhart et al., 2005. In view of the changes required in ages of beluga, the calculations from these earlier studies will have to be repeated.

Whales may range long distances from the communities where they are hunted, but the hunting itself is usually relatively close to the communities. Here, mammals are described only by the communities where the samples were obtained. Hence two or more communities may hunt the same stock if those communities are relatively near each other.

The raw data from several investigations are archived in the Freshwater Institute and comprise records of mercury levels in liver of 1077 arctic beluga, 240 walrus, 390 narwhal and 1011 ringed seals. Biologists obtain samples from hunter kills and those samples form the basis of most analyses. The archive grows through the collection and analyses of new samples and also through the analyses of archived samples from past collections. Over time, the accumulated data offer increasingly the means to detect geographic and temporal trends. Beluga ages in the archived data have been revised in keeping with the new information on growth layer formation.

## Activities in 2008/2009

This report covers data available by early mid-2009. The collection of samples is done independently of this project and is not described here. The analytical methods have been described in previous reports and have been continued to date. While the project is concerned with mercury, many of the same samples have analyzed for selenium and some for methylmercury and cadmium. Liver tissue of most animals was analyzed for mercury with smaller numbers of samples of kidney, muscle and muktuk analyzed.

Additional samples of beluga and walrus were analyzed for total mercury and usually for selenium during 2008/09. Virtually all of the new samples were from collections made in 2008. The new data available are listed in Table 1.

In addition, samples of ringed seals from Holman in 2008 were analyzed but those data are reported elsewhere.

Ages of beluga were determined from teeth on the basis of one growth layer per year following the study by Stewart et al. (2006a). Ages from old collections have been doubled to be consistent with the new calculations. Ages for some beluga are still to be determined. This change did not apply to ages of walrus.

## Results

### *Levels of mercury in liver and kidney of beluga*

The database is a cumulative one with each collection added to earlier data reported to NCP previously. The database currently includes information on 1115 beluga but 38 of these are from the St. Lawrence leaving 1077 from Arctic locations. A complete list of the records available and levels of mercury and selenium were updated in 2008. Additional analyses added in 2008/09 are listed in Table 1. New collections are reported from Pangnirtung, Sanikiluaq, Arviat and Hendrickson Island 2008. Two walrus from Grise Fiord were added also, one from 2007 and one from 2008. Ages for some recent collections of beluga are not available yet; they are being determined and will be added to the database when examinations of teeth have been completed. Lengths were recorded for most beluga and walrus and they are included in Table 1; although they are not adequate for adjusting mercury levels.

### **Pangnirtung**

Four new samples of liver and kidney were obtained from beluga taken at Pangnirtung in 2008 bringing the total for this location to 167 animals in 15 collections over the period from 1982 to 2008. Biological data on the four new whales will be added when they become available. Considering all 15 collections, the mean levels of mercury in liver from beluga from Pangnirtung ranged from 2.2 µg/g (1986) to 16.3 µg/g (2006). Ages, corrected



**Table 1. New data for mercury and selenium in organs of beluga and walrus added to the database during 2008/09**

Species	Location	Year	Organ	Mean Age (yr) and number of samples	Mean Length (cm) and number of samples	Mean Total mercury (ug/g) and number	Std. Dev. Total Hg	Mean selenium (ug/g) and number of samples
Beluga	Pangnirtung	2008	Liver	28.5 (2)		9.79 (4)	5.71	
Beluga	Pangnirtung	2008	Kidney	28.5 (2)		5.12 (4)	3.65	
Beluga	Sanikiluaq	2008	Liver	25 (13)	347.9 (12)	8.48 (13)	5.88	5.90 (13)
Beluga	Sanikiluaq	2008	Kidney	25 (13)	347.9 (13)	2.90 (13)	1.64	3.27 (13)
Beluga	Sanikiluaq	2008	Muscle	25 (13)	347.9 (13)	0.67 (13)	0.21	
Beluga	Arviat	2008	Liver	23.8 (15)	374.7 (15)	11.0 (14)	9.06	5.31 (13)
Beluga	Arviat	2008	Kidney	23.8 (15)	374.7 (15)	3.23 (15)	2.02	2.38 (15)
Beluga	Arviat	2008	Muscle	23.8 (15)	374.7 (15)	0.87 (15)	0.34	
Beluga	Hendrickson I.	2008	Liver		402.0 (28)	22.7 (28)	17.0	8.90 (28)
Beluga	Hendrickson I.	2008	Kidney		400.9 (27)	5.32 (27)	2.80	3.66 (27)
Beluga	Hendrickson I.	2008	Muscle		402.0 (28)	1.14 (28)	0.52	0.34 (28)
Beluga	Hendrickson I.	2008	Muktuk		402.0 (28)	0.37 (28)	0.19	3.55 (28)
Walrus	Hall Beach	2008	Liver		305.5 (4)	3.08 (4)	3.21	
Walrus	Hall Beach	2008	Kidney		305.5 (4)	0.29 (4)	0.09	
Walrus	Igloodik	2008	Liver		300.1 (7)	1.78 (7)	2.95	1.82 (7)
Walrus	Igloodik	2008	Kidney		300.1 (7)	0.30 (7)	0.20	9.89 (7)
Walrus	Grise Fiord	2007	Liver		289.6 (1)	6.37 (1)		3.31 (1)
Walrus	Grise Fiord	2007	Kidney		289.6 (1)	0.85 (1)		4.77 (1)
Walrus	Grise Fiord	2008	Liver		365.8 (1)	6.87 (1)		2.98 (1)
Walrus	Grise Fiord	2008	Kidney		365.8 (1)	1.04 (1)		7.73 (1)

according to Stewart et al. (2006a), are available for 134 of these from 1984 to 2008 and these ranged from 11.1 years (1991, N=10) to 30.6 years (2002, N=10). Age is an important contributor to levels of mercury with higher levels almost always found in older animals. Age data are not yet available for collections in 1982, 2005, 2006 and 2007. The ages for the samples from 1982 are unlikely to become available but the remaining ones will be added when we obtain them. The levels of mercury may be compared superficially among collections from the same location although more rigorous analysis will be undertaken when remaining age data become available. Box plots of mercury in beluga liver from Pangnirtung are shown in Figure 1 with statistical outlier points identified by NCSS shown in the panel at the

right. The line across each box represents the median value for that collection and the box spans the 25<sup>th</sup> to 75<sup>th</sup> percentiles or the interquartile range. The line extending above the box represents points that fall between the 75<sup>th</sup> percentile and the value calculated as the 75<sup>th</sup> percentile plus 1.5 times the interquartile range. Similarly the line extending below the box represent points that fall between the 25<sup>th</sup> percentile and the value calculated as the 25<sup>th</sup> percentile minus 1.5 times the interquartile range. The liver values suggest a trend to higher levels of mercury over the interval from 1982 to 2007 but the samples in 2006, 2007 and 2008 represent only 4 whales each and any suggestion of a temporal trend will have to be confirmed statistically after adjustment for differing ages.

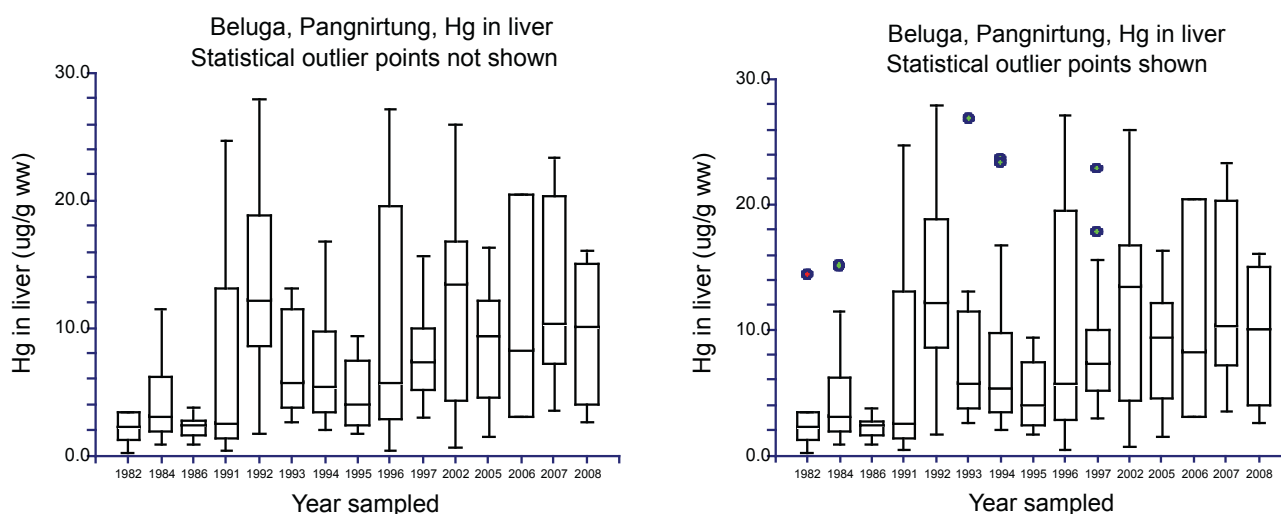


Figure 1. Median values and interquartile ranges of unadjusted levels of total mercury in liver of beluga from Pangnirtung over the interval from 1982 to 2008 comprising 15 collections of 167 individuals with outlier values excluded (left) and shown (right).

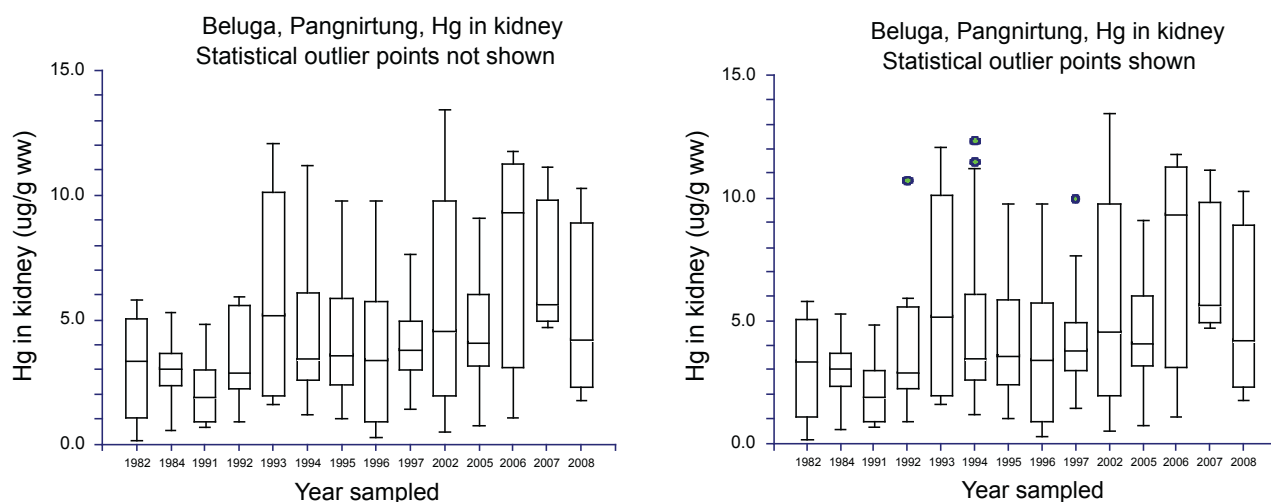


Figure 2. Median values and interquartile ranges of unadjusted levels of total mercury in kidney of beluga from Pangnirtung over the interval from 1982 to 2008 comprising 14 collections of 167 individuals with outlier values excluded (left) and shown (right).

A similar plot for mercury in kidney of 166 beluga from Pangnirtung is shown in Figure 2. Mean levels of mercury in kidney were about half those in liver (Table 1) but again there is a suggestion of a tendency to increase over the interval. As with liver, the last four collections in 2006, 2007 and 2008 consisted of only four individuals and hence merit limited weight.

Mercury levels in muscle of beluga from Pangnirtung are available for only 11 samples

from 1984 with a mean value of  $0.98 \mu\text{g/g}$ . We have no data on mercury in muktuk of beluga from Pangnirtung.

### Sanikiluaq

Fewer beluga data are available from Sanikiluaq and they span a shorter time interval. These data represent 130 individuals in 8 collections from 1994 to 2008. The mean levels of mercury in liver ranged from  $6.1$  to  $27.5 \mu\text{g/g}$ , generally exceeding the means found in beluga from Pangnirtung for

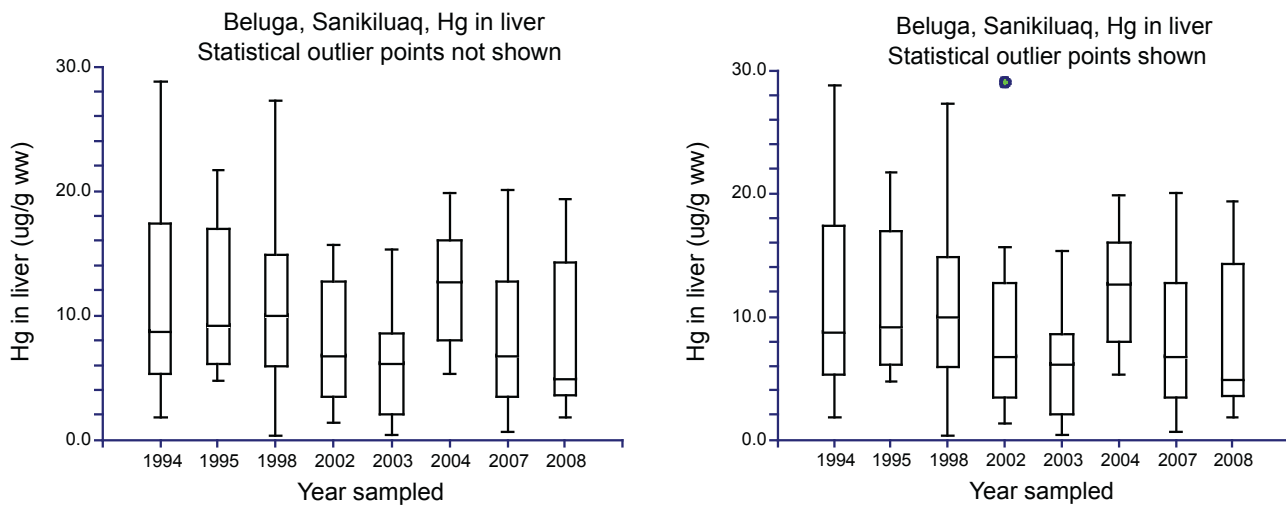


Figure 3. Median values and interquartile ranges of unadjusted levels of mercury in liver of beluga from Sanikiluaq over the interval from 1994 to 2008 comprising 8 collections of 130 individuals with outlier values excluded (left) and shown (right).

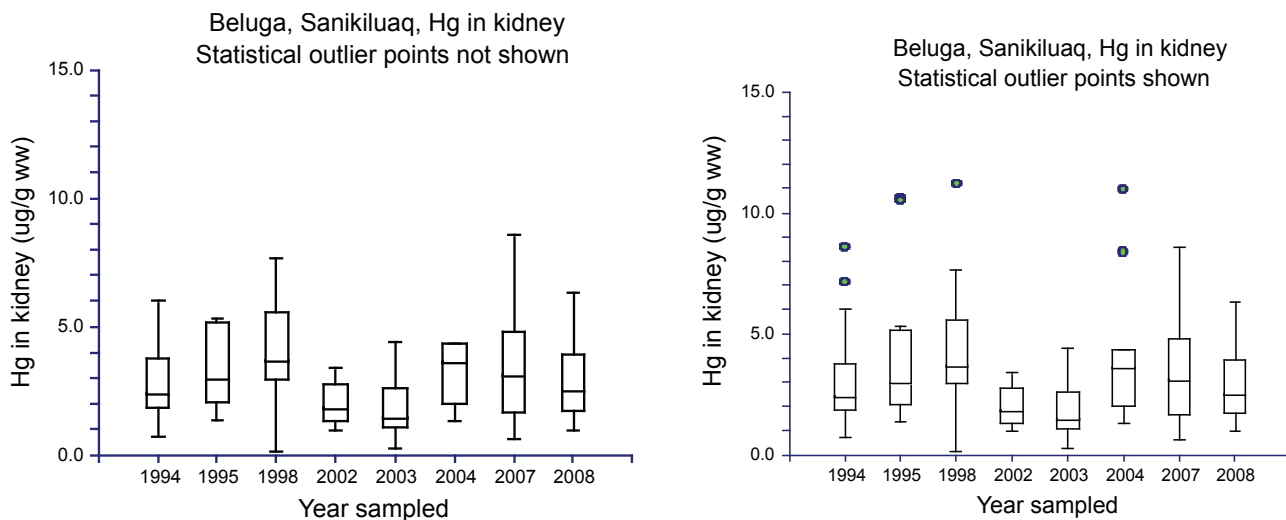


Figure 4. Median values and interquartile ranges of unadjusted levels of mercury in kidney of beluga from Sanikiluaq over the interval from 1994 to 2008 comprising 8 collections of 130 individuals with outlier values excluded (left) and shown (right).

which means ranged from 4.8 to 16.3  $\mu\text{g/g}$  over the same interval. The plots of medians and quartiles in Figure 3, shown for the same range of mercury values as Pangnirtung, offer no indication of a trend to higher values over the interval. The number of animals represented ranged from a low of 10 in 2002 to a high of 30 in 1994.

Similarly with kidney, the levels of mercury were generally lower from with means ranging from 1.8 to 4.1  $\mu\text{g/g}$  but the plot of medians and quartiles

(Figure 4) gives no evidence of any consistent trend to higher or lower levels.

Mercury content was lower yet in muscle of beluga from Sanikiluaq. We have only three collections for the years 1994, 2003 and 2008 with a total of 56 individual whales. Mean levels for the three collections were 0.98, 0.66 and 0.67  $\mu\text{g/g}$  respectively, considerably lower than those from liver or kidney. The plot shown in Figure 5 is too limited to suggest any trend.

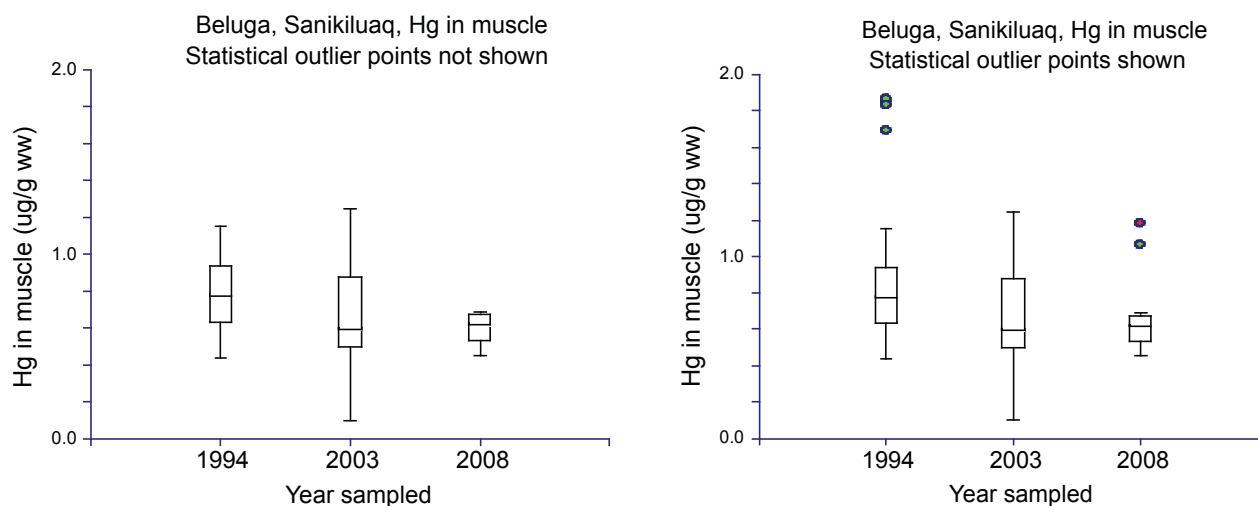


Figure 5. Median values and interquartile ranges of unadjusted levels of mercury in muscle of beluga from Sanikiluaq over the interval from 1994 to 2008 comprising 3 collections of 56 individuals with outlier values excluded (left) and shown (right).

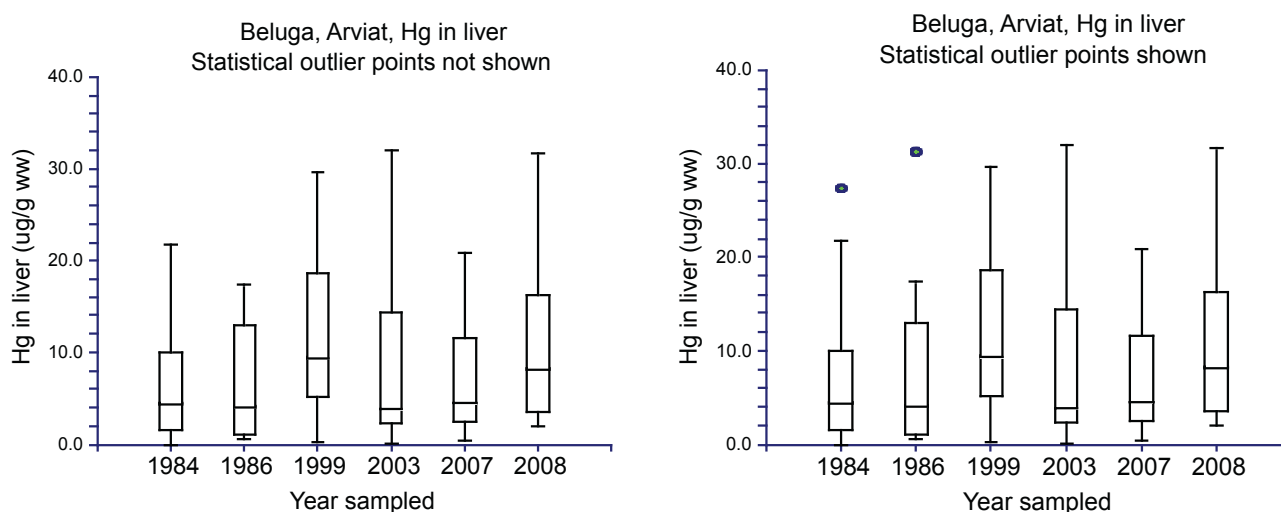


Figure 6. Median values and interquartile ranges of unadjusted levels of mercury in liver of beluga from Arviat over the interval from 1984 to 2008 comprising 6 collections of 136 individuals with outlier values excluded (left) and shown (right).

### Arviat

The database contains records of mercury in liver from 136 beluga in 6 collections from Arviat over the time from 1984 to 2008. Mean levels of mercury ranged from 6.6 to 12.7  $\mu\text{g/g}$  over the 6 collections. Ages are available for all collections and mean ages were very similar, ranging only from 19.6 to 23.8 years. Box plots showing median values and interquartile ranges for mercury in liver are shown in Figure 6. The number of samples available in each collection ranged from 14 to 37.

No obvious consistent trend is evident from inspection of these plots.

As with other locations, levels in kidney were below those in liver. Slightly fewer samples of kidney were available (127) with mean values ranging from 2.4 to 4.2  $\mu\text{g/g}$  over the 6 collections. Box plots are shown in Figure 7 and the medians were slightly higher in the last four collections than in the first two.

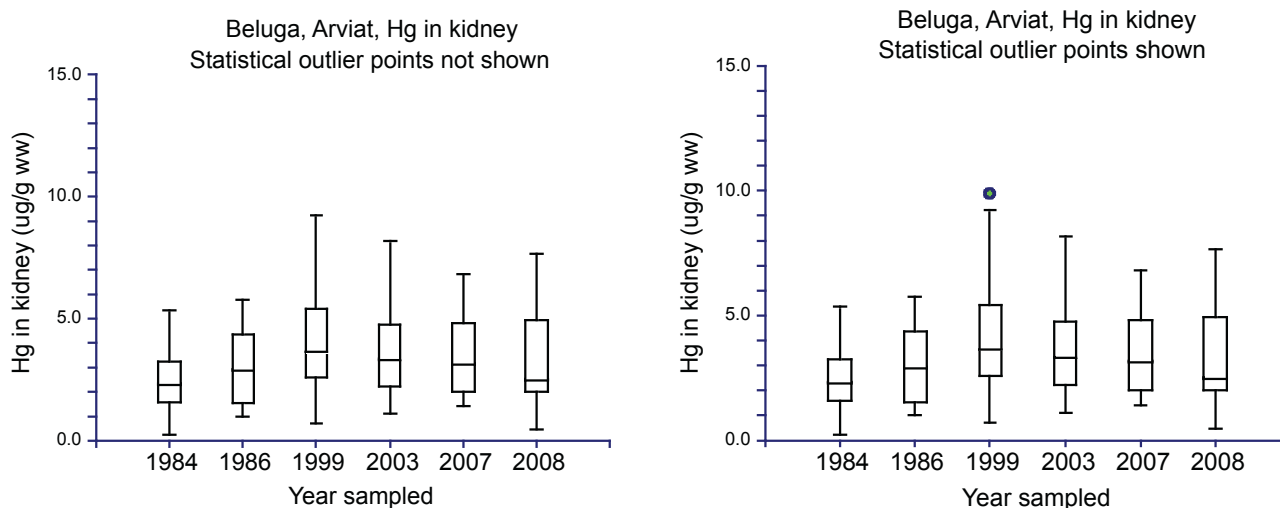


Figure 7. Median values and interquartile ranges of unadjusted levels of mercury in kidney of beluga from Arviat over the interval from 1984 to 2008 comprising 6 collections of 127 individuals with outlier values excluded (left) and shown (right).

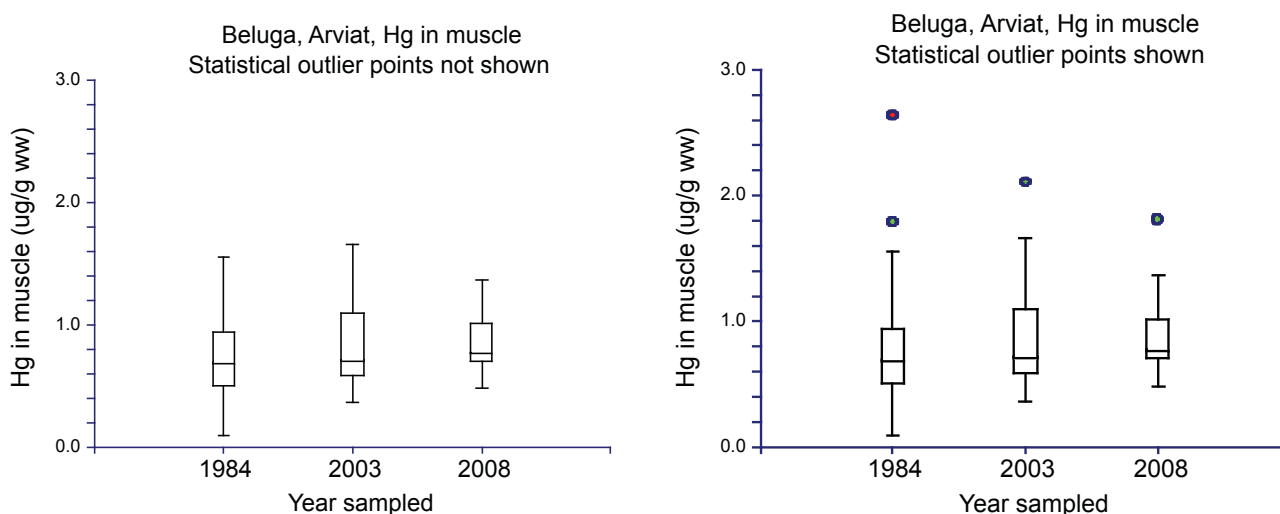


Figure 8. Median values and interquartile ranges of unadjusted levels of mercury in muscle of beluga from Arviat over the interval from 1984 to 2008 comprising 3 collections of 68 individuals with outlier values excluded (left) and shown (right).

The data from Arviat include three sets of analyses on muscle, those for 1984, 2003 and 2008, with mean levels of mercury in muscle of 0.84, 0.88 and 0.87  $\mu\text{g/g}$  respectively. The data describe 68 samples with reasonably good numbers in each collection ( $N=23$ , 30 and 15). As with several sets of data, more rigorous statistical analyses will be required after the ages are available to discern any trend.

#### (Mackenzie Delta) Hendrickson Island

Data from the Mackenzie Delta include collections from locations identified as Hendrickson Island,

East Whitefish, Kendall Island, Bird Camp, Kittigazuit, and Mackenzie Delta with most from Hendrickson Island. These locations have been grouped and considered as “Mackenzie Delta” and they include 357 individuals collected from 1977 to 2008 in 15 collections. (The earliest departmental data we have are samples taken in 1981. The collection from 1977 is cited from a report by Beak Consultants, 1978, and is included because of its historical importance.) The levels of mercury in liver from this region were consistently higher than those from the other locations described above. The mean levels for the collections ranged

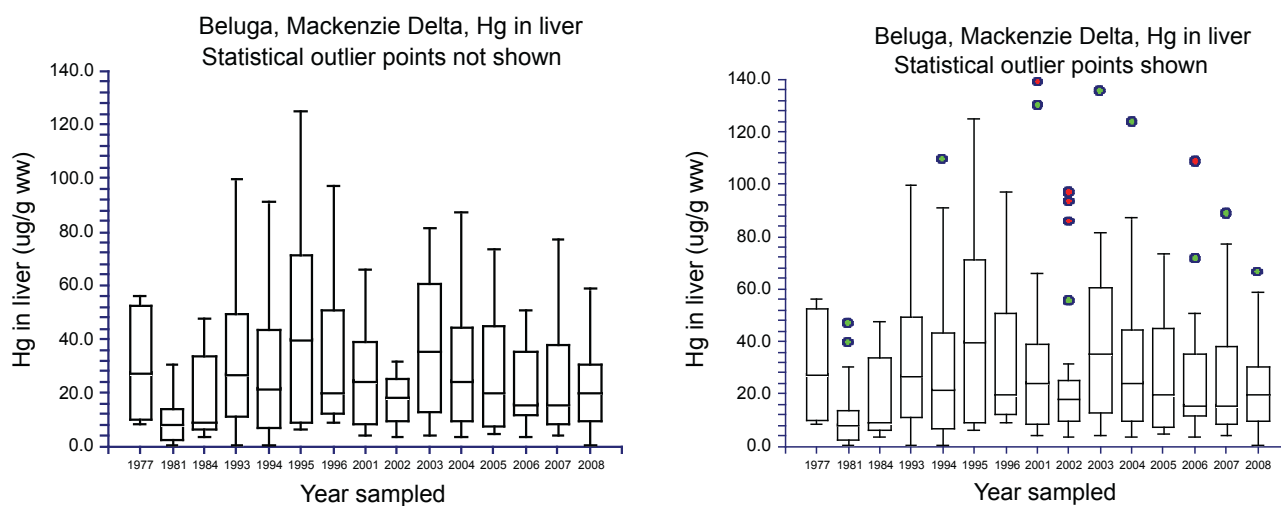


Figure 9. Median values and interquartile ranges of unadjusted levels of mercury in liver of beluga from the Mackenzie Delta over the interval from 1977 to 2008 comprising 14 collections of 329 individuals with outlier values excluded (left) and shown (right).

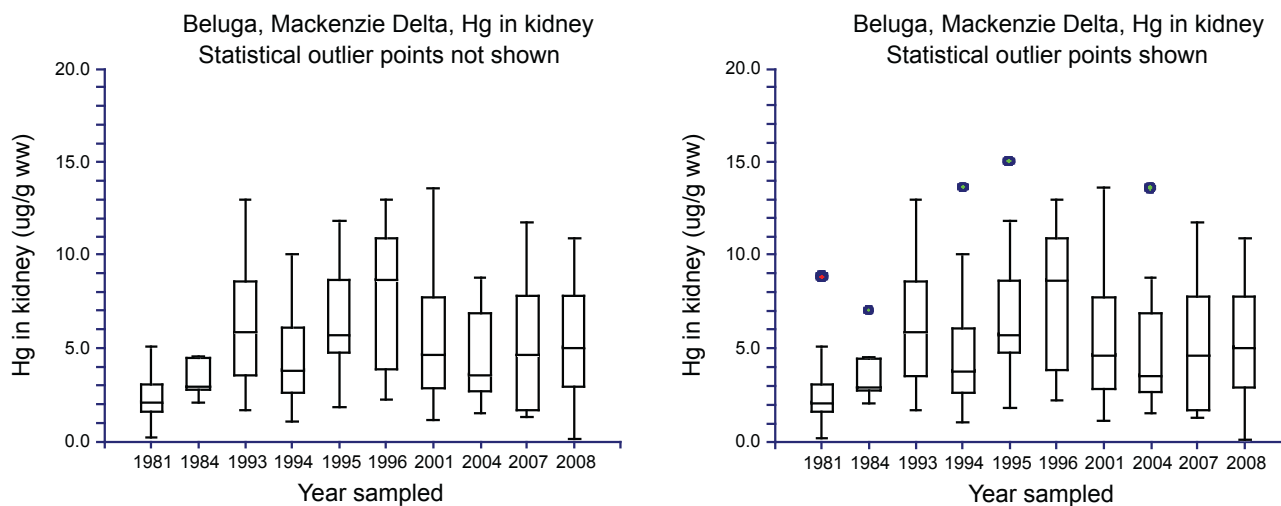


Figure 10. Median values and interquartile ranges of unadjusted levels of mercury in kidney of beluga from the Mackenzie Delta over the interval from 1981 to 2008 comprising 10 collections of 241 individuals with outlier values excluded (left) and shown (right).

from 10.7  $\mu\text{g/g}$  (1981) to 40.1  $\mu\text{g/g}$  (2003). There was variation in ages with mean ages ranging from 24.5 years in 1981 to 45.1 years in 1993; ages for the collections in 2007 and 2008 are not available yet. Inspection of the box plots in Figure 9 gives no indication of a consistent, linear trend over the period but rather suggests the possibility of alternating increases and decreases.

We have fewer analyses of liver (241) from the Mackenzie Delta collections but they still span the period from 1981 to 2008 in 10 collections.

Again, as with liver, there is no consistent linear trend obvious.

We have data on levels of mercury in muscle from almost all the beluga from the Mackenzie Delta (351). Mercury in muscle of these whales was relatively high with mean values for all 15 collections exceeding 1  $\mu\text{g/g}$ . Means ranged from 1.03  $\mu\text{g/g}$  in 1984 to 2.12  $\mu\text{g/g}$  in the 1977 collection cited from the report by Beak Consultants (1978). If the 1977 collection is ignored, the next highest mean mercury concentration was 1.75  $\mu\text{g/g}$  in 1996.



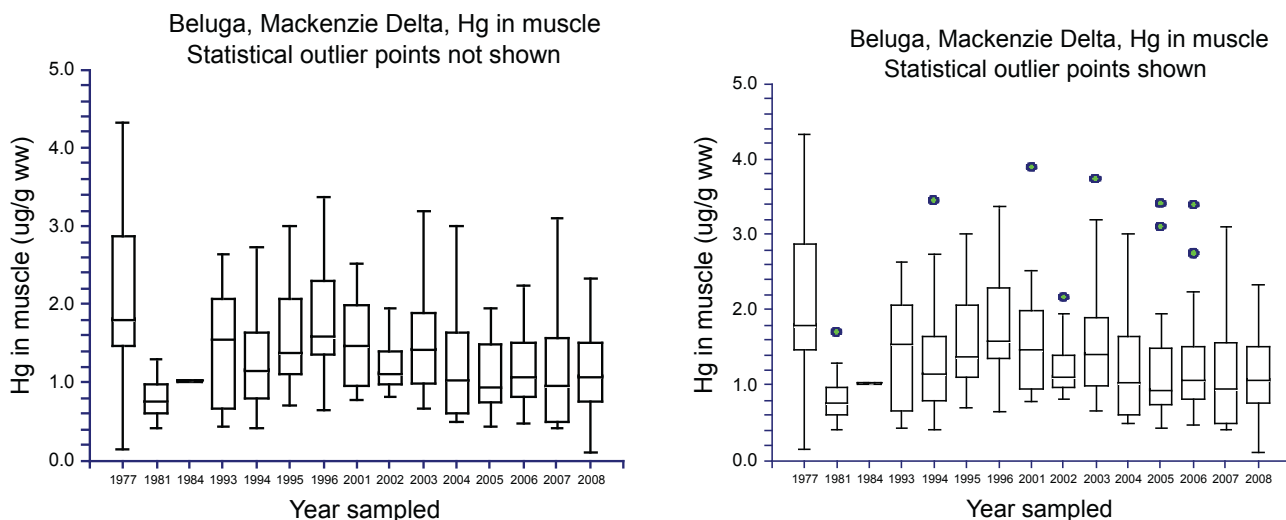


Figure 11. Median values and interquartile ranges of unadjusted levels of mercury in muscle of beluga from the Mackenzie Delta over the interval from 1977 to 2008 comprising 15 collections of 351 individuals with outlier values excluded (left) and shown (right). (The muscle sample in 1984 consisted of one individual.)

### *Levels of mercury in liver and kidney of walrus* Hall Beach

Four new samples of walrus were added in 2008 with a high mean mercury value of  $3.08 \mu\text{g/g}$  in liver. Mean levels of mercury in liver were relatively high in both 2007 ( $4.85 \mu\text{g/g}$ ) and 2008 when compared with earlier collections from this community ( $1.3\text{--}1.7 \mu\text{g/g}$ ). We do not have ages yet for the collections in 2007 and 2008 leaving open the possibility that the high values in those years are artifacts of sampling some older individuals.

Samples in 2007 and 2008 had a few individuals with very high values which affected the calculation of the means. The high degree of variation in these samples is evident from the long interquartile ranges (Figure 12). The possibility of any temporal trend in liver mercury will await determination of remaining ages. There was no similar upward spike in the values for mercury in kidney in 2007 and 2008. The mean value in kidney in 2007 was  $0.345 \mu\text{g/g}$  while that in 2008 was  $0.287 \mu\text{g/g}$ . These values may be compared with

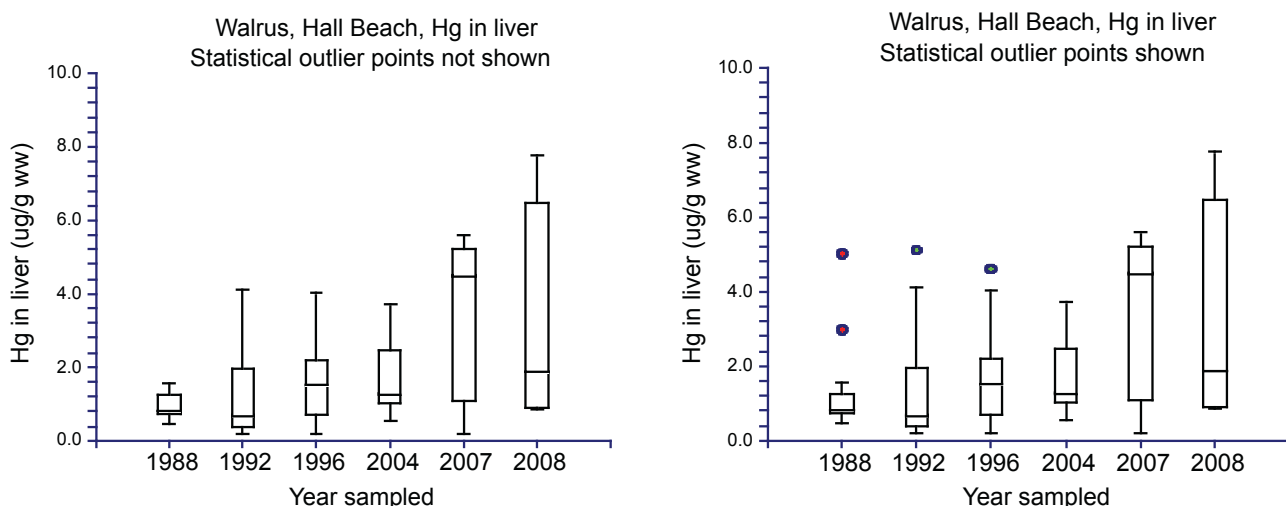


Figure 12. Median values and interquartile ranges of unadjusted levels of mercury in liver of walrus from Hall Beach over the interval from 1988 to 2008 comprising 6 collections of 73 individuals with outlier values excluded (left) and shown (right).

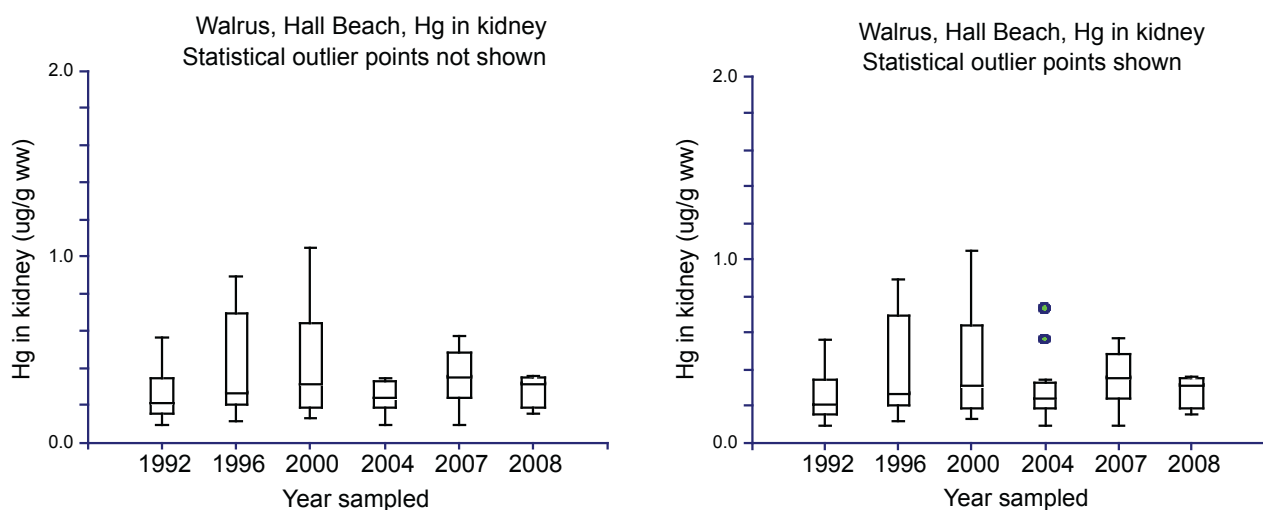


Figure 13. Median values and interquartile ranges of unadjusted levels of mercury in kidney of walrus from Hall Beach over the interval from 1999 to 2008 comprising 6 collections of 66 individuals with outlier values excluded (left) and shown (right).

the four previous collections for which mean values ranged from 0.26 to 0.44  $\mu\text{g/g}$ . The box plots in Figure 13 offer little evidence of any temporal trend of mercury in kidney.

### Igloodik

Seven new samples of walrus liver and kidney were added to the collections from Igloodik in 2008 bringing the total to 105 samples for liver and 61 for kidney. Ages for the walrus in 2008 are not available yet. The mean level of mercury in liver in 2008 was 1.78  $\mu\text{g/g}$  and that in kidney was 0.3  $\mu\text{g/g}$ . The mean mercury in liver in 2008 fell

within the range of means reported from previous 7 collections (0.78-2.41  $\mu\text{g/g}$ , 1982-1996) as did that for kidney for the four earlier collections (0.29-0.47  $\mu\text{g/g}$ , 1983-1996). Box plots of levels in liver are shown in Figure 14 and a number of outlier points were identified. No consistent trend to lower or higher values is evident from these plots.

We have fewer samples from kidney but they still span the period from 1983 to 2008 in 5 collections. The box plots for mercury in kidney are shown in Figure 15 and again no consistent linear trend is obvious.

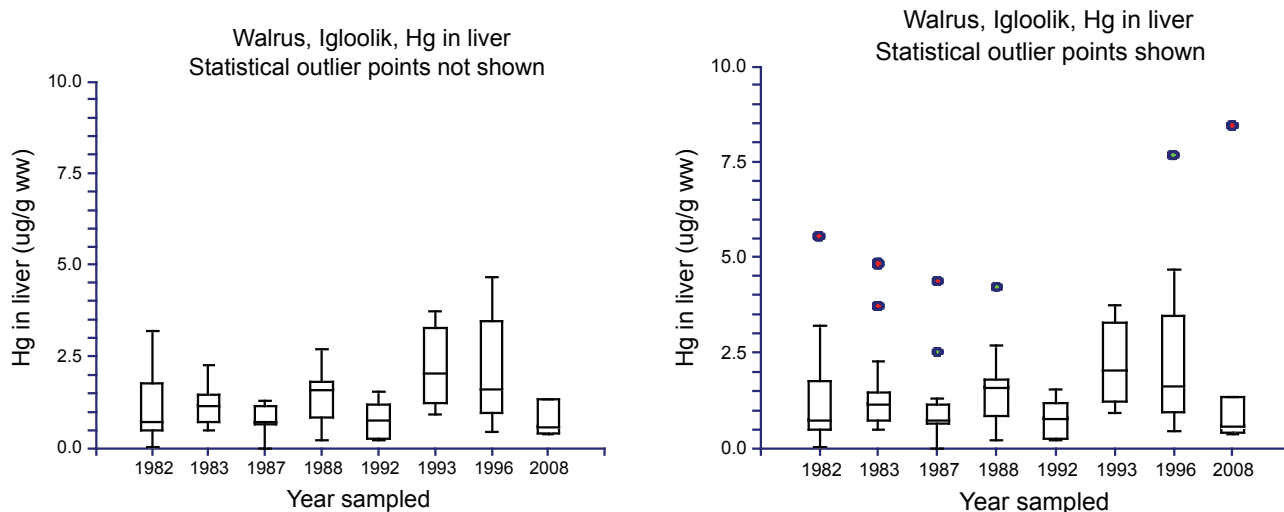


Figure 14. Median values and interquartile ranges of unadjusted levels of mercury in liver of walrus from Igloodik over the interval from 1982 to 2008 comprising 8 collections of 105 individuals with outlier values excluded (left) and shown (right).

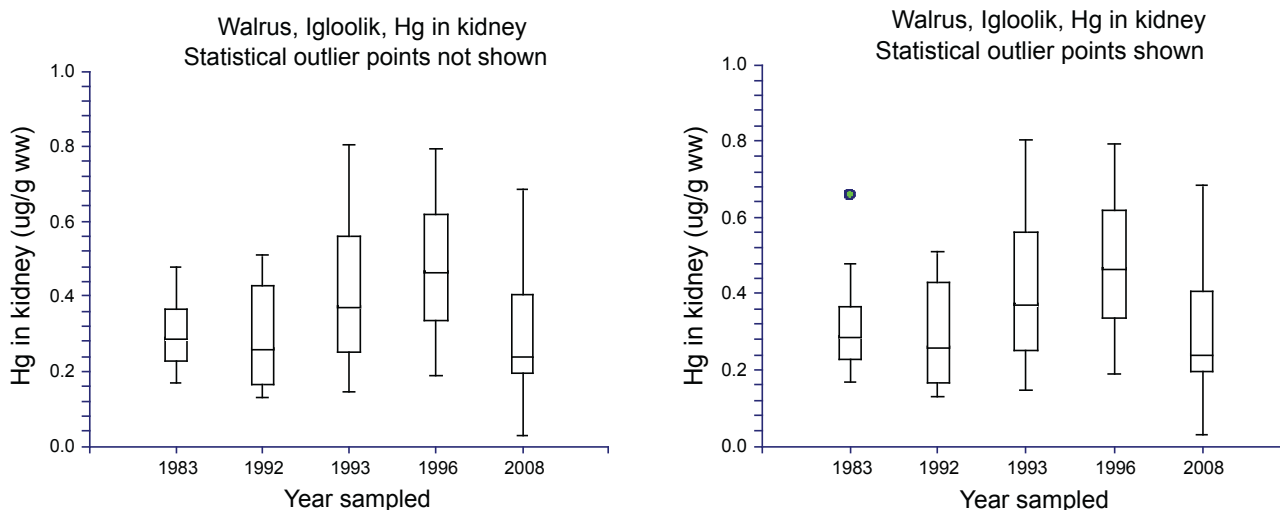


Figure 15. Median values and interquartile ranges of unadjusted levels of mercury in kidney of walrus from Igloolik over the interval from 1983 to 2008 comprising 5 collections of 105 individuals with outlier values excluded (left) and shown (right).

### Grise Fiord

We obtained two samples of walrus liver and kidney from Grise Fiord, one taken in 2007 and the other in 2008. We have had beluga and narwhal from this community on previous occasions but these are the first walrus. The concentrations of mercury and selenium in liver and kidney are shown in Table 1. While the new data represent only one walrus in each year, mercury levels in both liver samples were relatively high and additional samples from this community are desirable.

### Mercury and selenium

There is usually a strong statistical correlation between levels of mercury and levels of selenium in organs of arctic marine mammals. (See report for 2008 for the Hg/Se relationship in narwhal.) The micromolar concentrations of mercury and selenium in liver of 125 beluga from Pangnirtung for which we have both Hg and Se in the same sample are shown in Figure 16 (left). The robust regression technique of NCSS software discarded 5 samples as outliers but indicated a very strong relationship between Hg and Se. The slope of the regression was 0.75 indicating an excess of selenium over mercury and the R-squared value for the relationship was 0.88. Although the levels were lower, very similar results were obtained with the 116 samples of kidney available from the same location and interval. The robust regression slope was 0.73 with an R-squared value of 0.86.

The relationship between mercury and selenium in 104 samples of liver of walrus from Igloolik taken over the interval from 1982 to 2008 was less striking. Although the relationship still exists, the slope was lower than in the beluga shown in Figure 16. The values for walrus liver are shown in Figure 17 (left) and those for kidney in Figure 17 (right). The robust regression technique yielded a slope of only 0.08 and an R-squared value of only 0.21 for liver. We have fewer samples of kidney from walrus from Igloolik but the relationship between mercury and selenium was improved somewhat (R-squared = 0.52 from the robust regression) but the slope was even shallower (0.004). The data are shown in Figure 17 (right).

Walrus appear to differ somewhat from beluga or narwhal in the way they accumulate mercury and selenium. Walrus have very low ratios of mercury to selenium in kidney. The biological basis for and its implications for the animals, if any, are unknown but may offer a fruitful area for future study.

### Expected Project Completion Date

Mercury at the levels in these mammals remains a concern with regard to human dietary intakes. It also poses legitimate questions regarding effects on the mammals themselves. In view of the physical changes occurring in the Arctic, it seems likely

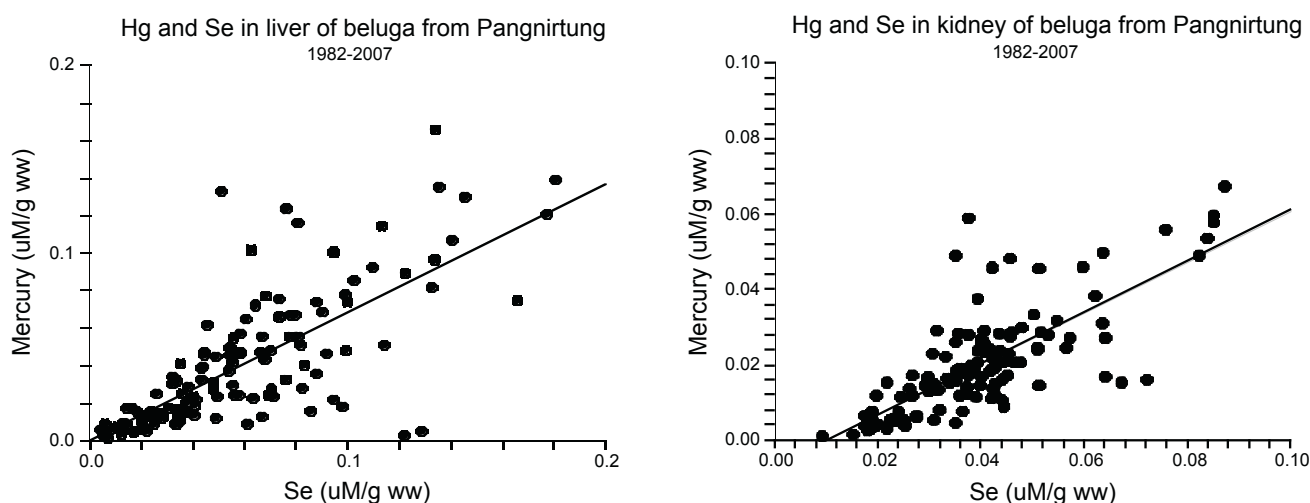


Figure 16. Micromolar concentrations of selenium and mercury in 125 samples of liver (left) and 116 samples of kidney (right) of beluga from Pangnirtung over the period 1983-2007 (left).

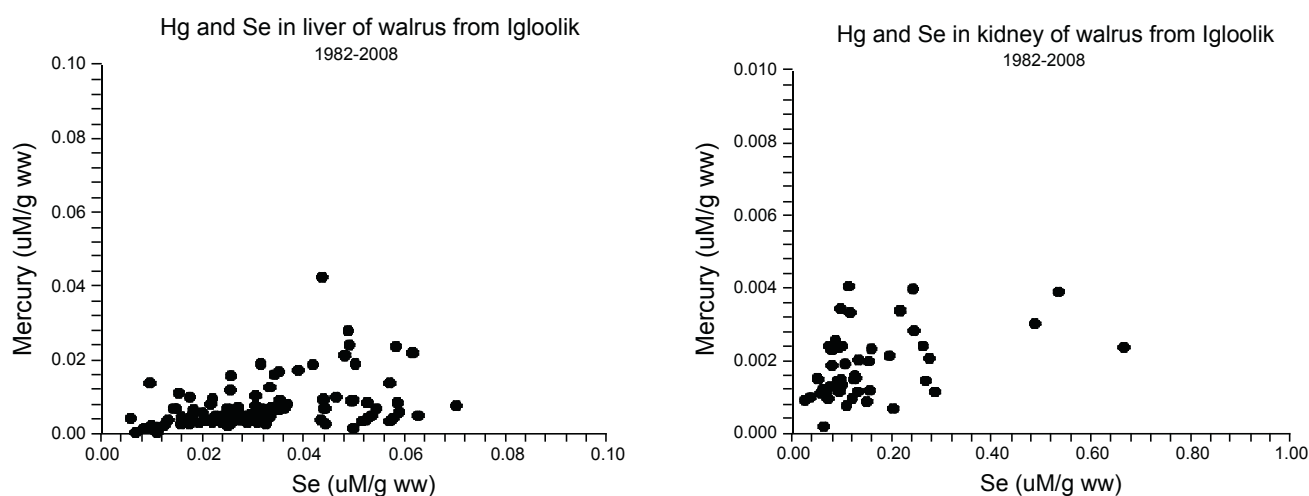


Figure 17. Micromolar concentrations of selenium and mercury in 104 samples of liver (left) and 51 samples of kidney (right) of walrus from Igloodik over the period 1982-2007 for liver and 1983-2008 for kidney. (Note that the vertical scale was compressed and the horizontal scale expanded to accommodate some low values for mercury and high values for selenium in kidney.)

that some form of this research will have to be continued for as long as marine mammals are hunted for human consumption.

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# Temporal trends of halogenated organic compounds in Canadian Arctic Beluga

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## Abstract

The objectives of this on going study are to maintain current data on contaminant levels in marine mammals and to continue to assess the temporal trends of halogenated organic compounds (HOCs). This will allow us to determine whether the levels of these compounds in the marine mammals, and hence exposure to Arctic people who traditionally consume them, are changing with time. These results will also help to test the effectiveness of international controls and, in conjunction with projects such as CFL (IPY & NSERC) and ArcticNet, to understand the effects that climate variation may have on these contaminant levels.

## Key Messages

- HCHs levels in the western Arctic beluga are not showing the declines observed atmospherically and in the Arctic Ocean since the ban in the usage of the technical mixture by China in 1983 and followed by India in 1990. Declines, however, were observed in the Panguitong animals beginning in the 2000s.

## Résumé

Cette étude permanente a pour but de tenir à jour les données sur les niveaux de contaminants dans les mammifères marins et de poursuivre l'évaluation des tendances temporelles des composés organiques halogénés (COH). Cela nous permettra de déterminer si les niveaux de ces composés chez les mammifères marins et, par conséquent, l'exposition des habitants de l'Arctique qui, traditionnellement, les consomment, changent avec le temps. Ces résultats permettront aussi de vérifier l'efficacité des contrôles internationaux et, de concert avec des projets comme l'étude sur le chenal de séparation circumpolaire (API et CRSNG) et ArcticNet, de comprendre les effets que les variations climatiques peuvent avoir sur ces niveaux de contaminants.

## Messages clés

- Les niveaux de HCH dans le béluga de l'Ouest de l'Arctique ne reflètent pas les baisses observées dans l'atmosphère et dans l'océan Arctique depuis l'interdiction des mélanges techniques par la Chine en 1983, puis par l'Inde en 1990. Des baisses ont toutefois été observées chez les animaux de Panguitong depuis les années 2000.



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- Eastern Baffin Island narwhal (Pond Inlet and Clyde River), with a few exceptions, seem to have the highest HOC level of the locations sampled in 2005/06.
  - In walrus, highest concentrations in the males and female animals were measured in the 2008 Grise Fiord and Hall Beach animals, respectively.
  - Le narval de l'Est de l'île de Baffin (Pond Inlet et Clyde River), à quelques exceptions près, semble avoir le plus haut niveau de COH de tous les sites étudiés en 2005-2006.
  - Chez le morse, les concentrations les plus fortes, chez le mâle et la femelle, ont été mesurées en 2008 chez les animaux du fjord Grise et de Hall Beach respectivement.
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## Objectives

To continue to assess long term trends and to maintain the current data-base on levels of halogenated organic compounds (e.g. PCBs, DDT, toxaphene) in marine mammals (beluga, narwhal, walrus) from selected locations across the Canadian Arctic.

## Introduction

Marine animals accumulate (relatively) high concentrations of halogenated organic compounds (HOCs). The objectives of this project, therefore, are to maintain current data on contaminant levels in marine mammals and to continue to assess the temporal trends of halogenated organic compounds (HOCs). This will allow us to determine whether contaminant levels in the marine mammals, and hence exposure to Arctic people who traditionally consume them, are changing with time. These results will also help to test the effectiveness of international controls and, in conjunction with projects such as the IPY CFL (Circumpolar Flaw Lead) System Study and ArcticNet Phase I and II to understand the effects that climate variation may have on the contaminant levels in these animals and the health of the stocks.

The raw data and samples from previous and ongoing investigations are archived in the Freshwater Institute and represent about 2000 marine mammals, mostly beluga, ringed seals, narwhal and walrus from 23 different locations across the Canadian Arctic. DFO scientists concerned with stock management, animal health and climate change studies obtain various samples from hunter kills and those samples form the basis of most of our analyses. For example, tissues from

eastern Arctic and Hudson Bay narwhal and beluga have been collected and analyzed for HOCs as part of DFOs stock management studies since 1996. In the western Arctic the collections have been supported by FJMC since 2002. The accumulating data resulting from these studies offer the means to detect both spatial and temporal trends of HOCs and heavy metals in Arctic marine mammals and most importantly to try and link the observed variation to physical and biological process and carbon and contaminants cycling within the Arctic Ocean.

## Activities in 2008/2009

Samples collected and analysed to date are in Table 1-4. Our quest to collect and analyse walrus samples has improved significantly this year as we had moderate responses from both Hall Beach and Iglood. We also received tissues from two Grise Fiord walrus. Difficulties in getting aging results for walrus and beluga have now been addressed but we are still in a catch up stage. We expect to have all the aging completed by fall, 2009.

## Results

As part of an ongoing whale sampling and stock identity program, supported by the Nunavut Wildlife Management Board (NWMB), FJMC, NIF and DFO, samples were collected by hunters during their subsistence hunts using standardized whale kits. Blubber, kidney, liver, ovaries and uterus, muscle and the lower jaw, as well as morphometric data were collected for each animal. All samples were shipped frozen to the Freshwater Institute and stored at - 20°C until analysis.

We now have a very unique long term data set for HOCs in western Arctic beluga; twelve time points

spanning nineteen years. As shown in Table 1, no real trends are observed. Of particular interest is the fact that HCHs are not showing the declines observed atmospherically and in the Arctic Ocean (Li et al., 2004) since the ban in the usage of the technical mixture by China in 1983 and followed by India in 1990. As was postulated for mercury, the lack of response of HCH in western Arctic beluga

to the declining levels in the Arctic atmosphere and Ocean could be, at least in part, attributed to recent changes in ice cover, which may, for example, alter the foraging of the beluga whales or their prey (Stern and Macdonald, 2005; Gaden et al. 2009; Loseto et al. 2006; 2008a,b; 2009). Conversely, HCH concentrations start to decline in the Pangnirtung animals in the early 2000s. These

**Table 1. Mean (stdev) of major HOC groups and compounds in blubber from western Arctic beluga (ng g<sup>-1</sup>, wet wt).**

Loc	Year	Sex	n	Age	%lipid	ΣCBz	ΣHCH	ΣCHL	ΣDDT	ΣPCB	ΣCHB	Dieldrin	Oxychlor
HL	1989	M	12	29.0 (15.0)	90.5 (3.7)	421.5 (185.9)	269.3 (111.3)	1857.6 (667.8)	2930.8 (1125.8)	3800.0 (1587.7)	4548.0 (1462.6)	297.5 (112.1)	487.7 (143.6)
HI	1994	M	10	33.4 (10.8)	93.3 (2.8)	669.6 (168.7)	215.8 (55.5)	1601.4 (495.5)	4283.8 (2591.7)	3949.1 (1819.5)	11207.4 (6735.6)	324.2 (118.4)	477.0 (109.9)
HI	1995	M	15	32.1 (10.4)	81.3 (3.3)	783.2 (380.4)	211.9 (40.6)	2076.2 (1025.8)	3907.7 (1753.7)	4176.6 (1473.3)	6701.0 (3207.3)	287.9 (141.7)	519.1 (348.2)
HI	1996	M	10	27.5 (6.8)	92.4 (4.6)	666.3 (117.5)	243.0 (23.3)	1803.1 (274.0)	4415.3 (2030.9)	4246.8 (1146.9)	10519.3 (4708.5)	397.7 (133.4)	498.3 (45.0)
HI	2001	M	18	32.3 (9.7)	82.2 (3.5)	518.7 (169.4)	205.5 (44.0)	1932.4 (808.5)	3445.1 (1633.7)	3986.7 (1737.5)	6266.4 (3634.6)	296.9 (103.9)	499.0 (225.6)
HI	2002	M	9	30.4 (8.1)	90.9 (2.2)	421.1 (109.1)	234.3 (34.8)	1408.9 (351.3)	2480.3 (1361.5)	2745.7 (1033.7)	7024.5 (3324.2)	376.0 (113.4)	439.9 (126.1)
HI	2003	M	9	27.2 (9.3)	91.3 (3.9)	518.3 (113.2)	243.1 (50.1)	1556.5 (361.5)	2736.0 (2134.3)	3084.4 (1462.9)	6188.5 (2600.1)	338.4 (100.7)	519.7 (119.3)
HI	2004	M	10	25.3 (6.0)	85.1 (6.8)	888.0 (189.0)	329.2 (50.8)	2932.8 (535.7)	3747.0 (1928.3)	4894.1 (948.8)	7554.1 (2360.1)	500.7 (99.4)	744.8 (192.5)
HI	2005	M	10	26.8 (9.6)	92.4 (3.4)	623.5 (196.3)	269.3 (68.2)	2568.5 (1715.6)	4545.8 (3595.3)	3633.1 (1906.5)	5680.1 (3776.0)	447.0 (186.8)	991.7 (684.7)
HI	2006	M	14	23.6 (7.2)	90.8 (6.2)	567.6 (197.7)	229.8 (33.6)	1498.8 (416.7)	2017.0 (851.8)	3100.2 (1081.7)	3673.6 (1614.5)	289.0 (80.4)	518.23 (162.2)
HI	2007	M	10	nd	86.0 (9.9)	650.1 (251.8)	254.7 (31.9)	2236.1 (853.1)	3805.4 (2124.4)	3744.1 (1391.8)	7226.1 (3276.8)	359.3 (114.0)	698.4 (281.2)
HI	2008	M	10	nd	80.1 (5.9)	392.4 (90.5)	121.5 (23.5)	1412.5 (582.7)	1348.7 (698.8)	3006.6 (1247.6)	4585.5 (2707.9)	209.2 (68.6)	290.5 (99.5)

HL = Husky Lakes; HI = Hendrickson Island; nd = not yet determined; ΣDDT = Sum of *p,p'*-DDT, *p,p'*-DDE, *p,p'*-DDD, *o,p'*-DDT, *o,p'*-DDE and *o,p'*-DDD; ΣHCH = α- β- and γ-HCH isomers; ΣCHL = all chlordane related compounds, including heptachlor; ΣCBz = Sum of 1245TCB, 1234TCB, P5CBz, HCBz; ΣPCB = Sum of CB1, 3, 4/10, 7, 6, 8/5, 19, 18, 17, 24/27, 16/32, 26, 25, 31, 28, 33, 22, 45, 46, 52, 49, 47, 48, 44, 42, 41/71, 64, 40, 74, 70/76, 66, 95, 56/60, 91, 84/89, 101, 99, 83, 97, 87, 85, 136, 110, 82, 151, 144/135, 149, 118, 134, 114,131, 146, 153, 132, 105, 141, 130/176, 179, 137, 138, 158, 178/129, 175, 187, 183, 128, 185, 174, 177, 171, 156, 201/ 157, 172/197, 180, 193, 191, 200, 170, 190, 198, 199, 196/203, 189, 208, 195, 207, 194, 205, 206, 209

**Table 2a. Mean (stdev) of major HOC groups and compounds in blubber from male Pangnirtung beluga (ng g<sup>-1</sup>, wet wt).**

Loc	Year	Sex	n	Age	%lipid	ΣCBz	ΣHCH	ΣCHL	ΣDDT	ΣPCB	ΣCHB	Dieldrin	Oxychlor
PG	1982	M	9	8.7 (5.8)	90.5 (2.4)	438.7 (79.0)	251.6 (57.5)	1614.2 (363.9)	5132.1 (2466.9)	4140.9 (1303.9)	9451.6 (2318.6)	604.7 (106.1)	271.7 (59.0)
PG	1986	M	16	5.8 (1.6)	90.9 (3.5)	376.8 (99.1)	259.4 (72.7)	1405.6 (318.2)	3233.8 (1051.4)	2569.8 (638.6)	7536.3 (1852.3)	449.4 (101.8)	290.5 (128.1)
PG	1991	M	3	8.7 (6.6)	94.7 (3.5)	549.2 (105.1)	348.6 (245.6)	2084.2 (836.1)	5387.5 (2074.2)	3368.0 (620.2)	15169.9 (9407.1)	1132.9 (937.1)	736.5 (280.9)
PG	1992	M	15	10.8 (5.6)	91.0 (2.2)	399.2 (203.6)	211.0 (77.7)	2067.0 (469.3)	5386.6 (1666.3)	3972.6 (986.4)	9801.8 (2522.5)	450.6 (170.6)	411.3 (124.0)
PG	1995	M	5	14.1 (8.6)	92.3 (3.3)	810.3 (550.0)	322.6 (205.5)	2628.9 (844.1)	4562.9 (3745.8)	4229.5 (1497.0)	16793.6 (8807.8)	1110.5 (565.3)	914.1 (338.6)
PG	1996	M	6	17.0 (5.5)	88.8 (2.9)	566.2 (188.2)	219.6 (57.6)	1685.3 (582.0)	6138.1 (798.6)	4783.1 (992.9)	12641.6 (3041.4)	666.2 (170.3)	194.9 (47.8)
PG	1997	M	9	12.1 (4.1)	91.2 (2.4)	789.2 (149.5)	233.6 (45.4)	1551.2 (772.2)	5290.5 (1879.9)	4015.6 (719.7)	9414.4 (3271.7)	1551.2 (772.2)	291.8 (180.8)
PG	2002	M	6	17.4 (3.8)	89.8 (2.4)	572.9 (220.1)	174.6 (66.6)	956.6 (439.1)	4072.3 (1203.9)	3772.9 (791.3)	6271.6 (1948.6)	359.8 (113.2)	145.4 (83.18)
PG	2005	M	7	nd	93.7 (2.0)	128.3 (83.9)	77.7 (25.7)	1145.3 (767.1)	2334.6 (1730.1)	1780.7 (1109.6)	3835.5 (1929.6)	295.9 (198.2)	357.5 (253.2)
PG	2006	M	3	nd	88.5 (1.9)	333.3 (159.5)	112.0 (33.4)	1422.3 (592.5)	1709.5 (849.5)	2368.7 (861.1)	4870.2 (1610.1)	420.6 (166.2)	431.7 (180.9)
PG	2007	M	1	nd	94.6	683.2	210.1	3129.2	4325.7	3772.4	15113.3	908.3	922.5
PG	2008	M	4	nd	74.1 (5.8)	154.7 (104.7)	55.3 (36.4)	1253.2 (749.9)	1265.9 (883.1)	2691.8 (1698.2)	6604.3 (5532.6)	224.7 (146.2)	253.5 (172.9)

na = not yet determined; ΣDDT = Sum of *p,p'*-DDT, *p,p'*-DDE, *p,p'*-DDD, *o,p'*-DDT, *o,p'*-DDE and *o,p'*-DDD; ΣHCH = α- β- and γ-HCH isomers; ΣCHL = all chlordane related compounds, including heptachlor; ΣCBz = Sum of 1245TCB, 1234TCB, P5CBz, HCBz; ΣPCB = Sum of CB1, 3, 4/10, 7, 6, 8/5, 19, 18, 17, 24/27, 16/32, 26, 25, 31, 28, 33, 22, 45, 46, 52, 49, 47, 48, 44, 42, 41/71, 64, 40, 74, 70/76, 66, 95, 56/60, 91, 84/89, 101, 99, 83, 97, 87, 85, 136, 110, 82, 151, 144/135, 149, 118, 134, 114, 131, 146, 153, 132, 105, 141, 130/176, 179, 137, 138, 158, 178/129, 175, 187, 183, 128, 185, 174, 177, 171, 156, 201/ 157, 172/197, 180, 193, 191, 200, 170, 190, 198, 199, 196/203, 189, 208, 195, 207, 194, 205, 206, 209; nd = not yet determined

**Table 2b. Mean (stdev) of major HOC groups and compounds in blubber from female Pangnirtung beluga (ng g-1, wet wt).**

Loc	Year	Sex	n	Age	%lipid	ΣCBz	ΣHCH	ΣCHL	ΣDDT	ΣPCB	ΣCHB	Dieldrin	Oxychlor
PG	1986	F	4	3.5	92.9 (0.6)	404.8 (127.8)	292.7 (72.0)	1454.3 (572.6)	3238.7 (1522.3)	2783.0 (818.7)	8230.1 (1353.6)	514.0 (232.3)	335.3 (164.6)
PG	1991	F	7	4.2	94.5 (3.5)	1375.0 (1065.5)	420.4 (189.6)	2956.4 (1401.6)	7611.0 (5961.2)	4271.4 (2107.6)	20274.1 (8170.6)	1454.4 (762.1)	1034.3 (531.7)
PG	1992	F	8	13.1	91.6 (2.8)	246.8 (173.4)	173.5 (58.9)	1252.1 (493.0)	2482.1 (913.8)	2391.9 (847.5)	5784.8 (2409.6)	281.0 (102.6)	238.2 (111.4)
PG	1995	F	5	7.8	89.8 (7.9)	801.8 (415.2)	325.1 (93.0)	2473.3 (793.7)	3157.3 (889.3)	3265.3 (745.9)	15755.3 (9116.7)	1149.9 (413.7)	961.4 (391.4)
PG	1996	F	10	14.1	87.5 (7.2)	251.0 (209.4)	113.5 (65.0)	904.7 (655.6)	2521.2 (1589.1)	2047.8 (1433.1)	5324.7 (3786.7)	298.0 (231.0)	101.7 (56.6)
PG	2005	F	2	nd	92.1 (1.4)	150.0 (9.6)	78.6 (1.0)	919.7 (24.5)	1954.0 (208.6)	1882.5 (24.4)	3527.7 (309.0)	293.9 (44.2)	282.8 (21.0)
PG	2006	F	1	nd	82.4	334.5	150.7	886.4	747.2	1348.3	2553.9	340.8	226.2
PG	2007	F	3	nd	97.9 (3.2)	73.5 (45.4)	70.3 (43.4)	665.2 (424.4)	736.5 (545.3)	985.1 (484.2)	3369.9 (1558.6)	158.3 (96.9)	135.9 (110.6)

na = not yet determined; ΣDDT = Sum of *p,p'*-DDT, *p,p'*-DDE, *p,p'*-DDD, *o,p'*-DDT, *o,p'*-DDE and *o,p'*-DDD; ΣHCH = α- β- and γ-HCH isomers; ΣCHL = all chlordanes related compounds, including heptachlor; ΣCBz = Sum of 1245TCB, 1234TCB, P5CBz, HCBz; ΣPCB = Sum of CB1, 3, 4/10, 7, 6, 8/5, 19, 18, 17, 24/27, 16/32, 26, 25, 31, 28, 33, 22, 45, 46, 52, 49, 47, 48, 44, 42, 41/71, 64, 40, 74, 70/76, 66, 95, 56/60, 91, 84/89, 101, 99, 83, 97, 87, 85, 136, 110, 82, 151, 144/135, 149, 118, 134, 114, 131, 146, 153, 132, 105, 141, 130/176, 179, 137, 138, 158, 178/129, 175, 187, 183, 128, 185, 174, 177, 171, 156, 201/ 157, 172/197, 180, 193, 191, 200, 170, 190, 198, 199, 196/203, 189, 208, 195, 207, 194, 205, 206, 209; nd = not yet determined

data, however; have not yet been corrected for age. As part of the IPY CFL project and ArcticNet Phase II a study is currently under way to try and determine the effects that climate change will have on the ocean-sea ice-atmosphere coupling of hexachlorocyclohexane (HCH) and transport within the western Arctic.

Data for narwhal and walrus are shown in Tables 3 and 4, respectively. Baffin Island narwhal (Pond Inlet and Clyde River), with a few exceptions, seem to have the highest HOC level of the locations sampled in 2005/06. In walrus, highest concentrations in the males and female animals were measured in the 2008 Grise Fiord and Hall Beach animals, respectively.

**Table 3. Mean (stdev) of major HOC groups and compounds in blubber from Narwhal (ng g<sup>-1</sup>, wet wt).**

Loc	Species	Year	Sex	n	Length	%lipid	ΣCBz	ΣHCH	ΣCHL	ΣDDT	ΣPCB	ΣCHB	Dieldrin	Oxychlor
PI	Narwhal	2006	M	5	410 (60)	93.9 (2.1)	698.0 (145.6)	185.2 (67.0)	2978.3 (901.8)	5784.9 (2829.2)	5255.3 (2159.6)	14867.6 (4857.8)	558.8 (99.5)	632.2 (187.5)
PI	Narwhal	2006	F	6	335 (35)	90.3 (3.4)	562.5 (333.9)	163.2 (96.4)	2395.1 (1511.5)	4044.8 (3246.3)	3748.8 (2687.5)	12818.0 (6541.4)	509.7 (268.3)	520.4 (363.7)
IG	Narwhal	2006	M	5	452 (21)	83.6 (11.0)	370.2 (99.1)	163.9 (45.4)	2178.0 (457.9)	4696.0 (995.6)	3679.7 (751.6)	10858.4 (1847.2)	424.5 (96.2)	445.1 (88.9)
IG	Narwhal	2006	F	3	399 (27)	91.5 (1.0)	352.1 (109.9)	142.0 (43.2)	1582.6 (744.7)	2209.4 (1127.7)	2210.5 (1026.2)	8081.4 (3643.2)	352.6 (123.6)	320.2 (168.9)
RE	Narwhal	2006	M	3	380 (72)	94.9 (4.1)	570.0 (194.7)	172.6 (29.4)	1672.2 (412.8)	3069.9 (479.1)	2302.9 (214.9)	8423.8 (5646.4)	509.2 (165.6)	440.4 (12.4)
RE	Narwhal	2006	F	4	363 (57)	92.6 (4.0)	320.6 (306.1)	119.3 (87.2)	1564.7 (1247.7)	2655.9 (2129.3)	2387.1 (1294.1)	8913.5 (6684.2)	323.8 (273.8)	368.0 (373.2)
RB	Narwhal	2006	M	5	429 (46)	89.0 (6.4)	341.3 (55.7)	174.5 (34.6)	2278.7 (883.8)	3480.7 (1157.6)	2933.6 (1085.1)	9458.6 (2552.0)	510.9 (112.5)	537.0 (252.8)
CR	Narwhal	2005	M	5	352 (40)	89.2 (93.3)	487.3 (213.3)	148.3 (20.0)	2382.1 (1275.6)	5848.7 (3673.8)	4358.2 (2276.0)	14875.9 (5467.1)	444.4 (169.5)	584.8 (268.2)
CR	Narwhal	2005	F	4	391 (75)	87.9 (4.6)	347.9 (234.0)	173.2 (124.6)	2046.6 (1698.9)	4652.9 (3896.3)	4184.4 (3634.5)	13187.2 (12218.9)	391.1 (235.6)	448.9 (336.1)

<sup>1</sup> Sexes of these animals need to be determined genetically as this information in many cases was not provided by the hunters; PI = Pond Inlet; RE = Resolute; IG = Iglulik; CR = Clyde River; RB = Repulse Bay; ΣDDT = Sum of *p,p'*-DDT, *p,p'*-DDE, *p,p'*-DDD, *o,p'*-DDT, *o,p'*-DDE and *o,p'*-DDD; ΣHCH = α- β- and γ-HCH isomers; ΣCHL = all chlordane related compounds, including heptachlor; ΣCBz = Sum of 1245TCB, 1234TCB, P5CBz, HCBz; ΣPCB = Sum of CB1, 3, 4/10, 7, 6, 8/5, 19, 18, 17, 24/27, 16/32, 26, 25, 31, 28, 33, 22, 45, 46, 52, 49, 47, 48, 44, 42, 41/71, 64, 40, 74, 70/76, 66, 95, 56/60, 91, 84/89, 101, 99, 83, 97, 87, 85, 136, 110, 82, 151, 144/135, 149, 118, 134, 114, 131, 146, 153, 132, 105, 141, 130/176, 179, 137, 138, 158, 178/129, 175, 187, 183, 128, 185, 174, 177, 171, 156, 201/ 157, 172/197, 180, 193, 191, 200, 170, 190, 198, 199, 196/203, 189, 208, 195, 207, 194, 205, 206, 209

Issues with aging of the beluga and walrus samples have now been resolved but results not expected until fall, 2009. Adjustment using ANCOVA will be undertaken when aging is complete.

## Expected Project Completion Data

This study, in conjunction with the trace metal work, is expected to be on going.

## References

De March. B, G.A. Stern. 2003. S. Stock separation of narwhal (*Monodon monoceros*) in Canada based on organochlorine contaminants. CSAS (Canadian Scientific Advisory Secretariat) Research Document 2003/079.

**Table 4. Mean (stdev) of major HOC groups and compounds in blubber from walrus (ng g<sup>-1</sup>, wet wt).**

Loc	Species	Year	Sex	n	Length	%lipid	ΣCBz	ΣHCH	ΣCHL	ΣDDT	ΣPCB	ΣCHB	Dieldrin	Oxychlor
HB	Walrus	2007	M	4	350 (55)	79.2 (5.5)	2.5 (0.6)	122.8 (71.1)	220.2 (107.5)	47.5 (21.0)	428.5 (155.8)	427.6 (234.3)	220.9 (142.2)	103.5 (51.0)
HB	Walrus	2007	F	3	219 (110)	78.7 (14.5)	1.8 (0.1)	48.5 (26.6)	26.6 (3.9)	14.2 (4.9)	130.6 (53.2)	87.3 (21.6)	52.4 (36.3)	7.2 (2.6)
HB	Walrus	2008	M	1	305	80.7	2.9	42.0	136.1	17.5	177.0	415.7	154.4	551.0
HB	Walrus	2008	F	3	306 (23)	68.6 (8.7)	11.3 (6.2)	55.6 (14.1)	122.9 (15.1)	14.6 (4.6)	156.6 (34.8)	347.6 (125.2)	175.8 (63.7)	67.4 (20.9)
GF	Walrus	2008	M	2	328 (54)	73.7 (1.9)	14.5 (2.1)	87.0 (1.1)	461.6 (69.0)	101.4 (32.8)	1307.0 (166.5)	591.4 (169.5)	284.0 (6.8)	326.6 (85.6)
IG	Walrus	2008	M	4	316 (18)	70.1 (4.7)	9.6 (5.9)	67.2 (9.9)	215.4 (86.6)	17.0 (7.7)	249.1 (85.0)	522.1 (211.6)	217.9 (57.8)	155.1 (74.5)
IG	Walrus	2008	F	3	279 (8)	73.9 (5.3)	12.7 (0.6)	22.5 (5.9)	37.4 (12.9)	12.6 (7.2)	102.9 (24.5)	259.1 (100.0)	39.5 (10.8)	12.2 (5.8)

<sup>1</sup> Sexes of these animals need to be determined genetically as this information in many cases was not provided by the hunters; IG = Iglulik; HB = Hall Beach; GF = Grise Fiord. ΣDDT = Sum of *p,p'*-DDT, *p,p'*-DDE, *p,p'*-DDD, *o,p'*-DDT, *o,p'*-DDE and *o,p'*-DDD; ΣHCH = α- β- and γ-HCH isomers; ΣCHL = all chlordane related compounds, including heptachlor; ΣCBz = Sum of 1245TCB, 1234TCB, P5CBz, HCBz; ΣPCB = Sum of CB1, 3, 4/10, 7, 6, 8/5, 19, 18, 17, 24/27, 16/32, 26, 25, 31, 28, 33, 22, 45, 46, 52, 49, 47, 48, 44, 42, 41/71, 64, 40, 74, 70/76, 66, 95, 56/60, 91, 84/89, 101, 99, 83, 97, 87, 85, 136, 110, 82, 151, 144/135, 149, 118, 134, 114, 131, 146, 153, 132, 105, 141, 130/176, 179, 137, 138, 158, 178/129, 175, 187, 183, 128, 185, 174, 177, 171, 156, 201/ 157, 172/197, 180, 193, 191, 200, 170, 190, 198, 199, 196/203, 189, 208, 195, 207, 194, 205, 206, 209

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# Temporal Trends of Contaminants in Arctic Seabird Eggs: Inter-year Variability

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## **Abstract**

Contaminants have been monitored in arctic seabird eggs collected from Prince Leopold Island in the Canadian High Arctic since 1975. In order to examine annual variation in the temporal trend data series, the fourth of five annual egg collections was made in 2008 from each of two species of seabirds from Prince Leopold Island. All of the eggs were analyzed for the legacy organochlorines (OCs) and mercury (Hg) as well as brominated flame retardants (BFRs) and perfluorinated compounds (PFCs). Annual sampling has shown that there can be considerable inter-annual variation in residue levels which would be missed in the regular sampling regime of every five years. As 2008 was a major sampling year, eggs from five seabird species were sampled from Prince Leopold Island and analyzed for OCs, BFRs, PFCs and Hg. There were significant differences in egg Hg concentrations among the five seabird species sampled with black-legged kittiwakes having the lowest Hg concentrations and glaucous gulls, the highest.

## **Résumé**

Les contaminants sont contrôlés dans les œufs d'oiseaux marins arctiques recueillis sur l'île Prince Leopold, dans l'Arctique canadien, depuis 1975. Afin d'étudier la variation annuelle des séries de données sur les tendances temporelles, les quatrième et cinquième collectes annuelles ont été faites en 2008 pour chacune des deux espèces d'oiseaux marins de l'île Prince Leopold. Tous les œufs ont été analysés quant aux anciens composés organochlorés (OC) et au mercure (Hg), ainsi qu'aux ignifugeants bromés (BFR) et aux composés perfluorés. Les échantillons annuels indiquent une variation considérable interannuelle des niveaux des résidus, qui pourrait passer inaperçue dans un échantillonnage ordinaire aux cinq ans. Étant donné que 2008 était une année d'échantillonnage importante, les œufs de cinq espèces d'oiseaux marins ont été recueillis sur l'île Prince Leopold et analysés quant aux OC, BFR, PFC et au Hg. D'importantes différences ont été observées dans les concentrations de Hg dans les œufs des cinq espèces d'oiseaux marins, ceux de la mouette tridactyle ayant les concentrations de Hg les plus faibles et ceux du goéland bourgmestre, les concentrations les plus élevées.

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## Key Messages

- Total mercury (Hg) concentrations in eggs of three arctic seabird species have increased significantly between 1975 and 2008.
- Annual monitoring of the thick-billed murre and northern fulmar eggs has shown that there can be considerable inter-annual variation in residue levels which would be missed in the regular sampling regime of every five years.
- There are significant differences in egg Hg concentrations among the five seabird species sampled at Prince Leopold Island in 2008. Eggs of black-legged kittiwakes had the lowest Hg concentrations and glaucous gulls, the highest.

## Messages clés

- Les concentrations de mercure total (Hg) dans les œufs de trois espèces d'oiseaux marins arctiques ont augmenté de façon importante entre 1975 et 2008.
- Le contrôle annuel des œufs du guillemot de Brünnich et du fulmar boréal révèle qu'il peut y avoir une variation considérable interannuelle des niveaux des résidus, qui pourrait passer inaperçue dans un échantillonnage ordinaire aux cinq ans.
- D'importantes différences ont été observées dans les concentrations de Hg dans les œufs des cinq espèces d'oiseaux marins de l'île Prince Leopold étudiés en 2008. Les œufs de la mouette tridactyle ont les concentrations de Hg les plus faibles et ceux du goéland bourgmestre, les concentrations les plus élevées.

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## Objectives

- In order to examine annual variation in the temporal trend data series, eggs are to be collected for contaminant analyses from each of two species of seabirds (northern fulmar, thick-billed murre) from Prince Leopold Island annually for five years starting in 2005. For comparative purposes, we are also making annual collections of thick-billed murre eggs from Coats Island in northern Hudson Bay (our Low Arctic monitoring colony since 1993) in parallel with the High Arctic collections.
- In 2008/09, which was a major monitoring year in the five-year cycle of the core seabird egg monitoring program, eggs were collected from each of five species of seabirds (northern fulmar, thick-billed murre, black-legged kittiwake, black guillemot, glaucous gull) from Prince Leopold Island as well as from thick-billed murres from Coats Island for contaminant analyses. These collections incorporate the collections for the two species being sampled annually. It was proposed that this opportunity be used to carry out an inter-species comparison of the newer groups of compounds (i.e. brominated flame retardants,

perfluorinated compounds) among the five species of seabirds being sampled.

## Introduction

Eggs of thick-billed murres (*Uria lomvia*), northern fulmars (*Fulmarus glacialis*) and black-legged kittiwakes (*Rissa tridactyla*) from Prince Leopold Island in the Canadian High Arctic have been monitored for contaminants since 1975 (Braune 2007) to provide an index of contamination of the arctic marine ecosystem and possible implications for seabird health. Past collections of arctic seabird eggs for contaminant analyses have been opportunistic but collections have been standardized to every five years since 1988. Most of the legacy persistent organic pollutants or POPs (e.g. PCBs, DDT) have been declining whereas total mercury (Hg) has been increasing (Braune 2007), as have the perfluorinated carboxylic acids (PFCAs) and, until recently, the polybrominated diphenyl ethers (PBDEs) (Braune 2008).

Interpretation of temporal trend data is often obscured by the “noise” associated with concentration measurements. The probability that a monitoring program will detect a temporal trend in concentrations, in spite of the “noise” in the data,

represents its statistical power. Long contaminant monitoring time series show random inter-year variations which are not part of a trend, which demonstrates the risks in using small, scattered sets of data on occasionally collected samples for interpreting environmental issues (Bignert et al. 1993, 1994, 1998; Olsson 1995, Hebert and Weseloh 2003). In order to examine the inter-year variation in contaminants data, and to improve the statistical power of the temporal trend data series for Canadian Arctic seabirds, we have been collecting eggs from each of two species of seabirds (northern fulmar, thick-billed murre) from Prince Leopold Island annually for five years starting in 2005. For comparative purposes, we have also been making annual collections of thick-billed murre eggs from Coats Island in northern Hudson Bay (our Low Arctic monitoring colony since 1993) in parallel with the High Arctic collections. Eggs are being analyzed for the normal suite of legacy POPs and total Hg, and the murre and fulmar eggs from Prince Leopold Island are being analyzed for PBDEs, hexabromocyclododecane (HBCD), polychlorinated dibenzo-*p*-dioxins (PCDDs), dibenzofurans (PCDFs), coplanar PCBs, and perfluorinated compounds (PFCs), as well.

However, we lack recent information for newer contaminants such as brominated flame retardants (BFRs) and PFCs in species of seabirds from the Canadian Arctic other than thick-billed murres and northern fulmars. Analyses of these compounds were not included as part of the Northwest Polynya food web project (see Buckman et al. 2004), nor have these compounds been analyzed in recent samples of other seabird species from Prince Leopold Island. As part of the core seabird egg monitoring program, we collected eggs from each of five species of seabirds (northern fulmar, thick-billed murre, black-legged kittiwake, black guillemot, glaucous gull) from Prince Leopold Island in 2008. This provided an opportunity to carry out a comparison of these new groups of compounds (BFRs, PFCs) among five seabird species which occupy quite different trophic levels (see Buckman et al. 2004).

## Activities in 2008/2009

### *Sample collection/analysis:*

Eggs were successfully collected from each of five species of seabirds (northern fulmar, thick-billed

murre, black-legged kittiwake, black guillemot, glaucous gull) from Prince Leopold Island (74°02'N, 90°05'W) in Lancaster Sound as well as from thick-billed murres on Coats Island (62°30'N, 83°00'W) in northern Hudson Bay for analyses of the legacy POPs, PBDEs, HBCD, PFCs, total Hg and stable isotopes. Additionally, the murre and fulmar eggs from Prince Leopold Island have been submitted for analyses of PCDDs, PCDFs and coplanar PCBs. Eggs are analyzed for the normal suite of legacy POPs (e.g. PCBs, DDT, chlordanes, chlorobenzenes, etc.), PBDEs, HBCD, and PFCs in pools of 3 eggs each (15 eggs per collection = 5 pools of 3 eggs each). Murre and fulmar eggs from Prince Leopold Island are also analyzed for PCDDs, PCDFs and coplanar PCBs in pools of 5 eggs each (15 eggs per collection = 3 pools of 5 eggs each) to conform with previous analyses. All eggs are individually analyzed for total Hg and stable isotopes of nitrogen (<sup>15</sup>N/<sup>14</sup>N) and carbon (<sup>13</sup>C/<sup>12</sup>C).

### *Analytical methods:*

Analyses of the legacy POPs, PBDEs, HBCD, PFCs and total Hg are carried out at the National Wildlife Research Centre (NWRC) laboratories at Carleton University in Ottawa, Ontario. The legacy POPs are analyzed by gas chromatography using a mass selective detector (GC/MSD) according to NWRC Method No. MET-CHEM-OC-06A. PBDEs and HBCD are analyzed using GC/MSD run in negative ion chemical ionization (NCI) mode. Total Hg is analyzed using an Advanced Mercury Analyzer (AMA-254) equipped with an ASS-254 autosampler for solid samples according to NWRC Method No. MET-CHEM-AA-03E. The method employs direct combustion of the sample in an oxygen-rich atmosphere. PCDDs, PCDFs and coplanar PCBs were analyzed by the Research and Productivity Council (RPC) in Fredericton, NB, which identified and quantified the compounds by high resolution gas chromatography coupled to a High Resolution Mass Spectrometer (HRGC/HRMS) using internal and external standards. The method is based on EPA Method 1613B in which specific congeners are targeted. Comparability with previous results generated by NWRC was assessed by analysis of two commercial Certified Reference Materials. PFCs are analyzed using HPLC/MS/MS in negative electrospray mode (ESI<sup>-</sup>) according to NWRC Method No. MET-WTD-PFC-01. Quality assurance/quality control (QA/QC) was monitored

by NWRC Laboratory Services which is an accredited laboratory through the CAEAL and has participated in the NCP's QA/QC Program. Stable isotope (C, N) analyses are done at the Department of Soil Science, University of Saskatchewan, Saskatoon. All samples are archived in the Environment Canada Specimen Bank housed at the National Wildlife Research Centre in Ottawa.

## Results

Analyses of the legacy organochlorines, PCDDs, PCDFs, or non-*ortho* PCBs, PBDEs, HBCD, and total Hg have been completed but we have received only the Hg data since the other data are still undergoing quality assurance review. We have also not yet received results for the PFCs and stable isotopes.

Although the Hg data have not yet been corrected for possible variations in trophic position over time as indicated by stable nitrogen ratios, trends of total Hg continue to show statistically significant increases for concentrations in eggs for thick-billed murres ( $n=50$ ,  $r=0.79$ ,  $p<0.00001$ ), northern fulmars ( $n=51$ ,  $r=0.45$ ,  $p<0.001$ ) and black-legged kittiwakes ( $n=26$ ,  $r=0.58$ ,  $p<0.003$ ) between 1975 and 2008 (Figure 1). The increase in Hg levels seems to be continuing, particularly in thick-billed murres whereas, after 2003, concentrations in the

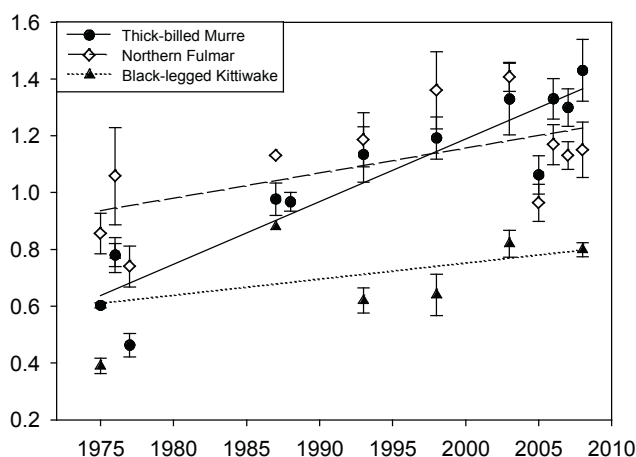


Figure 1. Mean concentrations ( $\pm$  standard error) of total Hg ( $\mu\text{g g}^{-1}$  dry wt) in eggs of northern fulmars, thick-billed murres and black-legged kittiwakes collected from Prince Leopold Island, 1975-2008.

fulmars, and possibly the kittiwakes, appear to be levelling off.

There were statistically significant differences in egg Hg levels among species in 2008 (ANOVA:  $F_{(4,15)} = 27.1$ ,  $p<0.0001$ ) with the black-legged kittiwakes having the lowest Hg concentrations and the glaucous gulls with the highest concentrations (Figure 2). The relative Hg concentration pattern among species in 2008 was the same as it was in 1993 (Figure 2) except that the differences

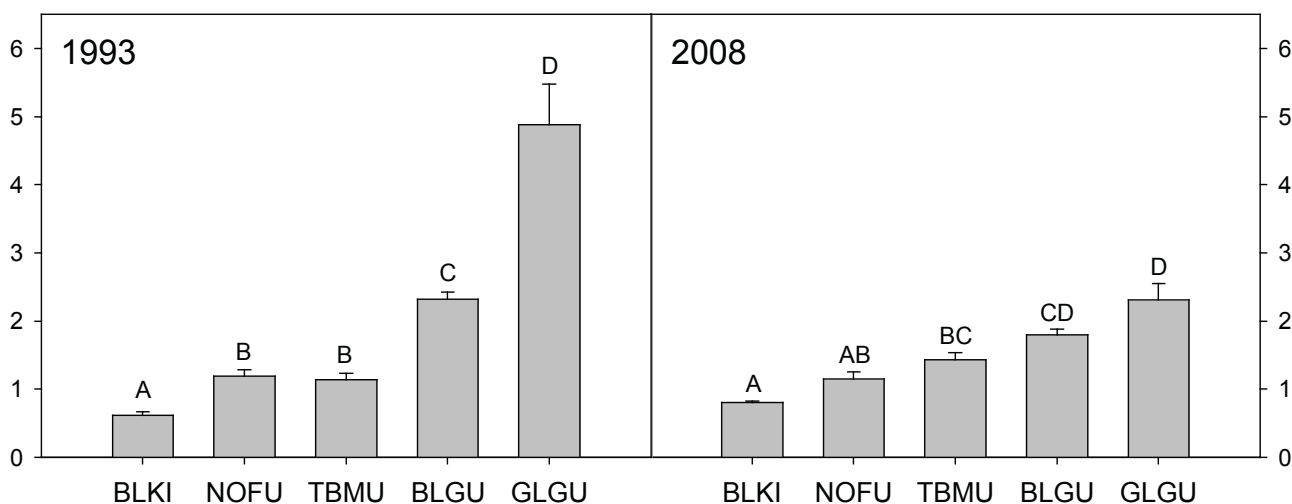


Figure 2. Mean concentrations ( $\pm$  standard error) of total Hg ( $\mu\text{g g}^{-1}$  dry wt) in eggs of black-legged kittiwakes (BLKI), northern fulmars (NOFU), thick-billed murres (TBMU), black guillemots (BLGU) and glaucous gulls (GLGU) collected from Prince Leopold Island in 1993 and 2008. For each year, different letters indicate statistically different Hg concentrations among species.

among species was more pronounced in 1993 with all species but the murres and fulmars having statistically significant difference Hg levels from each other (ANOVA:  $F_{(4,19)}=68.0$ ,  $p<0.00001$ ). Unlike the kittiwake and murre egg Hg levels, which increased from 1993 to 2008, Hg levels in the black guillemot and glaucous gull eggs significantly decreased from 1993 levels (BLGU:  $t_{(5)}=3.64$ ,  $p=0.015$ ; GLGU:  $t_{(5)}=2.80$ ,  $p=0.038$ ), and fulmar egg Hg levels in 2008 were down to values similar to 1993 levels.

## Discussion

After four years of annual sampling of murre and fulmar eggs, it is clear that there may be considerable inter-year fluctuation in Hg concentrations. Annual monitoring for the murre and fulmar eggs has allowed us to note that there was a drop in Hg levels in 2005 followed by an increase in 2006, although the fulmars have not gone back up to the 2003 levels. This level of detail would have been missed in the regular five-year sampling regime which called for sampling in only in 2003 and 2008, and not the years in between.

The relative pattern of egg Hg concentrations among species at Prince Leopold Island in 2008 was similar to that reported by Campbell et al. (2005) for birds collected from the Northwater Polynya in 1998. For those five species, Campbell et al. (2005) also found that the kittiwakes had the lowest Hg concentrations and glaucous gulls had the highest levels. Those authors attributed the differences to the birds feeding at different trophic levels with the glaucous gulls feeding at almost a full trophic level higher than the kittiwakes. At Prince Leopold Island, glaucous gulls feed their chicks mainly marine prey including fish and the eggs and nestlings of other seabirds (Nettleship et al. 1990), whereas breeding kittiwakes feed primarily on small surface-schooling fish as well as crustaceans and plankton at the surface (Baird 1994). However, many arctic seabirds overwinter in areas far from their breeding colonies and therefore, both differences in diet as well as overwintering areas may affect their exposure to contaminants.

## Expected Project Completion Date

March 31, 2011

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# Temporal trends and spatial variations in persistent organic pollutants and metals in sea-run char from the Canadian Arctic

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## Abstract

Our study is investigating metal and organic contaminant levels in sea-run char as they return from feeding in the ocean. In 2008, sea-run char were collected from Arviat, Cambridge Bay, Pond Inlet, Kangiqsualujjaq, Nain, and Puvirnituk. Fish varied in size and growth rates over the study area with the largest and oldest fish from Cambridge Bay and the smallest fish from Nain. Mercury concentrations were low with the highest concentrations associated with the largest fish. Mercury exhibited a weak trend on increasing concentrations at Cambridge Bay over 1992-2008. In the western Arctic, organochlorine concentrations declined in Rat River char from 1987 to 2003 with the notable exception of PCBs which occurred in higher concentrations in 2003. At Pond Inlet, in northern Baffin Island, only HCH showed a strong trend of decline over 1987-2007. For Kangiqsualujjaq, in northern Quebec, all organochlorine contaminants

## Résumé

Notre étude se penche sur les niveaux de métaux et de contaminants organiques qui s'accumulent dans l'omble anadrome alors qu'il revient après s'être nourri dans l'océan. En 2008, des ombles anadromes ont été recueillis à Arviat, Cambridge Bay, Pond Inlet, Kangiqsualujjaq, Nain et Puvirnituk. La taille et le taux de croissance des poissons variaient dans l'ensemble de la zone étudiée, les poissons les plus gros et les plus vieux étant ceux de Cambridge Bay et les plus petits, de Nain. Les concentrations de mercure étaient faibles, les plus élevées étant associées aux plus gros poissons. Le mercure indiquait une faible tendance vers une augmentation des concentrations à Cambridge Bay entre 1992 et 2008. Dans l'Arctique occidental, les concentrations de contaminants organochlorés ont diminué chez l'omble de la rivière Rat entre 1987 et 2003, à l'exception notable des BPC qui ont culminé en 2003. À Pond Inlet, au nord de l'île de Baffin, seuls les HCH

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occurred in lower concentrations over 2006-2007 than 1990. PBDEs occurred in low concentrations (1 ng/g) in sea run char from all locations. Community visits and consultations went well with a series of presentations given in Nain.

## Key Messages

- Mercury concentrations were very low in sea-run char caught in 2008, i.e., well below the 0.5 µg/g guideline for the commercial sale of fish and the 0.2 µg/g guideline for frequent consumers of fish. Highest concentrations were associated with the largest fish.
- There was a weak trend of mercury increase at Cambridge Bay when comparisons were based on 1992-2008 data. No trend was evident at Nain.
- Concentrations of legacy contaminants (PCBs, DDT, CBz, HCH, and chlordane) were low. PBDE concentrations were even lower.
- Persistent organic contaminants have generally declined in Rat River char from the western Arctic (PCB is the exception and has increased) and Kangiqsualujjuaq in northern Quebec while, at Pond Inlet, in northern Baffin Island, only HCH has declined in concentrations since the late 1980s or early 1990s.
- Fish appear healthy with only a few instances of disease and/or parasites noted in the majority of collections.

ont affiché une forte tendance à la baisse entre 1987 et 2007. Pour ce qui est de Kangiqsualujjuaq, dans le Nord du Québec, tous les contaminants organo-chlorés étaient présents en concentrations plus faibles en 2006-2007 qu'en 1990. Les PBDE étaient présents en faibles concentrations (1 ng/g) chez l'omble anadrome de tous les sites. Les visites dans les collectivités et les consultations auprès de ces dernières se sont bien déroulées, avec une série de présentations à Nain.

## Messages clés

- Les concentrations de mercure étaient très faibles chez l'omble anadrome attrapé en 2008, soit bien inférieures à la directive de 0,5 µg/g pour la vente commerciale de poisson et à celle de 0,2 µg/g pour la consommation fréquente de poisson. Les plus hautes concentrations se retrouvaient chez les plus gros poissons.
- Une légère tendance à l'augmentation du mercure a été constatée à Cambridge Bay lorsque les comparaisons se fondaient sur les données de 1992-2008. Aucune tendance n'a été constatée à Nain.
- Les concentrations d'anciens contaminants (BPC, DDT, CBz, HCH et chlordane) étaient faibles. Les concentrations de PBDE étaient encore moindres.
- Les contaminants organiques persistants ont en général baissé chez l'omble de la rivière Rat dans l'Ouest de l'Arctique (les BPC faisant exception et ayant augmenté) et à Kangiqsualujjuaq, dans le Nord du Québec, alors qu'à Pond Inlet, dans le Nord de l'île de Baffin, seules les concentrations d'HCH ont baissé depuis la fin des années 1980 ou le début des années 1990.
- Les poissons semblaient être en bonne santé, avec quelques cas seulement de maladie ou de parasite notés dans la majorité des ensembles.

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## Objectives

1. Determine levels of persistent organic pollutants (POPs) and metals (including mercury) as well as "new" POPs from sea-run char which are harvested by Arctic communities.
2. Investigate the role of factors such as fish age, trophic feeding, climate, and location in affecting contaminant body burdens and trends.
3. Contribute to AMAP's assessment of long-term trends in metals and POPs in the

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Arctic and Subarctic and the factors affecting such trends.

4. Provide and explain data to Arctic environmental contaminant committees, health committees, and local communities in a timely manner so that appropriate advice can be given on consuming sea-run char, a country food which is particularly low in contaminant concentration.

## Introduction

Sea-run arctic char (*Salvelinus alpinus*) is primarily an Arctic and Subarctic species, which spends a few weeks in summer feeding in marine waters before migrating inland (Scott and Crossman 1998). Sea-run char are important in traditional diets and the fish species most commonly consumed (Fisk et al. 2003; Van Oostdam et al. 2003). They are sufficiently abundant in some areas to be harvested commercially (e.g., Cambridge Bay, Pangnirtung and Nain) and also are important to the growing sports fishery. Thus, sea-run char have a multitude of economic values in addition to cultural values.

Sea-run char is the only marine fish being monitored for contaminant trends as part of the Northern Contaminant Program with this monitoring complementing the seal, beluga, walrus, and narwhal monitoring programs for the marine environment (Muir et al. 2008b; Stern and Lockhart 2008) and lake trout, burbot, and landlocked char for the freshwater environment (Evans and Muir 2008b; Muir et al. 2008a; Stern and Tomy 2008; Stern et al. 2008). Sea-run char were not included in NCP's monitoring blueprint until 2004 although some contaminant studies were conducted at earlier times, i.e., persistent organic contaminants in the late 1980s and the early 1990s (Muir et al. 1992; Braune et al. 1999) while mercury was measured more frequently as part of as part of the Canadian Food Inspection Service with some data going back to the 1970s (Lockhart et al. (2005). In the late 1990s, Muir et al. (1999) measured mercury in sea-run char from a number of northern Quebec and Labrador locations.

The NCP char monitoring program has two basic objectives. The first is to contribute to the assessment of contaminant trends in Canada's north

and the mechanisms driving these trends. With the implementation of various bans and restrictions on use, long-range atmospheric transport legacy organic contaminants such as HCH, DDT and PCBs should begin to decline although, because many of these compounds have half lives of 10, 20 or more years, this decline may be slow (Hung et al. 2005; Li and Macdonald 2005). Other contaminants may increase in concentration with their increasing usage, e.g., endosulfan (Muir and Köch 2003) or as new chemicals are developed, e.g., polybrominated diphenyl ethers which have properties allowing them to persist in the environment and be transported to the north (de Wit et al. 2006). Emissions of some compounds such as mercury may be increasing as India and China continue to develop economically with a growing number of coal-fired power plants driving this development. Climate change, generally viewed as manifesting itself in a warming trend in Canada's north, may be enhancing annual production with reverberations through aquatic and marine food webs (Rouse et al. 1997; Schindler 2001; Symon et al. 2005). These reverberations may have complex effects on contaminant levels in organisms being monitored for time trends (Stern et al. 2005; Gaden et al. 2009). Therefore, contaminant trend monitoring involves several considerations.

The second objective of the sea-run char program is to obtain detailed information on spatial concentrations in contaminants in sea-run char across their range in Canada's Arctic and Subarctic waters. Spatial coverage is needed to embrace the various geographic and climatic regions of northern Canada. This is especially important as the western Arctic appears to be experiencing a more pronounced warming trend than other parts of Canada and because the west is closer to atmospheric sources of mercury from Asia than the eastern Arctic.

## Activities in 2008/2009

In 2008, sea-run char were successfully collected at most communities including Cambridge Bay, Pond Inlet, and Arviat in Nunavut, Puvimuituq and Kangiqsualujjuaq in northern Quebec, and Nain in Labrador. There was little support for our study in Clyde River but, following the NCP Workshop in September, Holman Island agreed to participate

**Table 1. Mean ( $\pm$  standard deviation) fork length, age, weight, liver weight, liver somatic index, and mercury concentrations (wet weight basis) for sea-run char caught in 2008.**

Location	Fork length (mm)	Age (yr)	Weight (gm)	Liver Weight (gm)	LSI	Hg ( $\mu\text{g/g}$ )
Arviat	622 $\pm$ 88	6.7 $\pm$ 1.7	3,545 $\pm$ 1,207	118.9 $\pm$ 44.79	3.33 $\pm$ 0.46	0.08 $\pm$ 0.03
Cambridge Bay	648 $\pm$ 82	11.5 $\pm$ 2.7	3,545 $\pm$ 1,090	62.99 $\pm$ 24.52	1.83 $\pm$ 0.35	0.08 $\pm$ 0.02
Pond Inlet	530 $\pm$ 59	11.1 $\pm$ 1.9	1,932 $\pm$ 792	44.40 $\pm$ 26.21	2.22 $\pm$ 0.74	0.06 $\pm$ 0.03
Kanqisualujjaq	557 $\pm$ 61	7.4 $\pm$ 1.7	2,208 $\pm$ 716	54.55 $\pm$ 23.09	2.43 $\pm$ 0.54	0.04 $\pm$ 0.02
Nain	487 $\pm$ 44	7.8 $\pm$ 2.3	1,633 $\pm$ 459	43.86 $\pm$ 16.66	2.66 $\pm$ 0.43	0.02 $\pm$ 0.01
Puvirnitq	515 $\pm$ 56	5.1 $\pm$ 1.2	1,571 $\pm$ 392	31.87 $\pm$ 9.90	2.02 $\pm$ 0.29	0.04 $\pm$ 0.01

in our study. This was good news as we have been trying for some time to get a monitoring site west of Cambridge Bay.

#### ***General features of fish caught in 2008***

Our study design calls for 20 fish to be collected per location and these fish were either shipped whole to us (Cambridge Bay, Pond Inlet, Holman Island), as subsamples (Puvirnitq and Kangisualujjaq prepared by Dr. Kwan) or obtained when we visited the community (Arviat, Nain).

Length, weight, age, and gender were determined on all fish from each location. In addition, we have been measuring the liver, gonad, and stomach and noting the presence of parasites and/or disease. A fillet sample and the liver, stomach, and gonad were retained from each fish. Carbon and nitrogen isotope and percent moisture analyses are performed on all fish. Ten of the 20 fish from each location were selected for metals, legacy organic contaminants and PDBE and PFA analyses, with our sample archive consisting of various tissue samples from all 20 fish per location. Archived samples are maintained at  $-40^{\circ}\text{C}$  in a walk-in freezer at the National Hydrology Research Centre which has a monitoring system overseen by commissionaires 24-hours a day, 365 days a year. The majority of our 2008-caught samples are currently in queue at analytical laboratories. Highlights of our findings to date are as follows.

#### ***Results***

Fish varied markedly in their mean size with the smallest fish being from Nain and the largest from Cambridge Bay and Arviat. Pond Inlet fish typically

have been larger and older in previous collections than in 2008, i.e. a mean fork length of 618 mm and 13.3 years in 2005, 680 mm and 15.0 yr in 2006, and 683 mm and 14.1 yr in 2007. In 2008, fish may have been caught later in the summer with smaller fish possibly migrating inland later. As in previous years, the oldest fish were in the more northerly locations, i.e., Pond Inlet and Cambridge Bay. Fish growth rates also varied with fish reaching 600 mm fork length at 5.9 yr at Kanqisualujjaq and Puvirnitq, 6.7 yr at Arviat, 8.5 yr at Nain, 10.2 yr at Cambridge Bay, and 12.1 yr at Pond Inlet. While char growth rate appears to diminish with the generally expected pattern with increasing latitude, the slower growth rate at Nain in comparison to Kanqisualujjaq and Puvirnitq is unexpected. Many factors could account for this including food availability. In addition, Nain is near the southern limit of the arctic char range and these fish may not be living in their thermal optimum (Scott and Crossman 1998). Liver weight varied roughly as a function of animal weight with the liver somatic index (liver weight divided by body weight and multiplied by 100) varying from location to location; the lowest index was for Cambridge Bay. For comparisons, the mean LSI for lake trout from Lutsel K'e in Great Slave Lake is 1.7 and for West Basin lake trout is 1.3.

We continue to gather information on fish health noting pale and dark livers, gill parasites, skinny fish, etc. As our data base builds, we will be able to investigate how contaminant body burden and metrics of fish health are related. Skinny fish tend to be older fish with higher mercury concentrations.

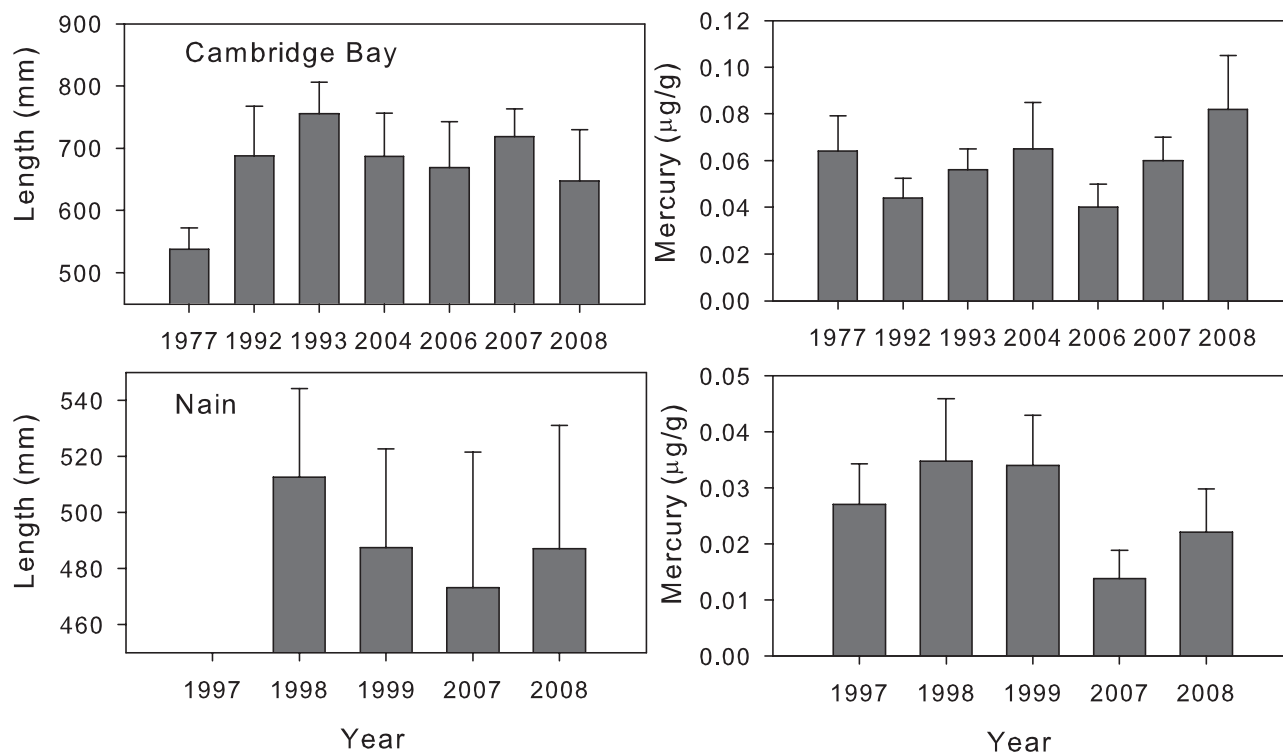


Figure 1. Time trends in fork length and mercury concentrations for sea-run char collected at Cambridge Bay and Nain.

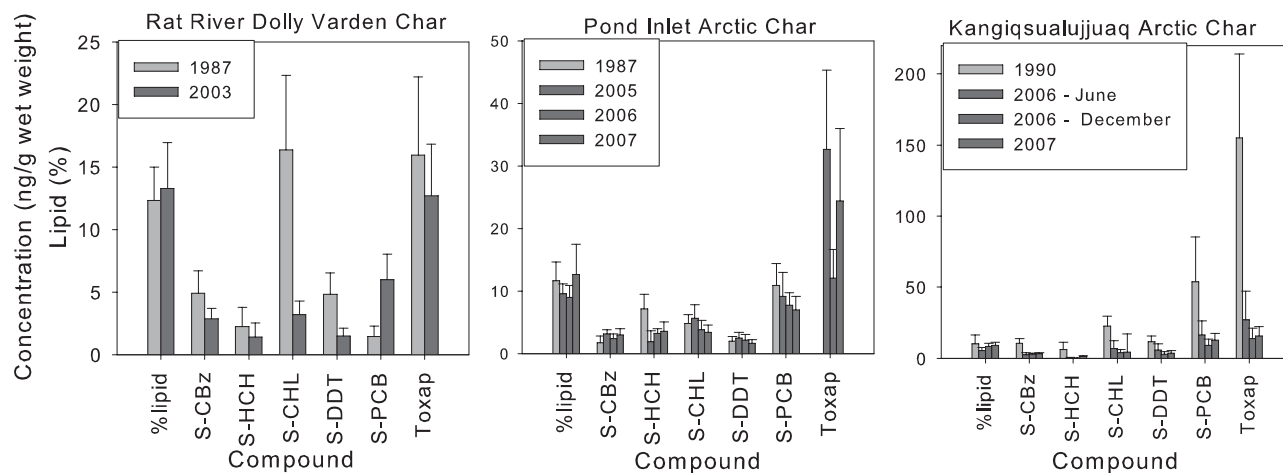


Figure 2. Time trends in lipids and persistent organic contaminants in sea-run char at Rat River, Pond Inlet, and Kangiqsualujuaq.

### Temporal variations in contaminant levels

Cambridge Bay has the longest record for investigating mercury time trends in sea-run char with Lockhart et al. (2005) reporting length and mercury data for 1977, 1992, and 1993 (Fig. 1). Fish caught in 1997 were small but had relatively high mercury concentrations for their length. Since 1977, mean char size has ranged from 648–755 mm with mercury concentrations ranging

from 0.04–0.08 µg/g. Excluding 1977, there is a weak trend for mercury to be increasing with time ( $r = +0.46$ ). There is less evidence of time trends in mercury at Nain. Fish caught in 1999 were of the same average size as fish caught in 2008 with lower mercury concentrations in 2008.

Time trends in persistent organic contaminants are being investigated (Fig. 2). For Rat River char,



**Table 2. PBDE concentrations (ng/g wet weight) in sea-run char collected in 2007. The two most abundant PBDEs for each location are shown in bold.**

	28	49	47	66	100	99	85	154	153	183	209	Sum
Cambridge Bay	0.130	0.023	<b>0.201</b>	0.038	0.057	0.102	0.005	0.050	0.014	0.006	<b>0.276</b>	0.902
Hall Beach	0.039	0.011	<b>0.081</b>	0.000	0.019	0.020	0.000	0.003	0.002	0.000	<b>0.331</b>	0.507
Hopedale	0.033	0.017	<b>0.091</b>	0.003	0.019	0.022	0.000	0.002	0.002	0.004	<b>0.043</b>	0.215
Kaniqsual.	<b>0.230</b>	0.049	<b>0.265</b>	0.003	0.067	0.066	0.000	0.012	0.010	0.004	0.004	0.694
Kangirsuk	<b>0.099</b>	0.017	<b>0.144</b>	0.000	0.033	0.043	0.000	0.010	0.008	0.004	0.090	0.444
Nain	<b>0.069</b>	0.020	<b>0.088</b>	0.006	0.020	0.026	0.000	0.011	0.011	0.007	0.003	0.260
Pond Inlet	0.050	0.031	<b>0.069</b>	0.000	0.017	0.020	0.000	0.000	0.015	0.000	<b>0.266</b>	0.266
Vittrekwa	0.062	0.011	<b>0.123</b>	0.018	0.031	0.046	0.000	0.000	0.000	0.002	<b>0.333</b>	0.333

in the western Arctic, lipid levels were similar in 1987 and 2003, but all organochlorines, with the exception of PCBs were substantially lower in the later period suggesting a decline in inputs; PCB concentrations were higher. At Pond Inlet, in northern Baffin Island, only HCH and possibly toxaphene appear to have declined in concentration since 1987. In contrast, all organochlorines are occurring in lower concentrations at Kaniqsualujjuaq when 2006 and 2007 data are compared with 1990 data.

PBDEs were detected in very low concentrations in sea-run char muscle samples with total concentrations ranging from 0.260 ng/g at Nain to 0.902 ng/g at Cambridge Bay (Table 2). The predominant PBDEs were PBDE 28 and 47 although PBDE 209 was abundant at Cambridge Bay, Hall Beach, Pond Inlet and in Vittrekwa River Dolly Varden char.

## Community Consultations and Presentations

Community consultations went well. While we were unable to obtain sea-run char from Clyde River, we were able to obtain fish from Holman Island, in the western Arctic; Holman Island agreed to participate in this study again in 2009 and ideally will become our western most long-term monitoring site. Arviat and Nain were visited. We could not visit Pond Inlet because accommodations were not available. While in Nain, Mary Denniston and John Lampe arranged for a radio

interview to discuss the char study, a demonstration of fish dissection techniques was offered to the general public, and an open house to further discuss the study. Results of our research to date were reported at the September 2008 NCP workshop. Cathy Aitoak, from the Cambridge Bay Hunters and Trappers was able to attend the NCP workshop and meet with us, other researchers, agencies, and community members involved with NCP. A presentation on this study also was given at the 35<sup>th</sup> Aquatic Toxicology Workshop in October 2008 (Evans et al. 2008c). In addition, we continue to contribute to the AMAP expert work groups for mercury and POPs including a December 2008 meeting in Quebec City.

## Acknowledgements

We thank Molleen Anaviapik (Mittimatalik Hunters and Trappers), Cathy Aitoak (Ekaluktutiak Hunters and Trappers), Annie Amauyak (Arviat Hunters and Trappers), Lillian Kanayok and Bessie (Holman Island Hunters and Trappers) and Mary Denniston and John Lampe (Nunavsiavut Government) for their support and participation in this study.

## Expected Completion Date

This project is ongoing with sea-run char to be collected annually for temporal trend analyses at 3-4 communities. Sea-run char will be collected annually from an additional two or three community to investigate POPs and inorganic metal

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contaminant concentrations to obtain information for communities not yet investigated or to revisit communities previously studied but not part of the intense temporal trend monitoring. The current NCP program runs to 2010 but we are hopeful that it and this study will be continued.

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# Temporal trends of Persistent Organic Pollutants and Mercury in Landlocked char in the High Arctic

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## Abstract

This long term study is examining trends over time of mercury and other trace elements, as well as legacy and new persistent organic pollutants (POPs) in landlocked Arctic char collected annually from three lakes near the community of Resolute Bay on Cornwallis Island (Amituk, Char and Resolute) and in Lake Hazen in Quttinirpaaq National Park on Ellesmere Island. In 2008, arctic char samples were successfully collected from Lakes Amituk, Char, Resolute, and Hazen. To assess temporal trends, results from 2008 were combined with previous results from the same lakes. Mercury concentrations have increased in Arctic char from lakes Amituk, Char and Resolute, however, the increases are statistically significant

## Résumé

Cette étude à long terme se penche sur les tendances temporelles du mercure et autres éléments en trace, ainsi que des polluants organiques persistants (POP), anciens et nouveaux, chez l'omble chevalier dulcicole recueillis au moyen d'échantillons tous les ans dans trois lacs près de la collectivité de Resolute Bay, sur l'île Cornwallis (Amituk, Char et Resolute) et dans le lac Hazen, dans le parc national Quttinirpaaq, sur l'île d'Ellesmere. En 2008, des échantillons d'omble chevalier ont été prélevés dans les lacs Amituk, Char, Resolute et Hazen. Afin d'évaluer les tendances temporelles, les résultats de 2008 ont été combinés aux résultats antérieurs des mêmes lacs. Les concentrations de mercure ont augmenté chez l'omble chevalier des lacs Amituk, Char et Resolute, mais

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only in Amituk Lake. Statistical analysis of trends of POPs in landlocked char from the four lakes in this study shows overall declining trends with average annual % change of  $\alpha$ -HCH (14%) > toxaphene (9%) >  $\Sigma$ DDT (7%) >  $\Sigma$ PCBs (5%). While concentrations of brominated diphenyl ethers increased in char in Resolute and Amituk Lakes until about 2005 they are now declining.

## Key Messages

- Mercury concentrations appear to be increasing slowly in arctic char from small lakes but not in Lake Hazen, the largest lake in this study
- Concentrations of most POPs such as toxaphene and DDT in landlocked char have decreased significantly in the study lakes
- Brominated diphenyl ether flame retardants have increased significantly in char from all four lakes over the past 10 years but are now declining in Resolute and Amituk lakes

ces augmentations ne sont significatives, sur le plan statistique, que dans le lac Amituk. L'analyse statistique des tendances des POP chez l'omble chevalier dulcicole des quatre lacs de cette étude indique des tendances générales à la baisse, avec un pourcentage de changement annuel moyen de  $\alpha$ -HCH (14 %) > toxaphène (9 %) >  $\Sigma$ DDT (7 %) >  $\Sigma$ PCBs (5 %). Même si les concentrations d'éthers de diphenyle bromés ont augmenté chez l'omble des lacs Resolute et Amituk jusqu'à environ 2005, elles sont maintenant à la baisse.

## Messages clés

- Les concentrations de mercure semblent augmenter lentement chez l'omble chevalier des petits lacs, mais pas dans le lac Hazen, le plus grand de cette étude.
- Les concentrations de la plupart des POP comme le toxaphène et le DDT chez l'omble chevalier dulcicole ont nettement diminué dans les lacs étudiés.
- Les ignifugeants à base d'éther de diphenyle bromé ont nettement augmenté chez l'omble des quatre lacs au cours des 10 dernières années, mais sont maintenant à la baisse dans les lacs Resolute et Amituk.

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## Objectives

1. Determine long term temporal trends of persistent organic pollutants (POPs) and metals in landlocked Arctic char from lakes in the Canadian high arctic islands by analysis of annual or biannual sample collections.
2. Investigate factors influencing contaminant levels in landlocked char such as the influence of sampling time, water temperature, diet and climate warming.
3. Determine levels of current POPs and metals as well as "new" potential POPs in fish from lakes of importance to the community of Resolute Bay (Qausuittuq) and provide this information on a timely basis.

## Introduction

The contaminant burdens of lakes in the high Arctic are derived from long-range atmospheric

transport and deposition to the lake surface and from the lake catchment (Macdonald et al 2000). Local catchments are typically the primary source of water for arctic lakes (Wrona et al 2005). An intense 2-3 week runoff period (freshet) in spring/summer and direct precipitation both represent significant fractions of lake water budgets.

The sedimentary records of persistent organic pollutants (POPs) in small arctic lakes show that they reflect global trends in emissions (Muir et al. 1996; Stern et al. 2005; Breivik et al. 2006). About 30% of PCB inputs, and 8-33% of organochlorine pesticide (OCP) inputs, to Amituk Lake from snow melt in 1994 were retained in the lake (Macdonald et al. 2000). Sedimentary records for mercury in 18 high arctic lakes showed overall increasing trends since the 1950s with anthropogenic fluxes in agreement with modeled estimates (Muir et al. 2009). Given that lake sediments appear to reflect global trends of POPs and mercury it follows that



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trends of these contaminants in lake food webs and in arctic char, the sole top predator fish in most high arctic lakes, should also be reflected relatively quickly in food webs and top predator fishes.

Over the past few years the number of landlocked char populations and their food webs surveyed for contaminants in the Canadian arctic has increased significantly as a result of the PhD research by Klaus Gantner (Gantner et al. 2009a; Gantner et al. 2009b; Gantner et al. 2009c). Total mercury concentrations in char from 24 lakes showed a positive correlation with catchment-to-lake area ratio but concentrations were not related to latitude or longitude or food chain length (Gantner et al. 2009b).

This study reports on results of continued annual sampling and contaminant analysis of char at Resolute Lake, Char and Amituk lakes on Cornwallis Island, as well as from Lake Hazen in Quttinirpaaq National Park on Ellesmere Island. Further details on past results from these study lakes are given in previous synopsis reports (Muir et al. 2005; Muir et al. 2006, 2007, 2008).

## Activities in 2008/2009

### *Sample collection:*

Samples were successfully collected in July 2008 from lakes Amituk, Char, Hazen, and Resolute by our field teams. Fish were dissected in Polar Continental Shelf Project labs at Resolute or in the field (Lake Hazen). Samples (skin- on fillets) were frozen in Resolute and then shipped to the Environment Canada labs, Burlington, Ontario, and stored at -20°C until analysis. Char otoliths were shipped to J. Reist (DFO Winnipeg). Aging of the char was done by J. Babaluk (DFO Winnipeg).

### *Chemical analysis:*

Analytical methods were unchanged from previous reports (Muir et al. 2006, 2007). All analyses were performed by the National Laboratory for Environmental Testing (NLET) at Canada Centre for Inland Waters in Burlington. Mercury and 31 other elements were analysed in Arctic char muscle (skinless). Organohalogen compounds were determined in homogenized char (muscle plus skin) samples. PCBs and OCPs were analysed in final cleaned up extracts were by gas chromatography with electron-capture detection (GC-ECD). All organohalogen analyses were conducted in the

NLET “ultraclean” room (carbon and HEPA filtered air; positively pressured) to minimize background contamination. Toxaphene, endo-sulfan, PBDEs and hexabromocyclododecane (HBCD) were analysed by low resolution GC-negative ion mass spectrometry (NIMS) using a HP 5975 MSD. Toxaphene was determined as “total” toxaphene using a technical toxaphene standard and also by quantification of individual chlorobornanes (see Muir et al. (2004) for further details on methods).

Perfluoroalkyl acids (PFAs) in char muscle were analysed using the method of Hansen et al which involved extraction with methyl tert-butyl ether (MTBE) and tetrabutylammonium hydrogen sulfate as an ion pairing reagent. The extract was cleaned up on a 100 mg carbon solid phase extraction cartridge. The sample was eluted with methanol and the eluant was then taken to dryness and reconstituted in 1 ml of 50/50 methanol/water. The samples was then analysed by liquid chromatography tandem mass spectrometry as described by Butt et al. (2007). Major compounds analysed were perfluorohexanesulfonate (PFHxS), perfluoroheptanesulfonate (PFHpSA), perfluorooctane sulfonate (PFOS), perfluorooctanoate acid (PFOA), perfluorononanoate (PFNA), perfluorodecanoate (PFDA), perfluoroundecanoate (PFUnA) and perfluorooctansulfonamide (PFOSA).

### *Stable isotope analyses:*

Muscle from all fish analysed for mercury and POPs were analysed for stable isotopes of carbon ( $\delta^{13}\text{C}$ ) and nitrogen ( $\delta^{15}\text{N}$ ) at University of Waterloo Environmental Isotope Lab in muscle samples using isotope ratio MS.

### *Quality assurance (QA):*

Certified reference materials (CRMs) for heavy metals included DOLT-2, DORM-2 and TORT-2 (National Research Council of Canada) and for PCBs, OCPs, and PBDEs either 1588a cod liver or 1974b mussels from NIST (National Institute of Standards and Technology). CRMs and reagent blanks were also run with each sample batch of 10 samples and all results for organohalogens compounds were blank subtracted. Blanks were generally <10% of measured values for OCPs, PCBs and PBDEs. NLET organics and metals labs are participants in the NCP Quality Assurance



Program. The NLET labs are accredited by the Standards Council of Canada through Canadian Environmental Analytical Laboratory program to the standard CAN-P-4D (ISO/IEC 17025).

#### **Statistical analyses:**

Non-detect concentrations were replaced with half the instrumental detection limit for calculation of arithmetic and geometric means and standard deviations. Results for each collection year were first tested for normality using the Shapiro-Wilk test. Results for all elements and POPs were log10 transformed in order to reduce coefficients of skewness and kurtosis to <2. Geometric mean concentrations and upper/lower 95% confidence intervals were calculated with log transformed data and back transformed for graphical presentation. Results for POPs were lipid adjusted by dividing by fraction lipid.

## **Results**

#### **Mercury and other elements:**

Average size and age of char as well as concentrations of mercury in muscle for 2008 are presented in Table 1. Not shown are results for 31 other elements which were determined in all samples.

We have reported on these previously. Generally 22 to 24 are above detection limits in char muscle.

Temporal trends of mercury concentrations in char were not assessed for this report. We have previously shown that mercury has increased (using weight adjusted or unadjusted data) in Amituk, Char and Resolute lakes over the past 10-18 years. However results were statistically significant only for Amituk (Muir et al. 2008).

Compared to previous years mercury concentrations have declined in insectivorous char from Lake Hazen. When the combined dataset for Lake Hazen char is adjusted for trophic position using  $\delta^{15}\text{N}$ , no decline in mercury in Arctic char from Lake Hazen (1992 – 2006) is observed (Gantner et al. 2009c). Thus interpretation of mercury trends in arctic char requires careful attention to covariates such as size and trophic position. Gantner et al. (2009b) have recently shown that mercury was significantly correlated ( $P < 0.05$ ) with length, weight, and  $\delta^{15}\text{N}$  in char from Lake Hazen and many other high arctic lakes. For 24 lakes, including the 4 reported on here, total mercury concentrations in char showed a positive correlation with catchment-to-lake area

**Table 1. Arithmetic and geometric ranges of length, weight and mercury (ug/g wet wt) mean concentration in muscle of landlocked Arctic char collected in 2008 from four Arctic lakes**

Lake	Stat	N	Length (cm)	Weight (g)	Mercury (ug/g wet weight)	$\delta^{13}\text{C}$ (‰)	$\delta^{15}\text{N}$ (‰)
Amituk	Mean	3	45.7	896	0.94	-21.2	11.8
	Geomean		43.4	559	0.83		
	Min		27.4	118	0.41	-21.6	11.2
	Max		61.0	1702	1.38	-20.9	13.0
Char	Mean	9	35.6	637	0.32	-23.5	11.2
	Geomean		32.1	199	0.27		
	Min		16.9	31	0.10	-24.8	9.4
	Max		61.0	2250	0.62	-21.3	12.7
Hazen	Mean	10	40.6	674	0.03	-19.6	10.7
	Geomean		40.2	616	0.03		
	Min		34.5	405	0.01	-21.4	9.0
	Max		50.4	1382	0.07	-17.8	13.7
Resolute	Mean	11	42.9	628	0.17	-22.1	11.3
	Geomean		42.7	612	0.16		
	Min		39.0	474	0.10	-23.3	10.2
	Max		51.0	1000	0.34	-21.6	13.4

ratio but were not related to latitude or longitude or food chain length (Gantner et al. 2009b).

#### **Persistent organohalogen compounds:**

In 2008-09 analyses continued for legacy POPs (PCBs, DDT, toxaphene, and other organochlorine (OC) pesticides and byproducts) as well as PBDEs and other brominated flame retardants in landlocked char from Amituk, Char, Hazen and Resolute Lakes. Overall, concentrations of PCBs, DDT and toxaphene reported in previous years (Muir et al. 2006, 2007, 2008) remain basically unchanged. A new addition in 2008 was the determination of endosulfan sulfate a recalcitrant degradation product of endosulfan using GC-MS. In char from Lake Hazen (N=10) the sulfate averaged  $0.012 \pm 0.005$  ng/g wet wt (ww) compared to  $0.019 \pm 0.010$  ng/g ww for  $\beta$ -endosulfan and  $0.011 \pm 0.005$  ng/g ww for  $\beta$ -endosulfan. Therefore it is a significant fraction of total endosulfan in char muscle.

Temporal trends for PCBs, DDT, toxaphene,  $\alpha$ - and  $\beta$ -HCH, and PBDEs for all results up to 2008 were examined using the PIA software package (Bignert 2007) that has been utilized for studies of trends and statistical power of Arctic datasets for POPs (Rigét et al. 2009) and mercury (Bignert et al. 2004). For this preliminary assessment all data (lipid normalized) were utilized and no other adjustments (e.g. for weight or trophic status) were performed. By using the PIA package we were able to assign an annual % decline or

increase for each POP (Table 2). The program also provided confidence limits on the % decline or increase and an estimate of the statistical power of the dataset (i.e. in this case the lowest detectable % change with a power of 80% at  $\alpha=0.05$ ). For clarity of presentation these are not shown in Table 2.

Toxaphene concentrations were found to have declined significantly ( $P < 0.05$ ) only in Amituk Lake (11%/yr) and Lake Hazen (12%) (Table 2). In Resolute Lake concentrations remained unchanged (non-statistically significant increase of 11% per year), however, toxaphene results were only available from 2003 to 2008 for this lake. PCBs (sum of 87 congeners and  $\Sigma$ DDT (sum of o,p'- and p,p'- isomers) declined significantly in lakes Amituk, Char and Hazen but not in Resolute Lake. The  $\alpha$ -HCH isomer declined significantly in all 4 lakes with the overall greatest annual changes (average of 14% for the 4 lakes) while  $\beta$ -HCH did not show significant declines and was generally higher in samples from the mid-00s than in the 1990s.

Total PBDEs (sum of BDE congeners in order of their elution on a GC column: 17, 28, 49, 47, 66, 100, 99, 85, 154, 153, 138, 183, 190, 209) increased significantly and rapidly in most of the lakes, however, the year for maximum concentrations varied. Maxima were found in 2003 in Amituk, 2005 in Resolute and 2007 in Lake Hazen. However, in Char Lake total PBDEs have continued to

**Table 2. Percent annual decline (negative) and increase in selected POPs trends<sup>1</sup> in arctic char from the four study lakes using the PIA program (Bignert 2007).**

	Time period	Sampling Years	Toxaphene	$\Sigma$ PCB	$\Sigma$ DDT	$\alpha$ -HCH	$\beta$ -HCH	$\Sigma$ PBDEs <sup>2</sup>
Amituk	1989-2008	9	-11	-6.6*	-9.9*	-19*	7.7	60*
	2003-2008	5	—	—	—	—	—	-30*
Char	1993-2008	8	-4.7	-8.4*	-9.1*	-16*	-5.5	4.5*
Hazen	1990-2008	7	-12*	-8.1*	-8.2	-11*	8.4	—
	1990-2007	6	—	—	—	—	—	59*
Resolute	1997-2008	10	11 <sup>3</sup>	0.8	-1.8	-10*	-1.1	21*
	2005-2008	4	—	—	—	—	—	-32*

<sup>1</sup> \* indicated statistically significant trend ( $P < 0.05$ )

<sup>2</sup> Results for PBDEs generally from the 1990s to mid-00s; dates shown in the time period column

<sup>3</sup> Results for toxaphene in Resolute Lake char are for 2003-2008 only

increase although at a much slower rate than in the other lakes.

Other brominated flame retardants in arctic char muscle samples were present at concentrations near or below method detection limits. These included hexabromocyclododecane (HBCD), pentabromoethylbenzene, bis(tribromophenoxy) ethane and decabromodiphenyl ethane. HBCD was detected in char from Amituk, Char and Resolute lakes, but not in Lake Hazen, at concentrations ranging from about 0.05 to 0.5 ng/g wet wt. Further confirmation of these results using LC-MS/MS is planned.

PFAs were previously reported for all lakes in 2007 (Muir et al. 2008). Analyses of the 2008 samples revealed very similar concentrations. Again concentrations, particularly of PFOS, were much higher in char from Resolute Lake than in all other lakes. Analyses of lake waters and sediments have shown that Amituk and Char have background concentrations that are expected in remote lakes receiving PFAs solely via atmospheric deposition (Stock et al. 2007) while Resolute Lake appears to have been contaminated with aqueous film forming foams which are used for fire fighting at most airports.

## Discussion and Conclusions

A detailed statistical analysis of trends of POPs in landlocked char from the four lakes in this study shows overall declining trends of legacy chemicals with annual % change of  $\alpha$ -HCH > toxaphene >  $\Sigma$ DDT >  $\Sigma$ PCBs. The greater rate for  $\alpha$ -HCH and toxaphene may reflect the complete cessation of their uses globally possibly as long as 10-20 years ago. In contrast PCBs continue to be used in electrical equipment in large quantities and are only slowly being phased out while DDT use also continues, including possibly in Russia according to a recent AMAP report (AMAP 2009). The annual rates of decline for  $\alpha$ -HCH and  $\Sigma$ DDT are quite similar to those reported in Arctic air from Alert and Zeppelin (Svalbard) (Hung et al. 2009).

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# Spatial and long-term trends in persistent organic contaminants and metals in lake trout and burbot from the Northwest Territories

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## Abstract

Our study is designed to find out whether contaminant levels are changing in lake trout and burbot in Great Slave Lake and mercury in lake trout in other lakes and why. Mercury levels in lake trout from Stark Lake were lower in 2008 than 2003 while mercury concentrations in Trout Lake have almost doubled with a 600-mm fish having 0.61  $\mu\text{g/g}$  mercury versus 0.35  $\mu\text{g/g}$  in 2003. Persistent organic contaminants were measured in lake trout from Great Bear Lake in 2007: NCP provided supplemental funds for these analyses. Legacy organic contaminant concentrations were 1.5 (CBz) to 2.8 (DDT) times lower in 2007 than 2002; mercury concentrations also were lower. PBDE concentrations were very low. In lake trout from Great Slave Lake, we are observing a general trend of a decline in lipid levels and an increase in littoral zone carbon in the diet with a concomitant increase in mercury concentrations and a decline in HCH and DDT; PCBs are showing a weak trend of increase in the East Arm but not the West Basin. Burbot is a more near-shore fish

## Résumé

Notre étude vise à déterminer si les niveaux de contaminants changent chez la truite grise et la lotte du Grand lac des Esclaves, et ceux du mercure chez la truite grise d'autres lacs, et pourquoi. Les niveaux de mercure chez la truite grise du lac Stark étaient moins élevés en 2008 qu'en 2003, alors que les concentrations de mercure dans le lac Trout ont pratiquement doublé, un poisson de 600 mm ayant 0,61  $\mu\text{g/g}$  de mercure comparativement à 0,35  $\mu\text{g/g}$  en 2003. Les contaminants organiques persistants ont été mesurés chez la truite grise du Grand lac de l'Ours en 2007 : le Programme de lutte contre les contaminants dans le Nord (PLCN) a fourni des fonds supplémentaires pour ces analyses. Les concentrations d'anciens contaminants organiques étaient de 1,5 (CBz) à 2,8 (DDT) fois plus faibles en 2007 qu'en 2002; les concentrations de mercure étaient aussi plus faibles. Les concentrations de PBDE étaient très faibles. Chez la truite grise du Grand lac des Esclaves, nous observons une tendance générale à la baisse des niveaux de lipides et une augmentation du carbone de la zone littorale



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and is showing a weak trend of increasing lipid content and increased feeding on littoral zone carbon. There is a strong trend of increasing mercury concentrations while HCH and DDT concentrations are declining; PCBs are showing a trend of increase (East Arm) or a slight decrease (West Basin). Overall, considering the two species and the trends observed, it appears that HCH and DDT inputs to the lake have decreased while PCBs have changed little. Some of the time trends being observed in the fish may be driven by changes in their biology which, in turn, could be driven by changes in broader features of the lake.

dans le régime, avec une augmentation concomitante des concentrations de mercure et une baisse de HCH et de DDT; les BPC montrent une faible tendance à l'augmentation dans le bras oriental, mais pas dans le bassin ouest. La lotte est un poisson qui vit plus près du rivage et elle montre une faible tendance à l'augmentation de la teneur en lipides et une alimentation accrue en carbone de la zone littorale. On a observé une forte tendance à l'augmentation des concentrations de mercure alors que celles de HCH et de DDT baissent. Les BPC montrent une tendance à la hausse (bras oriental) ou une faible baisse (bassin ouest). D'une façon générale, étant donné les deux espèces et les tendances observées, il semble que l'apport de HCH et de DDT dans le lac a baissé alors que celui des BPC a peu changé. Certaines des tendances temporelles observées chez le poisson peuvent être influencées par des changements biologiques qui, à leur tour, pourraient être influencés par des changements de caractéristiques plus générales du lac.

## Key Messages

- Mercury levels were lower in lake trout from Stark Lake in 2008 than 2003: a similar trend was observed for lake trout from Great Bear Lake. However, mercury levels have almost doubled in Trout Lake fish (2003 and 2008 comparisons); a similar trend was observed for Kelly Lake and Lac Ste. Therese in earlier studies. It appears that small lakes with larger watersheds located on Palaeozoic deposits may be most vulnerable to mercury increases.
- Mercury is showing a strong trend of increase in lake trout from two regions of Great Slave Lake. Burbot is showing an even more pronounced trend of increase. This increase is occurring as fish are relying more on littoral zone carbon.
- Legacy organic contaminant concentrations were 1.5-2.8 times lower in Great Bear Lake in 2007 than 2002. Contaminant concentrations were higher than in Great Slave Lake and more similar to those observed in Lake Laberge with the exception of which is higher. However, Great Bear Lake fish are older.

## Messages clés

- Les niveaux de mercure chez la truite grise du lac Stark étaient moins élevés en 2008 qu'en 2003 : une tendance similaire a été observée chez la truite grise du Grand lac de l'Ours. Toutefois, les niveaux de mercure ont pratiquement doublé chez les poissons du lac Trout (comparaisons entre 2003 et 2008); une tendance similaire a été observée lors d'études antérieures dans les lacs Kelly et Sainte-Thérèse. Il semble que les lacs plus petits, avec bassins d'alimentation plus grands et situés sur des dépôts paléozoïques soient plus sensibles aux augmentations de mercure.
- Le mercure montre une forte tendance à l'augmentation chez la truite grise de deux régions du Grand lac des Esclaves. La lotte montre une tendance à l'augmentation encore plus marquée. Cette augmentation survient alors que les poissons dépendent plus du carbone de la zone littorale.
- Les concentrations d'anciens contaminants organiques étaient de 1,5 à 2,8 fois moins élevées dans le Grand lac de l'Ours en 2007 qu'en 2002. Les concentrations de contaminants étaient plus élevées que dans le Grand lac des Esclaves et ressemblaient plus à celles observées dans le



- Lipid levels are declining in Great Slave Lake trout as is HCH and DDT: PCBs are showing a weak trend of decline in the West Basin and an increase in the East Arm.
- Lipid levels are showing a weak trend of increase in burbot but HCH and DDT concentrations are declining: PCBs are showing a weak trend of increase in the East Arm and a decline in the West Basin.
- Overall, HCH and DDT concentrations appear to be declining in Great Slave Lake while PCB concentrations are staying the same (West Basin) or increasing (East Arm). Changes in carbon feeding and, for lake trout, lipids, may be accentuating some of these trends and possibly masking others.
- Changes in the biology of lake trout and burbot are being driven by unknown changes in the broader features of Great Slave Lake.

lac Laberge, sauf qu'elles y étaient plus élevées. Mais les poissons du Grand lac de l'Ours sont plus vieux.

- Le niveau des lipides baisse chez la truite du Grand lac des Esclaves, tout comme celui de HCH et de DDT : les BPC montrent une faible tendance à la baisse dans le bassin ouest et une augmentation dans le bras oriental.
- Les niveaux des lipides montrent une faible tendance à la hausse chez la lotte, mais les concentrations de HCH et de DDT baissent. Les BPC montrent une faible tendance à la hausse dans le bras oriental et une baisse dans le bassin ouest.
- D'une façon générale, les concentrations de HCH et de DDT semblent baisser dans le Grand lac des Esclaves alors que les concentrations de BPC restent les mêmes (bassin ouest) ou augmentent (bas oriental). Les changements de l'alimentation en carbone et, chez la truite grise, des lipides, peuvent accentuer certaines de ces tendances et en masquer d'autres.
- Les changements biologiques qui se produisent chez la truite grise et la lotte sont influencés par des changements inconnus des caractéristiques plus générales du Grand lac des Esclaves.

## Introduction

The Northern Contaminants Program (NCP) began in the early 1990s when concerns were raised about the presence of persistent organic contaminants in arctic and subarctic waters. It has been renewed three times. Currently the focus is on trend monitoring and human health. Trend monitoring is required to track the success of international programs in reducing contaminant inputs to the environment, including the north, and to identify the presence of new contaminants in arctic and subarctic environments. NCP contributes to the Arctic Monitoring and Assessment Program (AMAP) which is an international program based on contaminant trend monitoring in the circumpolar north. The goal of this monitoring is to detect a 10% annual change in contaminant concentrations over a period of 10-15 yrs with a

power of 80% and a confidence level of 95%. In addition, this monitoring must distinguish trends in contaminant levels that are being driven by changes in the biology of the organism and in the ecosystem being considered versus changes in atmospheric inputs of these contaminants. This is particularly important given global warming and the observation that some areas of the circumpolar north are becoming warmer while other areas are becoming cooler. As a consequence geographic coverage is broad, spanning a wide range in longitude, latitude, climate, productivity, watershed, and geological features. Several biota, important in traditional diets, are included in the program including various species of fish, mammals, and birds in terrestrial, freshwater, and marine environments. Collectively the findings from these monitoring programs present a strong "weight

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of evidence” for time trends (or their lack) in a changing environment where the diet, migration patterns and harvesting pressure of individual species may be changing.

Contaminant studies have been conducted in Great Slave Lake with NCP support since 1993. At that time, the study investigated lake trout (*Salvelinus namaycush*), burbot (*Lota lota*) and lake whitefish (*Coregonus clupeaformis*) but has since been scaled back to lake trout and burbot (Evans et al 2005a). Lake trout (*Salvelinus namaycush*) is an omnivorous fish with a thermal optimum of ca. 10°C; as such it is confined to cold, deep and well-oxygenated waters during summer although it does venture into the littoral where the lake edge is steep-sided and deep (Rawson 1951; Scott and Crossman 1998). In Great Slave Lake, lake trout are captured as part of the whitefish fishery operating in the West Basin of the lake although they are not a target species. The commercial fishery does not operate in the East Arm and so lake trout do not experience the same harvesting pressures in this region of the lake. Lake trout are monitored for contaminant trends near Lutsel K’e in the oligotrophic waters of the East Arm where the majority of contaminants are believed to enter the lake from the atmosphere. They also are monitored in the West Basin which receives phosphorus-rich water from Alberta via the Slave River. The West Basin is more productive than the East Arm as a consequence of this input; its relatively shallow depth also is important (Rawson 1950; Fee et al. 1985). The Slave River, by virtue of its flow also is a significant source of contaminants to Great Slave Lake with concerns raised about contaminants from pulp and paper mills, agriculture, oil sands activities, and urban releases into the Peace and Athabasca rivers which together form the Slave River (Sanderson et al. 1997; McCarthy et al. 1997; NRBS 1996). In the West Basin, most of the lake trout monitoring has been conducted near Hay River, from the commercial fishery.

Burbot also is a predatory fish but is not a cold-water isotherm. It has a higher thermal tolerance 15.6°-18.3°C than lake trout and also resides in large northern rivers whereas lake trout is strictly a pelagic fish (Scott and Crossman 1998). In Great Slave Lake, burbot is found both in shallow

waters and deep waters down to depths of 100 m (Rawson 1950). Burbot are more commonly captured by hook and line suggesting that they are an “ambush” predator whereas lake trout are more traditional “search” predator. Lake trout, therefore, would expend more energy in seeking prey than burbot. Burbot, most likely because of their sedentary nature, are not a major component of the commercial gill net fishery. However, they are important in traditional diets, and are especially valued for their liver. About 5.6% of their body weight is liver versus 1.4% in lake trout. The liver is fat rich and can be high in persistent organic contaminants, e.g., in Lake Laberge in the 1990s (Kidd et al. 1995; Ryan et al. 2005).

While the NCP monitoring programs focus on annual temporal trend monitoring at a small number of sites, the program also allows for improved spatial coverage by monitoring other sites on a periodic basis, i.e., every ca. 5 years. For fish in the Northwest Territories, the chemical of concern is mercury. In the late 1990s and early 2000s, Lockhart conducted a series of studies determining mercury levels in lakes being assessed for their fish stocks: mercury exceeded commercial sale guidelines (0.5 µg/g) in many of these lakes (Stewart et al. 2003 a, b; Lockhart et al. 2005). These higher mercury levels were related to a combination of old fish living in small lakes (Evans et al. 2005b). The mercury investigations in these lakes form the foundations of NCP’s spatial monitoring program for mercury trends in lake trout. Colville Lake, Lake Belot, Kelly Lake, Lac Ste. Therese, Cli Lake, and Great Bear Lake have been investigated as part of the current NCP blueprint (Evans and Muir 2006, 2007, 2008) with Stark Lake and Kelly Lake sampled in 2003 (Evans 2004).

## Objectives

1. Determine temporal trends in persistent organic contaminants, mercury, and other metals in lake trout at two locations (West Basin near Hay River, East Arm at Lutsel K’e) in Great Slave through annual sampling, extending the 1993-2008 data sets to 2009 and beyond.
2. Determine temporal trends in persistent organic contaminants, mercury and other

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metals in burbot in the West Basin (offshore of Fort Resolution) extending the 1993-2008 data sets to 2009 and beyond. (Burbot monitoring was discontinued at Lutsel K'e after 2004).

3. Investigate factors affecting temporal variability in contaminants in lake trout and burbot including length, age, trophic feeding, and lipid levels. Also note potential health metrics including the appearance of skinniness, liver and gonad weight, discoloured livers, the presence of parasites, etc.
4. Continue the multi-year program based on a 5-year cycle of monitoring mercury trends in lake trout at 1 or 2 locations in the Northwest Territories. In 2008, Trout and Stark lakes were selected for monitoring.
5. Participate in and contribute information to AMAP expert work groups for trend monitoring for POPs (persistent organic pollutants) and mercury.
6. Communicate results to communities and the commercial fisheries in a timely manner and respond immediately to directed requests for information.

## Activities in 2008/2009

### *Great Slave Lake – collections and biological measurements*

In 2008-2009, 20 lake trout were collected from the Lutsel K'e area (East Basin) and northwest of Hay River (West Basin). In addition, 20 burbot were collected in the Fort Resolution area (near the Slave River inflow, West Basin). Collections were done by community members or by a commercial fisherman (Hay River). Fish were frozen and shipped whole to Environment Canada (Saskatoon) for processing. Total length, fork length (lake trout only), round weight, liver weight, gonad weight, stomach weight and gender were determined for all fish; features such as the presence of parasites, discolored liver, skinniness, and crude measures of stomach contents were noted. Aging structures (otoliths) were removed from all fish and age later determined. Approximately 100 gm of dorsal fillet, the liver and stomach were removed from all fish for analyses and/or archiving. A subsample of fillet was freeze-dried, percent moisture determined, and analyzed for carbon and nitrogen stable isotopes for all 20 fish from

each location. Ten fish from each location were selected for organic contaminant and metal analyses with organic contaminant analyses.

### *Spatial trend monitoring – collections and biological measurements*

Trout Lake and Stark Lake were selected for spatial trend monitoring of mercury in 2008/2009; these lakes were last sampled in 2003 (Evans 2003). Trout Lake was sampled as part of a Fisheries and Oceans stock assessment study of the lake with fish measurements and subsampling conducted as part of that work. Stark Lake was sampled by Lutsel K'e as part of the autumn lake trout sampling. Twenty fish were provided from each study lake.

### *Chemical analyses*

Organohalogen compounds: Organic contaminant analyses were conducted on lake trout muscle (skin on) and burbot liver. Organohalogen analyses of 2007/2008 fish are ongoing.

Mercury and other elements: Mercury and 31 other elements analyses were performed on lake trout and burbot muscle (skin off). All analyses were performed by the National Laboratory for Environmental Testing (NLET) at Environment Canada (Burlington). Analyses are complete.

## Results and Discussion

### *Great Bear Lake organic contaminant studies*

Great Bear Lake was sampled in 2007 as part of the mercury spatial trend monitoring. As reported in Evans and Muir (2008), average mercury concentrations were lower in 2007 than 2002 with fish being approximately the same mean age and length in both years (Table 1). Mid-year NCP funds were received to analyze these fish for persistent organic contaminants with these data now being reported; we previously measured organic contaminants in Great Bear Lake and Lac Ste. Therese with Northern Ecosystem Initiative funding (Evans et al. 2005a). We compare these data with lake trout data for two of our monitoring locations in Great Slave Lake .

Mean lipid levels were slightly lower in Great Bear Lake trout in 2007 than 2002 while legacy organic contaminant concentrations were substantially lower in 2007, i.e., 1.5 (CBZ) to 2.8 (DDT) times lower. Legacy contaminant concentrations were

**Table 1. Biological features, mercury and persistent organic contaminant concentrations in lake trout from Lac Ste. Therese, Great Bear Lake, and Great Slave Lake in various years.**

	L. Ste. Therese	Great Bear		Great Slave Lake 2007	
	2002	2002	2007	Hay River	Lutsel K'e
Length (mm)	708 ± 61	737 ± 123	736 ± 88.8	651 ± 70	680 ± 72
Age (yr)	24.9 ± 5.2	19.9 ± 5.1	18.9 ± 5.3	8.8 ± 2.4	16.7 ± 3.4
Lipid (%)	4.22 ± 3.16	6.28 ± 4.43	5.08 ± 2.92	9.80 ± 2.16	4.42 ± 2.22
Hg (µg/g)	0.77 ± 0.47	0.35 ± 0.31	0.28 ± 0.13	0.21 ± 0.07	0.19 ± 0.14
HCH	0.41 ± 0.74	1.13 ± 0.92	0.49 ± 0.36	0.18 ± 0.04	1.39 ± 0.68
CBz	1.67 ± 1.22	3.36 ± 2.08	2.25 ± 1.26	1.97 ± 0.64	0.37 ± 0.22
Chlordane	21.4 ± 6.68	85.5 ± 70.5	35.7 ± 28.0	3.40 ± 1.48	8.58 ± 6.04
DDT	14.0 ± 4.15	42.7 ± 36.5	15.51 ± 9.98	1.58 ± 0.75	6.11 ± 7.53
PCBs	44.9 ± 12.6	161 ± 152	65.3 ± 33.1	7.46 ± 2.27	26.5 ± 25.6
PBDE 17	n.d.	n.d.	0.01 ± 0.00	0.00 ± 0.00	0.00 ± 0.00
PBDE 28	n.d.	n.d.	0.01 ± 0.00	0.01 ± 0.00	0.01 ± 0.00
PBDE 49	n.d.	n.d.	0.02 ± 0.01	0.01 ± 0.01	0.01 ± 0.01
PBDE 47	n.d.	n.d.	0.32 ± 0.11	0.20 ± 0.10	0.23 ± 0.08
PBDE 66	n.d.	n.d.	0.02 ± 0.01	0.01 ± 0.00	0.02 ± 0.01
PBDE 100	n.d.	n.d.	0.10 ± 0.05	0.09 ± 0.04	0.11 ± 0.04
PBDE 99	n.d.	n.d.	0.16 ± 0.08	0.18 ± 0.08	0.19 ± 0.08
PBDE 85	n.d.	n.d.	0.00 ± 0.00	0.00 ± 0.00	0.00 ± 0.00
PBDE 154	n.d.	n.d.	0.05 ± 0.02	0.04 ± 0.02	0.05 ± 0.02
PBDE 153	n.d.	n.d.	0.03 ± 0.01	0.03 ± 0.01	0.03 ± 0.01
PBDE 138	n.d.	n.d.	0.00 ± 0.00	0.00 ± 0.00	0.00 ± 0.00
PBDE 183	n.d.	n.d.	0.00 ± 0.00	0.00 ± 0.00	0.00 ± 0.00
PBDE 190	n.d.	n.d.	0.00 ± 0.00	0.00 ± 0.00	0.00 ± 0.00
PBDE 209	n.d.	n.d.	0.09 ± 0.12	0.02 ± 0.04	0.07 ± 0.22
ΣPBDEs	n.d.	n.d.	0.80 ± 0.29	0.59 ± 0.26	0.72 ± 0.22

higher than in Lac Ste. Therese, a small, shallow lake southeast of Great Bear Lake where fish are similarly long-lived. Legacy organic contaminant concentrations also were higher in Great Bear Lake than in lake trout from Lutsel K'e with some of the differences associated with the slightly younger age, smaller size, and lower lipid content of East Arm lake trout. Differences may also be associated with the lower productivity of Great Bear Lake than Great Slave Lake waters (Rawson 1955; Johnson 1975 a, b); organic contaminants tend

to occur in higher concentrations on plankton (and other particulates) in lower than higher productivity waters (Larsson et al. 1998). With the exception of CBz, lowest legacy contaminant concentrations were found in Hay River trout. PCB, CBz, and HCH concentrations in 2002 were roughly similarly to those observed in Lake Laberge over 2001-2002 (Ryan et al. 2005), while DDT concentrations were lower and chlordane higher. PBDEs occurred in very low concentrations in Great Bear Lake and Great Slave Lake trout

with similar compositions and concentrations. PBDE concentrations (2007 data) were substantially higher in Lake Labarge (25.1 ng/g) and Kusawa (28.1 ng/g); Quiet Lake (0.3 ng/g) PBDE concentrations were most similar to Great Bear and Great Slave Lake concentrations (Ryan et al. 2005; Stern et al. 2008).

#### ***Mercury trend monitoring: Stark and Trout lakes***

Stark Lake is a medium-size (177 km<sup>2</sup>) lake immediately east of Lutsel K'e and on the Canadian shield; it is poorly mapped with respect to bathymetry (Falk 1979). Lutsel K'e has been concerned about the health of the fish in this lake for many years. Lake trout caught in 2003 were relatively old (mean age 22.0 yr), large (mean fork length 755 mm) with the higher mercury levels (mean 0.23 µg/g) expected of old and large lake trout (Table 2). Smaller and younger fish were collected in 2008 with, as expected, lower average mercury concentrations than in 2003. However, mercury concentrations also were lower in 2008 than 2003 when adjusted for age or length. This is in contrast to Great Slave Lake waters offshore of Lutsel K'e where length and age adjusted mercury concentrations increased between 2003 and 2008. It is not clear why mercury concentrations declined in Stark Lake fish to approach values now being observed in the Lutsel K'e area of Great Slave Lake. Possibly lake trout from Great Slave Lake moved into Stark Lake and/or the slight increase in growth rates of Stark Lake fish has resulted in mercury biodilution. Trophic feeding may also have changed.

Trout Lake is a large (520 km<sup>2</sup>) lake located west of Great Slave Lake. The lake was previously investigated by Swyripa et al. (1993) with Lockhart et al. (2005) reporting the results of various mercury analyses over 1977-1991. Lockhart et al. reported that 555-mm lake trout had an estimated mercury concentration of 0.31 µg/g in 1977, 0.32 µg/g in 1982, 0.20 µg/g in 1990, and 0.25 µg/g in 1991; it is not clear whether length is total length or fork length. In 2003, mercury concentrations in lake trout averaged 0.34 µg/g (Table 1) with a 555-mm fork length fish having an estimated concentration of 0.37 µg/g. Similar-size fish were caught in 2008 as 2003 but mercury concentrations were 1.6 times as high at 0.59 µg/g for a 555-mm (fork length) fish and 0.61 µg/g for a 600-mm fish. A similar increase in mercury concentration was noted for Kelly Lake (a 116 km<sup>2</sup> lake east of Norman Wells) with mercury concentrations increasing in 600-mm total length fish from 0.40 µg/g in 1999 to 0.71 µg/g in 2007 (Evans and Muir 2008). Mercury concentrations in 600-mm lake trout from Lac Ste. Therese, a medium size (127 km<sup>2</sup>) lake southeast of Great Bear Lake, increased from 0.66 µg/g in 2002 to 2.23 µg/g in 2007. It is not known why mercury levels have increased so dramatically in Trout, Kelly and Lac Ste. Therese but appears to be related to a combination of small lake size and a larger watershed to lake area ratio; location on Palaeozoic deposits versus the Canadian Shield also may be important.

**Table 2. Biological features and mercury concentrations in lake trout from the Lutsel K'e area of Great Slave Lake, Stark lake, and Trout Lake at different sampling times.**

	Lutsel K'e		Stark Lake		Trout Lake	
	2002	2008	2003	2008	2003	2008
Fork length (mm)	535 ± 70.2	563 ± 51.6	755 ± 92.4	612 ± 51.9	645 ± 35.5	633 ± 69.5
Age (yr)	13.0 ± 4.1	13.4 ± 2.4	22.0 ± 5.7	13.8 ± 4.3	17.2 ± 5.5	16.2 ± 7.7
Hg (µg/g)	0.13 ± 0.07	0.18 ± 0.08	0.43 ± 0.37	0.14 ± 0.06	0.34 ± 0.08	0.61 ± 0.17
Predictors						
Hg (µg/g) at 15 yr	0.14	0.18	0.23	0.15	0.33	0.55
Hg (µg/g) at 600 mm	0.14	0.20	0.29	0.16	0.35	0.61
Age at 600-mm	14.3	14.7	17.0	15.9	16.5	16.0

### Great Slave Lake – lake trout

In earlier reports (Evans and Muir 2006, 2007, and 2008) we noted that the biological features of lake trout (and burbot) populations in Great Slave Lake were exhibiting temporal variability that may be masking and/or enhancing contaminant trends. In order to obtain a better understanding of biological drivers, we have been examining temporal and spatial variability in the biological features of the fish and, in the process, making adjustments in some of our data and analyses. In 1999 and 2000, lake trout initially were aged using the scale rather than the otolith; this resulted in underestimates of age when data were compared with earlier and later fish aged using their otoliths; this was a particular problem for the Lutsel K'e fish which are relatively old and

slow growing. We have adjusted our data using otoliths to age fish from our archive collection and using length–otolith age regressions to estimate the ages of scale-aged fish. In addition, for our West Basin studies, lake trout were caught at two locations over the course of our study. Fish initially (1993) were caught in an area immediately west of Fort Resolution in December by a commercial fisherman. They were caught by Fort Resolution in summer or autumn 2000 and 2001; these latter two collections were made in the East Arm area because lake trout are uncommon in the immediate Slave River area during the ice-free season. Lake trout were collected out of the Hay River area by commercial fishermen in 1999, 2001, 2002, and annually since 2004. In previous reports, we combined all the West Basin data, noting

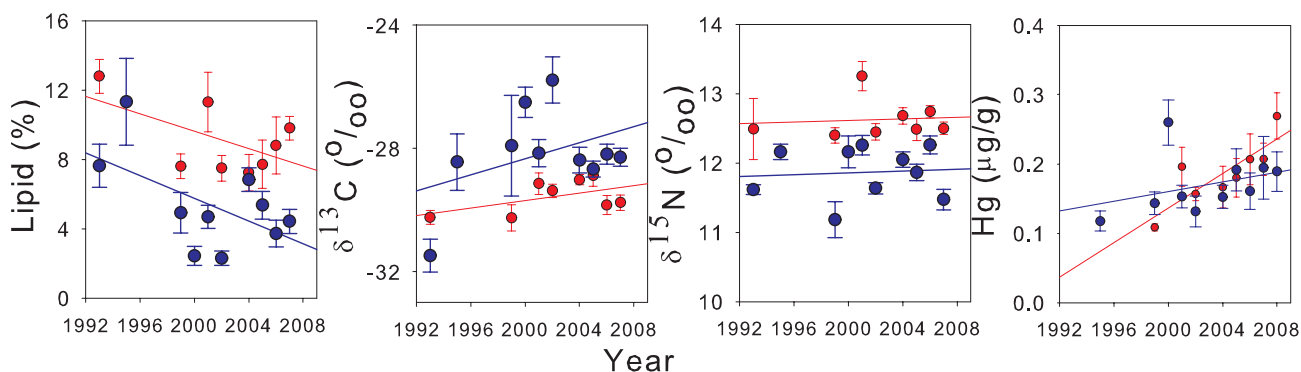


Figure 1. Long term trends (mean  $\pm$  1 standard error) in lake trout %lipid, carbon isotope, nitrogen isotope and mercury for lake trout caught from the Hay River area of the West Basin (red) and Lutsel K'e East Arm (blue) of Great Slave Lake. Also shown is the regression line. All analyses are of fillet.

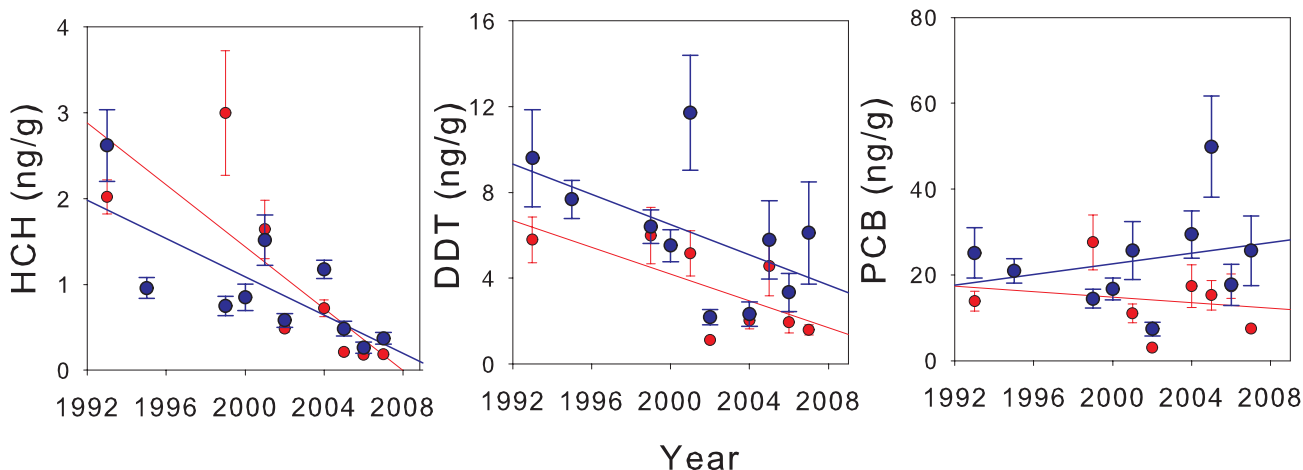


Figure 2. Long term trends (mean  $\pm$  1 standard error) in HCH, DDT, and PCBs for lake trout caught from the Hay River area of the West Basin (red) and Lutsel K'e East Arm (blue) of Great Slave Lake. Also shown is the regression line. All analyses are on fillet.



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some outliers. However, in examining the biological data more closely, we note that lake trout have different biological features between the three collection areas with Lutsel K'e being the slowest growing, lightest, and feeding to the greatest extent on littoral zone carbon and having the lowest lipid content. Hay River fish are the fastest growing, most lipid-rich, and youngest fish while fish caught in the Simpson Island area of the Fort Resolution-East Arm area show intermediate features. Therefore, we have reconstructed our West Basin data base to exclude collections made in 2000 and 2001 out of Fort Resolution and in the general area of the western end of the East Arm. Fortunately, we arranged for the collection of lake trout from both Hay River and Fort Resolution in 2001 and so only lose 2000 as a data point.

Considering first lipid content, there was been a general decline in lipids both for West Basin and East Arm lake trout with the regression lines almost parallel. A similar decline in energy content of lake trout has been reported for the Great Lakes; this decline has been related to competition for food resources with stocked salmonids (Paterson et al. 2009). Lipid content of Lake Laberge fish also has shown a temporal decline (Ryan et al. 2005) which has been related to a relaxation in the commercial fishery. It is not known to what extent commercial fishing pressures have changed on Great Slave Lake in recent years although there is anecdotal information that lake trout are becoming more prevalent in the Fort Resolution area. However, there is no commercial fishery in the main body of the East Arm and so fishing pressures are less likely to be a causal factor for the lipid trend at Lutsel K'e. Carbon isotope has increased in values in lake trout for both locations suggesting that trout are relying more on littoral zone carbon. Nitrogen isotope has shown little change with time while mercury has increased markedly with time, particularly in the West Basin. Average mercury concentrations remain below commercial sale guidelines.

HCH has been declining in concentration in lake trout in both regions of Great Slave Lake and at a more rapid rate than the decline in lipids; a similar situation occurs for DDT. Thus, while a major fraction of the decline in HCH and DDT appears to be related to changes in the lipid content of the lake trout, a smaller component of the decline

appears to be due to other factors. This will be investigated through multivariate analysis and through plotting trend data on a lipid basis. PCBs are showing weak evidence of a decline in the Hay River area and a trend of an increase at Lutsel K'e. Since lipid concentrations are declining in both locations, PCBs are increasing on a lipid-adjusted basis in both regions of Great Slave Lake.

#### ***Great Slave Lake – burbot***

Burbot were first measured for contaminants in 1993 with fish being caught at Lutsel K'e and Fort Resolution. They were sampled periodically until 1999 when the annual monitoring began and were sampled until 2003 when monitoring did not occur. Burbot monitoring resumed in 2004 at Fort Resolution and Lutsel K'e but then burbot monitoring at Lutsel K'e after 2004 because of cost considerations and the fact that fish are relatively sparse in the area. Burbot were collected again at Lutsel K'e in 2008 as part of our second Great Slave Lake project which is based on enhanced investigations of the lake. We wanted a second confirmation on trends in an area of the lake using a species that was not showing a trend of declining lipid but had been showing a trend of increasing mercury concentration in the East Arm.

Unlike lake trout, there is no evidence of a decline in lipid concentration in burbot with time although we do not have Lutsel K'e data for 2008 and fish were not studied over 2005-2007. Moreover, lake trout lipid determinations are based on fillet and burbot on liver and so we cannot be certain whether differences in lipid trends are associated with differences in the tissue analyzed or the fish species. However, we should be able to examine trends in percent moisture in the burbot fillet to estimate percent lipid; whole body percent moisture and lipids are strongly correlated with one another in lake trout (Paterson et al. 2009) and presumably similar correlations occur for lake trout fillet and fillet of other species such as burbot. Like lake trout, burbot are showing a trend of increasing carbon isotope values, suggesting a greater reliance on littoral zone carbon; carbon isotopes are heavier than for lake trout, reflecting the greater reliance on near shore carbon for burbot. There was no trend in nitrogen isotope. Mercury showed a pronounced and similar trend of increase both at Lutsel K'e and Fort Resolution.

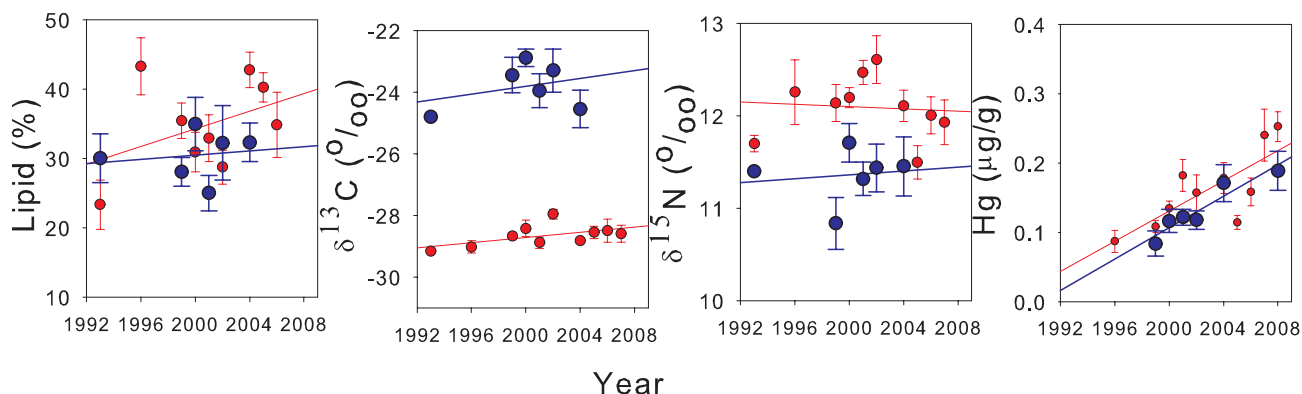


Figure 3. Long term trends (mean  $\pm$  1 standard error) in burbot %lipid, carbon isotope, nitrogen isotope and mercury for lake trout caught from the Fort Resolution area of the West Basin (red) and Lutsel K'e East Arm (blue) of Great Slave Lake. Also shown is the regression line. Lipid analyses are of the liver and all other analyses are of fillet.

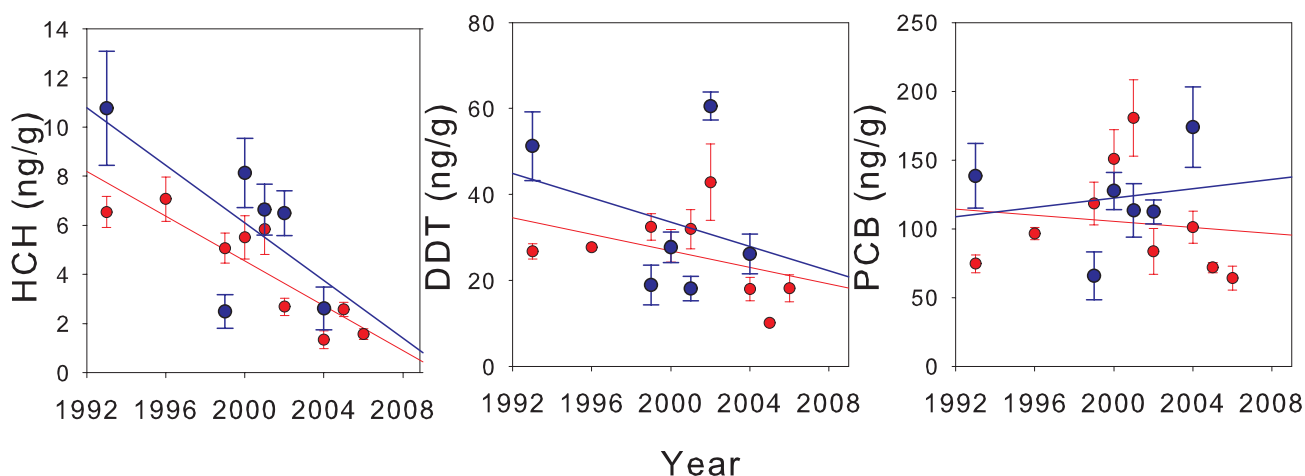


Figure 4. Long term trends (mean  $\pm$  1 standard error) in HCH, DDT, and PCBs for burbot caught from the Fort Resolution area of the West Basin (red) and Lutsel K'e East Arm (blue) of Great Slave Lake. Also shown is the regression line. All analyses are on liver.

This is remarkable given the fact that the Fort Resolution site is so strongly influenced by the Slave River. Burbot provided by Lutsel K'e are smaller than those from Fort Resolution so, on a length basis, concentrations are higher in East Arm burbot.

HCH is showing a strong trend of decline in burbot at both monitoring locations. Because percent lipid and nitrogen isotopes are not changing appreciably, this trend is highly suggestive of a decline in HCH inputs: DDT is showing a weaker trend of decline. As with lake trout, PCB is decreasing only slightly in concentration in the West Basin and showing some evidence of an increase in the East Arm.

## Community Interactions

Community interactions on this project continue to be very good. We have had an excellent long-term working relationship with Fort Resolution, Lutsel K'e and Shawn Buckley at Hay River. Charlie Catholique from Lutsel K'e attended the NCP workshop in September which provided an opportunity to discuss our work in more depth. Lutsel K'e has expressed a desire to have a workshop in the community to discuss our findings and this request has been brought to the attention of NCP. While in Fort Simpson working on a different research study (Mackenzie Gas Project) we gave a small presentation at Wood Buffalo

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National Park which including our findings on burbot trends at Fort Resolution. There have been various communications with the Northwest Territories Environmental Contaminants Committee meeting about the mercury findings and these communications will continue.

## Expected Project Completion Date

This study is expected to be completed on March 31, 2010 under the current 5-year NCP program. However, because the program is based on long-term monitoring and trends are beginning to be observed in this and other studies, it is our expectation that it will continue. The collection of such data are necessary to better understand the effectiveness of management plans implemented to reduce the inputs of contaminants into the Arctic and how other factors such as contaminant change and increasing urban and technological growth in Asia may be impacting such trends.

## Acknowledgments

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# Temporal trend studies of trace metals and halogenated organic contaminants (HOCs), including new and emerging persistent compounds, in Mackenzie River burbot, Fort Good Hope, NWT.

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## Abstract

Tissues from burbot collected at Fort Good Hope (Rampart Rapids) in December 2008 were analysed for organohalogen contaminants (OCs/PCPs/BFRs/FOCs) and heavy metals (Hg/Se/As). Data from this time point was combined with the existing metal (1985, 1989, 1993, 1995, 1999, 2000, 2001, 2002, Jan 2004 (2003), 2004, 2005, 2006, 2007) and OC (1988, 1994, 1999, 2000, 2001, 2002, Jan 2004 (2003), 2004, 2005, 2006, 2007) data covering time spans of 23 and 20 years, respectively. No significant correlation between length and mercury concentration was observed with muscle or liver for either sex. Mean Hg concentrations in muscle and liver over the entire data sets were  $0.343 \pm 0.138$  (n=385) and  $0.086 \pm 0.068$  (n=370)  $\mu\text{g g}^{-1}$ , respectively. Muscle mercury levels are below the recommended guideline level of  $0.50 \mu\text{g g}^{-1}$  for commercial sale. Major PBDE congener levels have increased significantly over the 19 year period from 1988 to 2008 but, are currently still about one order of magnitude less than those of PCBs. Since 1986, a consistent decline was observed in both PFOA and PFOS concentrations.

## Résumé

Des tissus de lotte prélevés à Fort Good Hope (rapides Rampart) en décembre 2008 ont été analysés pour déterminer s'ils contenaient des contaminants organohalogénés (pesticides organochlorés, biphenyle polychloré [BPC], produits ignifuges bromés et composés organiques fluorés) et des métaux lourds (Hg/Se/As). Les données de ce point dans le temps ont été combinées à celles qui existaient sur les métaux (1985, 1989, 1993, 1995, 1999, 2000, 2001, 2002, janv. 2004 (2003), 2004, 2005, 2006, 2007) et les composés organochlorés (1988, 1994, 1999, 2000, 2001, 2002, janv. 2004 (2003), 2004, 2005, 2006, 2007) sur une durée de 23 et de 20 ans respectivement. Aucune corrélation significative entre la durée et les concentrations de mercure n'a été observée dans les muscles et le foie des deux sexes. Les concentrations moyennes de Hg dans les muscles et le foie des ensembles complets de données étaient de  $0,343 \pm 0,138$  (n=385) et de  $0,086 \pm 0,068$  (n=370)  $\mu\text{g g}^{-1}$ , respectivement. Les niveaux de mercure dans les muscles se situent sous le niveau recommandé dans les directives, qui est de  $0,50 \mu\text{g g}^{-1}$  pour la vente commerciale. Les niveaux

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Conversely, PFDA concentrations show a consistent increase overtime. PFNA and PFUA levels peaked in 2003.

## Key Messages

- Mean Hg concentrations in muscle and liver over the entire data sets were  $0.343 \pm 0.138$  (n=385) and  $0.086 \pm 0.068$  (n=370)  $\mu\text{g g}^{-1}$ , respectively.
- Since the 1980s, a 1.6 and 2.5-fold increase in mercury concentrations has been measured in Fort Good burbot muscle and liver, respectively
- Muscle liver and mercury levels are below the recommended guideline level of  $0.50 \mu\text{g g}^{-1}$  for commercial sale.
- Significant declines, 10- and 4-fold, occurred for both  $\alpha$ - and  $\gamma$ -HCH over 20 year time period between 1988 and 2008.  $\Sigma\text{CBz}$  levels decreased by 3.0-fold over the same time period.
- Brominated flames retardant such as PBDEs have increase from 8.1 (PBDE 47) to 25.8-fold (PBDE 154) over the 20 year period from 1988 to 2008.
- Current  $\Sigma\text{PBDE}$  levels are approximately one order of magnitude less than those of PCBs.
- Since 1986, a consistent decline was observed in both PFOA and PFOS concentrations. Conversely, PFDA concentrations show a consistent increase overtime. PFNA and PFUA levels peaked in 2003.

de substances organoleptiques des principaux PBDE ont augmenté de façon significative au cours de la période de 19 ans entre 1988 et 2008, mais sont encore actuellement d'un ordre de grandeur de moins que ceux des BPC. Depuis 1986, une diminution régulière des concentrations de PFOA et de PFOS a été observée. En revanche, les concentrations de PFDA affichent une augmentation constante avec le temps. Les niveaux de PFNA et de PFUA ont été à leur plus haut en 2003.

## Messages clés

- Les concentrations moyennes de Hg dans les muscles et dans le foie de tout l'ensemble de données étaient de  $0,343 \pm 0,138$  (n=385) et de  $0,086 \pm 0,068$  (n=370)  $\mu\text{g g}^{-1}$ , respectivement.
- Depuis les années 1980, une augmentation de 1,6 et de 2,5 fois des concentrations de mercure a été mesurée dans les muscles et dans le foie, respectivement, de la lotte de Fort Good.
- Les niveaux de mercure dans les muscles et dans le foie se situent sous le niveau recommandé dans les directives, qui est de  $0,50 \mu\text{g g}^{-1}$  pour la vente commerciale.
- Des baisses importantes, de 10 et de 4 fois, des  $\alpha$ - et  $\gamma$ -HCH sont survenues au cours de la période de 20 ans entre 1988 et 2008. Les niveaux des  $\Sigma\text{CBz}$  ont diminué de 3 fois pendant cette même période.
- Les ignifugeants bromés comme les PBDE ont augmenté de 8,1 (PBDE 47) à 25,8 fois (PBDE 154) au cours de la période de 20 ans entre 1988 et 2008.
- Les niveaux actuels de  $\Sigma\text{PBDE}$  sont environ d'un ordre de grandeur moindre que ceux des BPC.
- Depuis 1986, on observe une baisse régulière des concentrations de PFOA et de PFOS. En revanche, les concentrations de PFDA affichent une augmentation constante avec le temps. Les niveaux de PFNA et de PFUA ont été à leur plus haut en 2003.

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## Objectives

To continue to assess long term trends and to maintain current data on levels of bioaccumulating

substances such as trace metals (e.g. mercury, selenium, arsenic, lead and cadmium), organo-chlorine contaminants (e.g. PCBs, DDT, toxaphene)



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and new contaminants (e.g. brominated flame retardants, fluorinated organic compounds) in Mackenzie River burbot at Rampart Rapids (Fort Good Hope).

## Introduction

With a few exceptions, minimal or no direct temporal trend information on organohalogen (OCs/PCPs/BFRs/FOCs) contaminants and heavy metals (Hg/Se/As) in fish are available in either the Arctic marine or freshwater environments. Due to a lack of retrospective samples and of past studies, much of the temporal trend data that are available are too limited to be scientifically credible because they are based on 2 or at most 3 sampling times. In addition, much of this is confounded by changes in analytical methodology as well as variability due to age/size, or dietary and population shifts. By comparison, temporal trend data for contaminants in Lake Ontario lake trout (Borgmann and Whittle 1991) and in pike muscle from Storvindeln Sweden are available over a 15 to 30 year period.

In the Mackenzie Basin over the last 150 years a steady increase in temperatures has been recorded. In particular, over the last 35 years temperatures have increased about a degree a decade, in the centre of the basin (Rouse et al., 1997). Rising temperatures in the region may be responsible for the increasing Hg levels in the FGH burbot (see Results) for several reasons: (a) melted permafrost, increased erosion and forest fires may release increasing amounts of Hg into the river; (b) the rate of Hg methylation processes may be increased by increasing temperature and nutrients, particularly in the wetlands and peatlands in the basin; and (c) possible changes in food web structure may have an effect on methylmercury (MeHg) biomagnification.

As outlined in the Northern Contaminants Program 2008-2009 call for proposals, the goal of temporal trend monitoring is to be able to detect a 10% annual change in contaminant concentration over a period of 10-15 years with a power of 80% and a confidence level of 95%. This requires sample collection and analysis of a minimum of 10 fish annually for a period of 10 to 15 years. Because of the importance of burbot to the subsistence diet of northerners residing in the Sahtu Region and because of the availability of current data sets and

archived samples (1986-2008), Fort Good Hope (and the continued analysis of burbot) was selected as one of the priority sampling location for long-term temporal trend studies.

FWI currently maintains a very extensive archive of Fort Good Hope burbot sample tissues and data on trace metals (22 years and 12 time points; 1985, 1993, 1995, 1999, 2000, 2001, 2002, Jan04 (2003), 2004, 2005, 2006, 2007) and POPs (21 years and 12 time points; 1986, 1988, 1994, 1999, 2000, 2001, 2002, Jan04 (2003), 2004, 2005, 2006, 2007).

## Activities in 2008/2009

In December 2008, 37 burbot were collected from the Mackenzie River at Fort Good Hope (Rampart Rapids) by community residents. Heavy metal and HOC analyses for these samples are now complete and the results discussed below.

## Results

### *Hg, Se, As:*

Currently heavy metal (mercury, selenium and arsenic) time trend data from Fort Good Hope (Rampart Rapids) burbot tissues cover 23 years and 13 time points (1985, 1993, 1995, 1999, 2000, 2001, 2002, Jan04 (2003), 2004, 2005, 2006, 2007 and 2008). Mean Hg concentrations in muscle and liver over the entire data sets were  $0.343 \pm 0.138$  ( $n=385$ ) and  $0.086 \pm 0.068$  ( $n=370$ )  $\mu\text{g g}^{-1}$ , respectively. Muscle mercury levels in muscle are below the recommended guideline level of  $0.50 \mu\text{g g}^{-1}$  for commercial sale.

Mean mercury, selenium and arsenic concentrations for burbot muscle and liver samples for each collection year are shown in Tables 1 and 2, respectively. No significant correlation between length and mercury concentration was observed with muscle or liver for either sex. Mercury trends and levels in male and female burbot muscle and liver follow quite closely from the early 1990's to 2008. Figure 1 shows a 1.6 and 2.5-fold increase in mercury concentrations in Fort Good burbot muscle and liver, respectively, since the 1980s. For selenium and arsenic no trends were observed in either the muscle or liver (Tables 1 and 2). The highest measured As concentration,  $17.16 \mu\text{g g}^{-1}$ , occurred in a muscle sample from a female burbot collected in 1999.

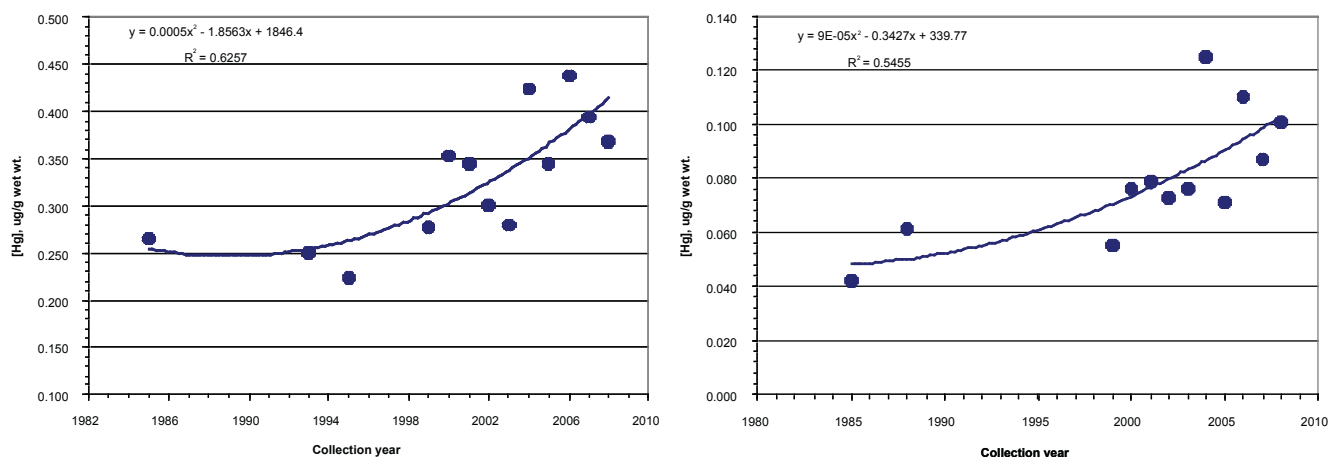


Figure 1. Mean Hg concentrations in muscle (top) and liver (bottom) from Fort Good Hope burbot (males 1 females).

**Table 1. Mean (standard deviation) concentrations of mercury, selenium and arsenic in Fort Good Hope burbot muscle ( $\mu\text{g g}^{-1}$ ).**

Collection	Sex	n	Length	Hg	Se	As
Apr-85 <sup>1</sup>	M	10	633 (84)	0.222 (0.035)	0.358 (0.087 )	—
Dec-93	M	7	677 (109)	0.231 (0.113)	0.534 (0.163)	2.291 (3.151)
Sept-95	M	2	—	0.265 (0.035)	—	—
Dec-99	M	21	676 (107)	0.286 (0.095)	0.395 (0.107)	0.637 (0.637)
Dec-00	M	21	699 (104)	0.345 (0.097)	0.478 (0.136)	1.333 (1.944)
Dec-01	M	10	720 (164)	0.342 (0.151)	0.581 (0.272)	3.106 (3.897)
Dec-02	M	12	699 (92)	0.297 (0.139)	0.427 (0.132)	1.555 (2.746)
Jan-04	M	9	705 (79)	0.336 (0.179)	0.377 (0.061)	3.324 (4.506)
Dec-04	M	17	681 (112)	0.413 (0.130)	0.523 (0.199)	1.011 (1.680)
Dec-05	M	13	616 (67)	0.301 (0.118)	0.434 (0.420)	1.663 (2.271)
Dec-06	M	17	700 (78)	0.389 (0.118)	0.401 (0.080)	0.873 (0.913)
Dec-07	M	16	642 (61)	0.420 (0.110)	0.520 (0.132)	0.522 (0.717)
Dec-08	M	15	624 (75)	0.410 (0.115)	0.506 (0.157)	0.310 (0.294)
Apr-85 <sup>1</sup>	F	6	714 (140)	0.337 (0.136)	0.480 (0.126)	—
Dec-93	F	3	812 (133)	0.297 (0.035)	0.321 (0.009)	6.450 (0.984)
Sept-95	F	2	—	0.180 (0.085)	—	—
Dec-99	F	21	735 (101)	0.259 (0.108)	0.219 (0.104) <sup>2</sup>	2.626 (3.815)
Dec-00	F	15	732 (127)	0.364 (0.140)	0.460 (0.175)	1.929 (1.621)
Dec-01	F	10	747 (122)	0.336 (0.180)	0.304 (0.096)	1.098 (1.821)
Dec-02	F	17	727 (118)	0.294 (0.126)	0.400 (.297)	2.704 (3.258)
Jan-04	F	22	726 (98)	0.254 (0.179)	0.376 (0.125)	2.827 (3.425)
Dec-04	F	18	708 (115)	0.432 (0.138)	0.451 (0.114)	1.562 (2.075)
Dec-05	F	25	710 (104)	0.350 (0.112)	0.409 (0.120)	1.587 (1.942)
Dec-06	F	21	695 (106)	0.477 (0.174)	0.435 (0.121)	0.958 (1.179)
Dec-07	F	25	671 (111)	0.376 (0.115)	0.466 (0.152)	0.533 (0.777)
Dec-08	F	22	689 (118)	0.339 (0.114)	0.433 (0.156)	0.570 (0.706)

<sup>1</sup> Wagemann 1985;

<sup>2</sup> n = 20

**Table 2. Mean (standard deviation) concentrations of mercury, selenium and arsenic in Fort Good Hope burbot liver ( $\mu\text{g g}^{-1}$ ).**

Collection	Sex	n	Length	Hg	Se	As
Apr-85 <sup>1</sup>	M	9	643 (82)	0.044 (0.019)	1.759 (0.558)	—
Dec-88	M	8	706 (84)	0.054 (0.026)	1.230 (0.555)	3.119 (1.725)
Dec-93	M	7	677 (109)	—	—	1.016 (1.328)
Dec-99	M	21	676 (107)	0.046 (0.024)	1.071 (0.628) <sup>2</sup>	0.607 (0.326)
Dec-00	M	21	699 (104)	0.064 (0.026)	1.646 (0.733)	0.585 (0.412)
Dec-01	M	10	720 (164)	0.063 (0.048)	1.434 (1.278)	0.839 (0.822)
Dec-02	M	12	699 (92)	0.063 (0.031)	1.437 (0.808)	0.771 (0.539)
Jan-04	M	9	705 (79)	0.126 (0.179)	1.981 (1.370)	1.994 (1.447)
Dec-04	M	17	681 (112)	0.111 (0.065)	3.267 (2.437)	0.496 (0.605)
Dec-05	M	13	616 (67)	0.053 (0.047)	1.677 (0.782)	0.527 (0.540)
Dec-06	M	17	700 (78)	0.094 (0.064)	1.939 (1.117)	—
Dec-07	M	16	642 (61)	0.076 (0.035)	2.090 (0.837)	—
Dec-08	M	15	324 (75)	0.114 (0.055)	3.416 (1.722)	0.335 (0.300)
Apr-85 <sup>1</sup>	F	6	714 (140)	0.097 (0.098)	1.272 (0.715)	—
Dec-88	F	2	623 (86)	0.072 (0.035)	1.460 (1.529)	1.280 (1.018)
Dec-93	F	3	812 (129)	—	—	1.062 (0.546)
Dec-99	F	20	749 (77)	0.064 (0.069)	0.687 (0.552) <sup>2</sup>	1.353 (0.811)
Dec-00	F	15	732 (127)	0.094 (0.056)	1.203 (0.469)	0.632 (0.349)
Dec-01	F	10	747 (122)	0.098 (0.108)	1.235 (0.720)	1.074 (1.227)
Dec-02	F	17	727 (118)	0.082 (0.067)	1.488 (1.203)	1.063 (0.890)
Jan-04	F	22	726 (98)	0.057 (0.033)	1.245 (0.511)	1.522 (1.348)
Dec-04	F	17	700 (112)	0.138 (0.081)	2.616 (2.030)	0.489 (0.335)
Dec-05	F	25	710 (104)	0.080 (0.050)	1.585 (1.013)	0.489 (0.585)
Dec-06	F	21	695 (106)	0.125 (0.076)	1.906 (1.006) <sup>3</sup>	—
Dec-07	F	24	674 (113)	0.094 (0.098)	2.064 (1.096)	—
Dec-08	F	22	689 (118)	0.092 (0.059)	1.690 (1.095)	0.451 (0.401)

<sup>1</sup> Wagemann 1985;

<sup>2,3</sup> n = 19

### Organohalogenes:

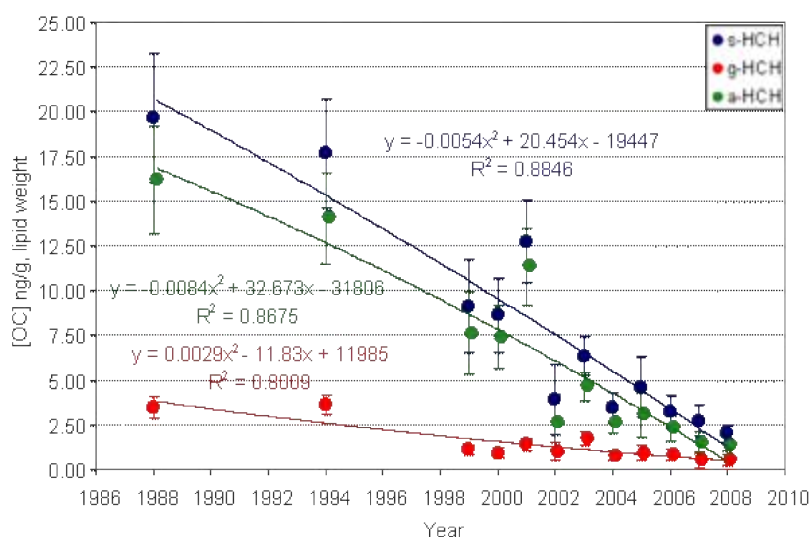
Table 3-7 list the mean wet weight of major HOC group concentration for collection periods between 1988 and 2008. After lipid normalization, significant declines, 10 - and 4-fold, occurred for both  $\alpha$ - and  $\gamma$ -HCH over this 19 year time period (Figure 2).  $\beta$ -HCH concentrations were below the detection limit in most samples.  $\Sigma\text{CBz}$  levels decreased by 3-fold.

Major PBDE congener and homologue concentrations in selected burbot liver samples are listed in Table 5 (1988 to 2008). PBDE 47 is the most predominant PBDE congener residue in the burbot liver followed by PBDE 99, 100, 153 and 154.

Results for perfluoroalkyl compounds are shown in Table 6.

**Table 3. OCs in Burbot liver from Fort Good Hope (mean and standard deviation, ng g<sup>-1</sup>, ww)**

Year	sex	n	% Lipid	ΣCBz	ΣHCH	ΣCHL	ΣDDT	ΣPCB	ΣCHB	HCBz	Oxychlor
1988	M + F	10	30.20 (13.47)	13.63 (4.21)	5.53 (1.71)	23.83 (7.37)	16.17 (5.25)	58.11 (18.45)	121.66 (38.62)	13.07 (4.06)	9.46 (1.58)
1994	M + F	9	30.56 (11.59)	8.63 (2.63)	5.13 (1.53)	17.34 (6.14)	18.96 (8.28)	50.05 (17.55)	93.70 (28.92)	8.17 (2.48)	9.23 (1.42)
1999	M + F	21	42.10 (13.31)	10.04 (3.81)	3.78 (1.38)	21.00 (8.04)	22.84 (8.59)	62.77 (22.29)	108.06 (40.74)	5.43 (2.17)	8.49 (1.70)
2000	M + F	20	36.22 (15.22)	8.72 (5.24)	3.29 (1.98)	19.02 (12.50)	21.24 (14.92)	54.62 (36.25)	94.02 (58.08)	4.78 (2.89)	8.28 (2.44)
2001	M + F	20	30.14 (15.00)	6.36 (3.06)	3.79 (1.67)	13.68 (6.99)	8.99 (5.96)	41.88 (21.26)	75.36 (48.54)	4.33 (1.90)	10.60 (2.67)
2002	M + F	12	27.33 (16.06)	4.69 (2.93)	1.40 (0.94)	17.83 (10.10)	22.18 (12.19)	37.97 (16.50)	143.61 (119.82)	4.54 (2.85)	17.64 (14.33)
Jan-04 (2003)	M + F	10	24.90 (5.77)	3.83 (3.08)	1.62 (0.57)	17.25 (18.71)	15.19 (12.72)	29.95 (21.29)	118.13 (109.79)	3.80 (3.00)	12.82 (11.27)
2004	M + F	9	24.73 (14.27)	4.05 (3.72)	0.87 (0.45)	25.35 (21.84)	35.65 (26.15)	57.62 (32.22)	201.65 (167.60)	3.90 (2.66)	24.89 (20.06)
2005	M + F	10	24.50 (12.12)	4.71 (2.14)	1.09 (0.61)	22.16 (12.40)	19.46 (9.28)	29.23 (8.49)	110.33 (67.35)	4.42 (2.01)	42.50 (23.38)
2006	M + F	10	32.74 (15.87)	3.77 (1.99)	1.00 (0.46)	21.42 (19.01)	35.53 (15.68)	61.84 (44.44)	158.00 (149.07)	3.59 (2.00)	5.25 (4.86)
2007	M + F	9	31.89 (10.25)	7.53 (3.20)	0.90 (0.44)	24.67 (11.80)	42.13 (15.73)	38.19 (17.57)	119.16 (72.60)	6.96 (3.03)	5.79 (2.78)
2008	M + F	9	39.09 (9.74)	8.83 (2.41)	1.19 (0.44)	15.26 (10.06)	38.59 (17.71)	101.86 (55.71)	289.16 (210.32)	6.52 (1.74)	3.38 (2.58)

**Figure 2. Lipid normalized ΣHCH, α-, γ-HCH concentrations in FGH burbot liver (1988–2008).**

**Table 4. Lipid normalized OCs concentrations in Burbot liver from Fort Good Hope (mean and standard deviation, ng g<sup>-1</sup>)**

Year	sex	n*	ΣCBz	ΣHCH	α-HCH	γ-HCH	ΣCHL	ΣDDT	ΣPCB	ΣCHB	Oxychlor
1988	M + F	10	48.50 (9.02)	19.67 (3.61)	16.19 (3.00)	3.48 (0.62)	84.67 (15.40)	57.34 (11.15)	206.05 (37.45)	215.97 (40.74)	9.46 (1.58)
1994	M + F	9	30.29 (8.48)	17.66 (3.03)	14.05 (2.51)	3.61 (0.52)	58.92 (11.35)	61.55 (6.42)	168.80 (22.62)	160.38 (27.00)	9.23 (1.42)
1999	M + F	21	23.55 (3.87)	9.12 (2.61)	7.64 (2.31)	1.10 (0.25)	49.18 (8.10)	53.58 (7.76)	148.85 (28.67)	126.74 (22.84)	8.49 (1.70)
2000	M + F	20	22.40 (5.31)	8.64 (2.09)	7.41 (1.79)	0.91 (0.26)	47.77 (13.83)	52.98 (16.76)	137.50 (40.16)	119.41 (32.92)	8.28 (2.44)
2001	M + F	19	21.05 (3.64)	12.76 (2.31)	11.34 (2.14)	1.42 (0.35)	44.58 (7.37)	27.52 (6.72)	138.19 (7.59)	117.55 (21.82)	10.91 (2.34)
2002	M + F	12	15.63 (11.88)	3.89 (1.96)	2.66 (1.56)	1.02 (0.48)	80.63 (64.60)	95.62 (67.66)	162.67 (107.09)	487.58 (523.81)	17.64 (14.33)
Jan-04 (2003)	M + F	10	14.59 (9.64)	6.34 (1.08)	4.63 (0.82)	1.71 (0.38)	63.29 (58.34)	57.27 (41.69)	113.99 (62.86)	446.99 (350.00)	12.82 (11.27)
2004	M + F	9	16.75 (9.42)	3.39 (0.91)	2.62 (0.66)	0.76 (0.26)	133.85 (124.50)	168.22 (103.73)	257.46 (159.14)	883.24 (823.31)	25.38 (21.39)
2005	M + F	8	18.81 (8.02)	4.63 (1.69)	3.08 (1.24)	0.94 (0.39)	83.67 (41.03)	69.12 (36.06)	103.47 (46.49)	408.43 (208.31)	42.50 (20.38)
2006	M + F	8	16.62 (4.37)	3.20 (0.95)	2.30 (0.74)	0.85 (0.29)	62.22 (54.07)	112.70 (80.80)	151.22 (105.33)	445.42 (410.90)	15.04 (13.90)
2007	M + F	9	23.56 (6.03)	2.73 (0.87)	1.53 (0.59)	0.52 (0.41)	78.62 (30.26)	143.95 (68.04)	129.20 (59.81)	363.09 (168.67)	18.40 (6.81)
2008	M + F	8	17.13 (4.37)	3.03 (0.75)	1.69 (0.38)	0.96 (0.78)	41.20 (7.52)	102.71 (52.23)	283.38 (200.27)	803.57 (648.00)	9.54 (8.29)

\*only liver samples with lipid > 10 % included.

## Expected Project Completion Data

Temporal trend studies are long-term propositions and thus annual sampling is projected into the foreseeable future.

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**Table 5. Major PBDE congener concentrations in Burbot liver from Fort Good Hope (mean and standard deviation, pg g<sup>-1</sup> ww)**

Year	Sex	n	% Lipid	PBDE 47	PBDE 99	PBDE 100	PBDE 153	PBDE 154
1988	M + F	10	30.2 (13.5)	226.3 (280.3)	84.5 (130.6)	35.2 (46.7)	29.4 (44.7)	20.5 (28.9)
1999	M + F	4	35.0 (9.6)	582.8 (522.3)	370.1 (269.6)	207.7 (154.6)	161.3 (124.8)	157.5 (116.4)
2000	M + F	11	33.3 (13.1)	620.3 (628.9)	319.7 (273.9)	180.5 (182.7)	135.2 (133.9)	81.3 (84.2)
2002	M + F	10	24.8 (14.5)	680.5 (305.4)	383.3 (258.3)	200.6 (87.1)	111.9 (74.2)	191.4 (95.7)
Jan-2004 (2003)	M + F	10	28.1* (11.5)	814.7 (618.9)	745.3 (583.2)	297.7 (190.9)	435.5 (330.1)	311.4 (216.0)
2005	M + F	10	17.3 (9.4)	718.4 (370.7)	516.0 (248.9)	210.7 (102.9)	111.7 (60.2)	170.1 (62.2)
2006	M + F	9	21.7 (16.3)	1822.9 (1913.5)	1281.5 (717.7)	1010.4 (522.9)	539.6 (359.8)	529.7 (344.7)
2007	M + F	9	23.56 (6.03)	800.4 (878.0)	709.1 (967.0)	361.8 (314.4)	86.8 (192.8)	72.7 (39.0)
2008	M + F	9	39.1 (9.74)	498.5 (228.0)	105.1 (110.7)	48.3 (62.7)	21.4 (25.6)	264.7 (64.1)

\*Some sample not the same as for OCs in Table 3

**Table 6. FOC levels in Burbot liver from Fort Good Hope (mean and standard deviation, ng g<sup>-1</sup> ww)\***

Year	Sex	n	PFOA	PFNA	PFOS <sup>a</sup>	PFDA	PFUA	Total
1986	M + F	10	4.59 (6.84)	0.89 (0.72)	10.44 (5.90)	1.91 (2.13)	2.25 (4.90)	20.08
1999	M + F	10	4.03 (6.57)	3.89 (9.29)	9.89 (10.16)	1.20 (1.73)	1.44 (2.92)	20.45
2000	M + F	10	1.58 (5.01)	0.98 (3.11)	5.62 (7.81)	2.59 (2.66)	0.75 (1.65)	11.52
2001	M + F	10	1.44 (2.62)	1.57 (3.00)	4.52 (7.75)	36.85* (94.21)	0.70 (1.71)	45.08*
Jan-2004 (2003)	M + F	10	2.03 (3.28)	7.97 (8.03)	9.88 (10.16)	4.28 (3.96)	5.31 (3.96)	29.47
2006	M + F	10	1.07 (1.10)	4.71 (4.47)	1.93 (0.78)	8.27 (9.39)	mdl	15.98
2007	M + F	9	0.44 (0.99)	1.01 (1.13)	1.39 (1.25)	7.47 (7.52)	1.36 (1.42)	11.67
2008	M + F	10	7.62 (9.70)	6.74 (7.21)	20.73 (12.21)	7.07 (6.94)	mdl	42.16

PDDoDA = mdl = 0.05; PFUA (mdl = 0.05); \*Higher value due to one sample with a measured concentration of 304.24 ng g<sup>-1</sup>. If this value is excluded then the mean value for PFDA and total FOCs for the 2001 samples are 7.15 (7.47) and 15.38 ng g<sup>-1</sup>, respectively.



# Trace metals and Organohalogen contaminants in Fish from Selected Yukon Lakes: A Temporal and Spatial Study

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## Abstract

Lake trout muscle samples collected from two Yukon Lakes, Kusawa and Laberge, were analysed for a range of organohalogen (OCs/PCBs/BFRs/FOCs) and heavy metals (Hg/Se/As) contaminants. Currently heavy metal time trend data from Laberge and Kusawa Lake trout muscle cover 15 years, 12 and 10 time points, respectively. Mean Hg levels over the entire data sets for the Laberge and Kusawa samples were  $0.48 \pm 0.22$  (n=113) and  $0.40 \pm 0.26$  (n=94)  $\mu\text{g g}^{-1}$ , respectively. In both lakes, levels are just below the recommended guideline level of  $0.50 \mu\text{g g}^{-1}$  for commercial sale. In Lake Laberge, mercury levels in the trout muscle decreased significantly after 1993 but seemed to rise again after 2002. In Kusawa Lake trout Hg levels declined after 1999 but a steady increase has been observed over the period from 2001 to 2007. In 2007, length corrected THg concentrations were highest in Lake Laberge followed by Kusawa and Fox. Only Laberge trout had a mean THg concentration higher than  $0.5 \mu\text{g/g}$ . As was observed

## Résumé

Les échantillons de muscles de touladis prélevés dans deux lacs du Yukon (Kusawa et Laberge) ont été analysés pour détecter un vaste éventail de composés organiques halogénés (CO/BPC/IB/COF) et des contaminants de métaux lourds (Hg/Se/As). Actuellement, les données sur les tendances temporelles des métaux lourds dans les muscles des touladis des lacs Laberge et Kusawa couvrent 15 ans, 12 et 10 points temporels, respectivement. Les niveaux moyens de mercure pour l'ensemble des données concernant les échantillons des lacs Laberge et Kusawa s'élevaient à  $0,48 \pm 0,22$  (n=113) et à  $0,40 \pm 0,26$  (n=94)  $\mu\text{g g}^{-1}$ , respectivement. Dans les deux lacs, les niveaux se situent juste en dessous des niveaux recommandés par les lignes directrices pour la vente commerciale de  $0,50 \mu\text{g g}^{-1}$ . Au lac Laberge, les niveaux de mercure dans les muscles des truites diminuaient considérablement après 1993 mais semblaient augmenter de nouveau après 2002. Chez les truites du lac Kusawa, les niveaux de mercure reculaient après 1999 mais une augmentation

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with the mercury, after a rapid decline, the lipid adjusted OC concentrations seem to start to increase again around 2003/04.

## Key Messages

- Currently heavy metal (mercury, selenium and arsenic) time trend data from Laberge and Kusawa Lake trout cover 15 years, 12 and 10 time points, respectively
- The mean Hg levels in the Laberge and Kusawa trout muscle samples over the entire data sets were  $0.48 \pm 0.22$  (n=113) and  $0.40 \pm 0.26$  (n=94)  $\mu\text{g g}^{-1}$ , respectively. In both lakes, levels are just below the recommended guideline level of  $0.50 \mu\text{g g}^{-1}$  for commercial sale.
- In Lake Laberge, mercury levels in the trout muscle decreased significantly after 1993 but seemed to rise again after 2002. In Kusawa Lake trout Hg levels declined after 1999 but a steady increase has been observed over the period from 2001 to 2007.
- In 2007 that length corrected THg concentrations were highest in Lake Laberge followed by Kusawa and Fox. Only Laberge trout had a mean THg concentration higher than  $0.5 \mu\text{g/g}$ .
- As was observed with the mercury, after a rapid decline, the lipid adjusted OC concentrations seem to start to increase again around 2003/04.

constante a été observée au cours de la période comprise entre 2001 et 2007. En 2007, les concentrations totales de mercure étaient les plus élevées au lac Laberge, suivi des lacs Kusawa et Fox. Seules les truites du lac Laberge présentaient une concentration moyenne de mercure total supérieure à  $0,5 \mu\text{g/g}$ . De même qu'il avait été observé avec le mercure, les concentrations de CO semblaient recommencer à augmenter vers 2003-2004 après un recul rapide.

## Messages clés

- Les données actuelles sur les tendances temporelles des métaux lourds (mercure, sélénium et arsenic) chez les touladis dans les lacs Laberge et Kusawa couvrent 15 ans, 12 et 10 points temporels, respectivement.
- Les niveaux moyens de mercure dans les échantillons de muscle des touladis des lacs Laberge et Kusawa pour l'ensemble des données s'élevaient à  $0,48 \pm 0,22$  (n=113) et à  $0,40 \pm 0,26$  (n=94)  $\mu\text{g g}^{-1}$ , respectivement. Dans les deux lacs, les niveaux se situent juste en dessous des niveaux recommandés par les lignes directrices pour la vente commerciale de  $0,50 \mu\text{g g}^{-1}$ .
- Au lac Laberge, les niveaux de mercure dans les muscles des touladis ont considérablement diminué après 1993 mais semblaient augmenter de nouveau après 2002. Au lac Kusawa, les niveaux de mercure des touladis reculaient après 1999, cependant une augmentation constante a été constatée au cours de la période comprise entre 2001 et 2007.
- En 2007, les concentrations totales de mercure corrigées selon la longueur étaient les plus élevées au lac Laberge, suivi des lacs Kusawa et Fox. Seules les truites du lac Laberge présentaient une concentration totale de mercure supérieure à  $0,5 \mu\text{g/g}$ .
- Tel qu'observé avec le mercure, les concentrations de CO rajustées selon les lipides semblaient augmenter de nouveau vers 2003-2004 après un recul rapide.

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## Objectives

The objective of this project is to maintain current data on contaminants levels in lake trout from two Yukon lakes (Laberge and Kusawa) to continue to

assess the temporal trends of bioaccumulating substances such as trace metals (e.g. mercury, selenium, arsenic), organochlorine contaminants (e.g. PCBs, DDT, toxaphene), selected current

use chemicals such as brominated flame retardants (e.g. PBDEs, HBCDD), and fluorinated organic compounds (e.g. PFOS and its precursors) so as to determine whether the levels of these contaminants in fish (health of the fish stock) and thus exposure to people who consume them are increasing or decreasing with time. These results will also help to test the effectiveness of international controls.

## Introduction

Historical studies have demonstrated that halogenated organic contaminants (HOCs) and mercury levels in top predators can vary considerably from lake to lake within a small geographic region but temporal trends of these contaminants have rarely been monitored in a sub-Arctic area for a long period of time. This study examines concentrations of a wide range of HOCs and trace metals in lake trout from two Yukon lakes (Laberge, Kusawa), over a span of 13 years (1993-2006). Recently, Ryan et al. (2005) reported that OC pesticide and PCB concentration were declining at various rates in lake trout (*Salvelinus namaycush*) in three different Yukon lakes (Laberge, Kusawa and Quiet). For example,  $\Sigma$ DDT concentrations have decreased 39%, 85% and 84% in Kusawa, Quiet and Laberge lakes respectively. Spatial variations in OC/PCB levels were quite evident as Lake Laberge trout continued to maintain the highest levels over the 10 year period from 1992 to 2003 followed by Kusawa and then Quiet. These differences were related to a variety of factors especially the species morphological characteristics such as log age, log weights and fish lipid content. A decreasing trend in Quiet and Laberge lake trout lipid content, coupled with fluctuating condition factors and increases in body masses, suggest biotic changes may be occurring within the food webs due to fish population variations related to the cessation of commercial fishing or potentially an increase in lake plankton productivity related to annual climate variation.

Because of the importance of lake trout and burbot to the subsistence diet of northerners, the need to continue to assess the effect of climate variation on fish contaminant levels, the availability of current data sets and archived samples, Lakes Laberge and Kusawa were selected as the priority Yukon sampling location for long term temporal trend studies.

## Activities in 2008/2009

INAC (Whitehorse)/DFO (Winnipeg) together maintain a very extensive archive of fish tissues and data for Hg, Se, As, and HOCs in Yukon lakes (see Tables 1-4). In 2008, 10 lake trout were collected each from Kusawa, Laberge and Teslin Lake (the selected roving lake for 2008/09).

## Results and Discussion

### Hg, Se, As:

Currently heavy metal (mercury, selenium and arsenic) time trend data from Laberge and Kusawa Lake trout cover 15 years, 12 and 10 time points, respectively (Table 1). Mean Hg concentrations in the Laberge and Kusawa muscle samples over the entire data sets were  $0.48 \pm 0.22$  ( $n=113$ ) and  $0.40 \pm 0.26$  ( $n=94$ )  $\mu\text{g g}^{-1}$ , respectively. In both lakes, levels are just below the recommended guideline level of  $0.50 \mu\text{g g}^{-1}$  for commercial sale.

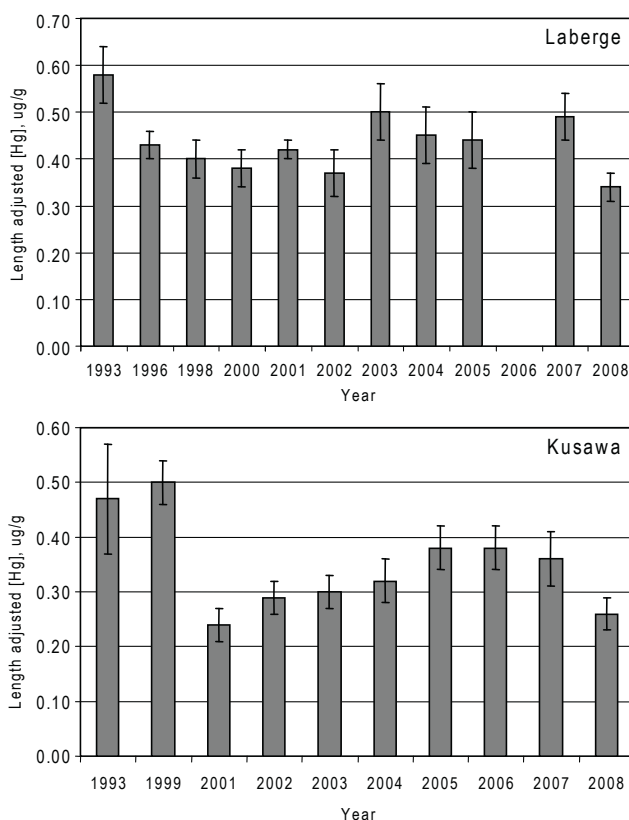


Figure 1. Length adjusted Hg concentrations in trout muscle from Lake Laberge (1993-2008) and Kusawa (1993-2008). Only Kusawa trout less than 700 mm in length were used in the ANCOVA.

**Table 1. Mean (standard deviation) concentrations of mercury, selenium and arsenic in laketrout muscle from Laberge and Kusawa Lakes. All levels are in µg/g.**

	Year	n	Length	Hg	Se	As
Laberge	1993	13	483 (110)	0.44 (0.11)	0.45 (0.08)	0.15 (0.04)
	1996	18	472 (93)	0.32 (0.10)	0.32 (0.12)	0.12 (0.06)
	1998	7	700 (125)	0.61 (0.24)	0.42 (0.07)	0.18 (0.12)
	2000	6	590 (108)	0.43 (0.21)	0.66 (0.14)	0.13 (0.04)
	2001	22	639 (92)	0.54 (0.23)	0.57 (0.13)	0.10 (0.04)
	2002	5	570 (120)	0.38 (0.15)	0.61 (0.12)	0.11 (0.05)
	2003	8	593 (98)	0.56 (0.25)	0.47 (0.10)	0.10 (0.03)
	2004	5	614 (68)	0.54 (0.23)	0.38 (0.09)	0.09 (0.04)
	2005	10	606 (97)	0.50 (0.19)	0.47 (0.09)	0.06 (0.03)
	2006	1	800	0.68	0.45	0.08
	2007	9	674 (109)	0.70 (0.27)	0.42 (0.05)	0.08 (0.03)
	2008	10	580 (78)	0.37 (0.19)	0.43 (0.07)	0.06 (0.02)
Kusawa	1993	3	535 (72)	0.54 (0.21)	0.43 (0.17)	na
	1999	14	515 (106)	0.51 (0.17)	0.46 (0.11)	0.12 (0.07)
	2001	9	551 (108)	0.29 (0.11)	0.52 (0.09)	na
	2002	10	500 (74)	0.29 (0.09)	0.55 (0.07)	0.02 (0.01)
	2003	10	487 (90)	0.35 (0.13)	0.35 (0.24)	0.03 (0.02)
	2004	9	553 (117)	0.39 (0.13)	0.64 (0.14)	0.03 (0.01)
	2005	10	510 (118)	0.43(0.31)	0.60 (0.11)	0.01 (0.01)
	2006	9	568 (168)	0.56 (0.38)	0.59 (0.17)	0.02 (0.01)
	2007	10	446 (80)	0.36 (0.24)	0.57 (0.08)	0.02 (0.01)
	2008	10	471 (94)	0.24 (0.07)	0.54 (0.08)	0.02 (0.01)
Fx	2007	10	454 (83)	0.24 (0.10)	0.34 (0.08)	0.03 (0.02)
Ts	2008	10	589 (48)	0.13 (0.06)	–	–

Fx = Fox Lake; Ts = Teslin Lake

A significant correlation between length and muscle mercury concentration was observed in the Laberge ( $[HgT] = m \cdot \text{length} + b$ ,  $m=0.0013$ ,  $b=-0.2892$ ,  $r^2 = 0.59$ ,  $p<0.001$ ,  $n=103$ ) and Kusawa ( $[Hg] = m \cdot \text{length} + b$ ,  $m=0.0018$ ,  $b=-0.5046$ ,  $r^2 = 0.52$ ,  $p<0.001$ ,  $n=84$ ) trout. ANCOVA was used to assess the effects of year to year collections (temporal trends), length and length\*year interactions (homogeneity of the slope between length and [Hg]). In Lake Laberge, mercury levels in the trout muscle decreased significantly after 1993 but seemed to rise again in 2003. Levels in 2005 are again significantly lower than those measure in the 1993 trout (Figure 1).

Only one trout was collected from Lake Laberge in 2006. In Kusuwa Lake, after a significant drop in the trout muscle mercury concentrations in 2001, levels have increased consistently but, are still below those measured in 1993 and 1999 sample (Figure 1). Figure 2 shows that in 2007 that length corrected THg concentrations were highest in Lake Laberge followed by Kusawa and Fox. Only Laberge trout had a mean THg concentration higher than 0.5 ug/g.

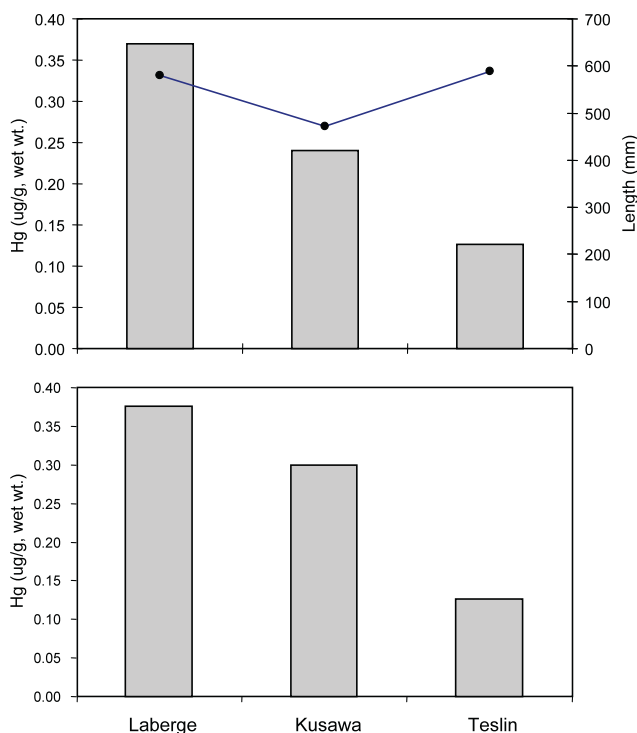


Figure 2. Wet weight HgT concentrations (top) and length corrected HgT concentrations (bottom) in 2008 lake trout muscle from Lakes Laberge, Kusawa and Teslin

#### Organohalogenes:

Tables 2 and 3 list the mean wet weight HOC concentration in trout from Lake Laberge and Kusawa Lake, respectively, over the 15 year time period from 1983 to 2008. Figure 3 show the lipid

adjusted concentration for several of the HOC groups in trout from both lakes. As was observed with the mercury, after a rapid decline, the lipid adjusted OC concentrations seem to start to increase again around 2003/04.

Major PBDE congener and  $\Sigma$ PBDE concentrations in Lake trout from Lakes Laberge, Kusawa and Quite are shown in Table 4. Levels in trout from Quite Lake are 1 to 2 orders of magnitude lower than those from Laberge and Kusawa.

FOC levels in Kusawa and Laberge lake trout liver are noted below:

#### Laberge

2006 (n=1); PFOS = 2.18 ng g<sup>-1</sup>, wet wt.

2007 (n=9); PFOS = 2.47 (1.86); PFNA = 5.78 (6.33); PFDA = 32.40 (30.34) ng g<sup>-1</sup>, wet wt.

2008 (n=10); PFOS = 1.28 (2.31); PFNA = 0.06 (0.14); PFOSA = 1.31 (1.24) ng g<sup>-1</sup>, wet wt.

#### Kusawa

2006 (n=9); PFOA = 2.93 (7.78) ng g<sup>-1</sup>, wet wt.

2007 (n=9); PFOS = 0.50 (0.54); PFNA = 0.36 (1.08); PFDA = 12.78 (16.93) ng g<sup>-1</sup>, wet wt.

2008 (n=9); PFOS = 0.44 (0.88); PFNA = 0.06 (0.14); PFDA = 0.10 (0.24); PFOSA = 0.32 (0.65), wet wt.

**Table 2. Mean (S.D.) HOC levels (ng/g wet wt.) in lake trout muscle from Lake Laberge**

Laberge	N	Age	% lipid	$\Sigma$ PCB	$\Sigma$ DDT	$\Sigma$ CHL	$\Sigma$ HCH	$\Sigma$ CHB	$\Sigma$ CBz
1993	24	15 (2)	7.9 (0.9)	328.28 (121.49)	391.54 (132.69)	47.60 (8.84)	4.69 (0.78)	310.96 (62.36)	3.92 (0.57)
1996	13	22 (5)	9.6 (1.4)	209.32 (52.08)	236.51 (41.39)	53.38 (13.74)	6.50 (1.79)	212.23 (28.31)	4.90 (1.24)
2000	6	12 (2)	3.7 (0.8)	138.95 (60.89)	96.46 (14.21)	22.36 (5.84)	2.30 (1.08)	207.33 (49.90)	2.26 (0.59)
2001	16	14 (2)	4.9 (0.5)	139.71 (53.75)	89.46 (14.04)	26.37 (5.14)	0.80 (0.07)	154.20 (60.46)	2.11 (0.17)
2002	5	12 (4)	4.2 (0.9)	48.60 (8.81)	54.50 (11.58)	7.26 (1.59)	1.58 (0.50)	139.23 (16.88)	1.15 (0.25)
2003	8	12 (1)	4.7 (0.8)	81.01 (29.83)	61.48 (8.55)	7.44 (2.24)	0.54 (0.10)	179.31 (42.79)	1.21 (0.28)
2004	6	12 (4)	8.7 (3.9)	48.93 (34.30)	94.09 (60.68)	7.46 (4.90)	0.19 (0.09)	79.92 (52.01)	0.49 (0.28)
2005	10	14 (7)	2.0 (1.22)	28.94 (20.27)	50.91 (30.27)	2.61 (1.28)	0.16 (0.10)	34.50 (19.97)	0.35 (0.27)
2006	1	21	1.0	25.52	31.25	4.82	0.07	76.87	0.35
2007	9	14 (5)	1.2 (0.80)	37.36 (25.89)	43.98 (29.93)	5.32 (4.05)	0.10 (0.09)	25.78 (14.58)	0.27 (0.80)
2008	10		2.3 (1.1)	50.23 (36.89)	70.06 (41.29)	4.04 (2.88)	0.18 (0.08)	24.48 (16.85)	0.77 (0.23)

**Table 3. Mean (S.D.) OC levels (ng/g wet wt.) in lake trout muscle from Kusawa Lake**

Kusawa	N	Age	% lipid	ΣPCB	ΣDDT	ΣCHL	ΣHCH	ΣCHB	ΣCBz
1993	10	19 (2)	1.8 (1.6)	85.62 (26.07)	44.16 (21.50)	17.33 (2.78)	1.21 (0.36)	120.80 (24.94)	1.15 (0.28)
1999	14	18 (1)	4.6 (3.0)	91.09 (11.85)	139.16 (19.72)	17.82 (2.74)	1.68 (0.23)	148.38 (29.29)	1.52 (0.20)
2001	9	12 (1)	2.4 (1.4)	48.55 (7.91)	56.58 (15.30)	7.45 (2.35)	0.91 (0.14)	61.03 (8.55)	0.84 (0.14)
2002	10	12 (1)	1.4 (0.8)	32.45 (3.66)	26.66 (4.15)	3.01 (0.48)	0.62 (0.08)	43.47 (5.02)	0.61 (0.09)
2003	9	9 (3)	5.8 (3.6)	8.16 (5.86)	8.21 (15.67)	3.50 (2.28)	0.14 (0.08)	45.05 (32.20)	0.44 (0.30)
2004	9	13 (4)	7.9 (4.7)	11.29 (3.78)	5.70 (3.70)	4.52 (2.16)	0.15 (0.07)	49.73 (30.17)	0.50 (0.27)
2005	10	15 (6)	0.61 (0.51)	5.48 (4.84)	2.35 (3.02)	1.17 (0.88)	0.03 (0.03)	12.37 (11.57)	0.12 (0.10)
2006	9	12 (4)	1.82 (1.49)	6.28 (4.58)	2.97 (2.57)	2.49 (1.84)	0.09 (0.06)	42.63 (34.97)	0.47 (0.26)
2007	9	10 (4)	1.52 (1.43)	9.88 (9.93)	2.35 (1.88)	2.78 (2.90)	0.10 (0.06)	22.44 (23.88)	0.42 (0.33)
2008	10		1.16 (0.42)	18.30 (27.27)	2.35 (0.94)	1.30 (0.40)	0.13 (0.26)	22.55 (7.87)	0.47 (0.13)

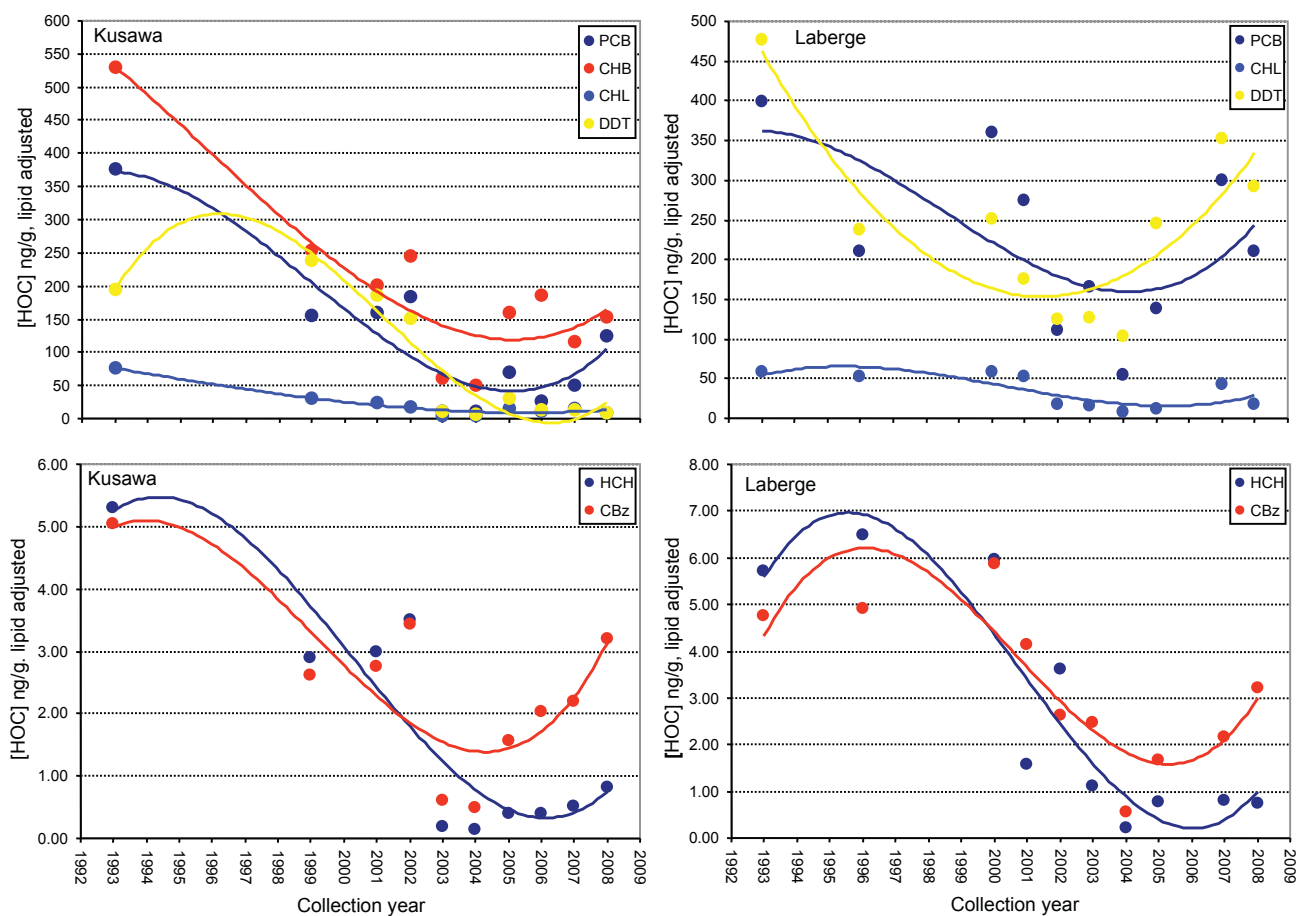


Figure 3. Lipid adjusted OC group concentrations in trout muscle from Kusawa and Laberge (1992-2008).



**Table 4. Mean (S.D.) PBDE levels (pg g<sup>-1</sup>, wet wt.) in lake trout muscle from Lakes Laberge, Kusawa and Quiet Lakes**

	Laberge	n	% Lipid	BDE 47	BDE 49	BDE 99	BDE 100	BDE 153	BDE 154
Laberge	1993	10	1.99 (1.69)	1481 (728)	348 (112)	2943 (1531)	700 (341)	642 (491)	1530 (1009)
	2000	6	0.45 (0.25)	4900 (1680)	2100 (240)	8590 (1170)	3380 (630)	5740 (1320)	4460 (1190)
	2003	8	0.41 (0.19)	3170 (1430)	1290 (750)	5890 (2860)	2450 (1200)	3920(4050)	3200 (2810)
	2005	10	2.04 (1.22)	2659 (1977)	165 (117)	4093 (2389)	1848 (1235)	740 (580)	986 (732)
	2006	1	1.01	24920	1630	35900	11370	4120	3240
	2007	9	1.20 (0.79)	5500 (901)	1100 (1130)	9680 (1627)	6700 (5850)	200 (560)	1900 (1450)
	2008	10	2.3 (1.1)	2389 (1207)	2175 (1476)	1721 (1222)	139 (99)	258 (152)	560 (211)
Kusawa	1999	10	3.01 (2.20)	4377 (2490)	nd	3636 (2011)	2573 (1623)	894 (622)	1495 (895)
	2001	10	2.78 (1.63)	700 (990)	130 (160)	720 (1090)	250 (250)	260 (480)	230 (330)
	2003	5	0.16 (1.12)	960 (1220)	360 (47)	2630 (3510)	950 (1260)	1180 (1590)	870 (1150)
	2006	9	1.82 (1.49)	1103 (1231)	66 (99)	824 (911)	446 (514)	136 (140)	202 (236)
	2007	9	1.60 (1.41)	9900 (1216)	300 (700)	12300 (1271)	3900 (5990)	1100 (830)	600 (790)
	2008	10	1.16 (0.42)	4178 (1781)	6475 (2398)	1653 (1394)	294 (113)	58 (103)	1653 (609)
Quiet	1992	6	2.52 (0.71)	25 (20)	nd	18 (31)	3 (5)	6 (6)	11 (14)
	2001	5	0.94 (0.94)	28 (40)	nd	19 (32)	8 (10)	11 (7)	12 (14)
	2003	5	0.12 (0.09)	51 (106)	7 (4)	127 (273)	18 (41)	32 (71)	17 (35)

nd = non detect; Laberge 2004 and Kusawa 2004, 2005 samples are currently be re-analyzed. Results will be available by mid-July 2007.

## Expected Project Completion Date

Temporal trend studies are long-term propositions and thus annual sampling is projected until well into the future.

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# Arctic Caribou and Moose Contaminant Monitoring Program

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## **Abstract**

Moose and caribou provide an important food resource for Northerners across the Arctic, and have been designated in the NCP blueprint as key species for monitoring contaminants in the terrestrial Arctic ecosystem. Two barren-ground caribou herds, one from the eastern and one from the western Arctic, have been designated for annual sampling, and five additional caribou herds and two moose populations have been designated for sampling every five years. In 2008/9 tissue samples were collected from Yukon moose and the Porcupine and Qamanirjuaq caribou. Analysis of samples collected in 2005-8 showed that renal copper levels are declining in the Bluenose East caribou. This could potentially affect the health of the animals if they become severely copper deficient. Renal lead concentrations are increasing in the Bathurst caribou herd. It is unclear why this is happening, so further research is being conducted. Renal mercury concentrations are increasing in female Porcupine and Bathurst caribou and to a lesser extent in male Porcupine caribou. Female caribou are considered to be more sensitive indicators of changes in available environmental mercury. None of the elements measured were present at levels that would be expected to have toxic effects in caribou.

## **Résumé**

L'orignal et le caribou représentent une ressource alimentaire importante pour les habitants de l'Arctique et ont été désignés dans le plan directeur du Programme de lutte contre les contaminants dans le Nord (PLCN) comme espèces clés pour la surveillance des contaminants dans l'écosystème arctique terrestre. Deux troupeaux de caribous de la toundra, l'un de l'Est de l'Arctique et l'autre de l'Ouest, ont été désignés pour un échantillonnage annuel et cinq autres troupeaux de caribous et deux populations d'originaux ont été désignés pour un échantillonnage aux cinq ans. En 2008-2009, des tissus ont été prélevés chez l'orignal du Yukon et chez les caribous de la Porcupine et de Qamanirjuaq. L'analyse des tissus prélevés en 2005-2008 indique que les niveaux de cuivre dans les reins ont baissé chez le caribou de Bluenose-Est. Une carence grave en cuivre risquerait de nuire à la santé des animaux. Les concentrations de plomb dans les reins augmentent dans le troupeau de caribous de Bathurst. Nous ne savons pas pourquoi, ce qui fait que d'autres recherches sont menées. Les concentrations de mercure dans les reins augmentent chez les caribous femelles de la Porcupine et de Bathurst et, dans une mesure moindre, chez les caribous mâles de la Porcupine. La femelle est considérée comme un indicateur plus sensible des changements dans le

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mercure disponible dans l'environnement. Aucun des éléments mesurés n'était présent dans des concentrations qui pourraient avoir des effets toxiques chez le caribou.

## Key Messages

- Copper levels are declining in the Bluenose East caribou herd which could affect the health of the caribou.
- Lead levels are increasing in the Bathurst caribou although it is unclear why. Ongoing studies will try to determine why this trend is occurring.
- Mercury concentrations are changing over time in some herds, and increasing in female Porcupine and Bathurst caribou. Ongoing studies will help to determine why these trends are occurring.
- None of the elements measured were present at levels that would be expected to have toxic effects in caribou.

## Messages clés

- Les niveaux de cuivre sont à la baisse dans le troupeau de caribous de Bluenose-Est, ce qui pourrait nuire à la santé de ces animaux.
- Les niveaux de plomb augmentent chez le caribou de Bathurst sans que l'on sache trop pourquoi. Des études en cours essaieront de déterminer les causes de cette tendance.
- Les concentrations de mercure se modifient avec le temps dans certains troupeaux, et augmentent chez les caribous femelles de la Porcupine et de Bathurst. Des études en cours essaieront de déterminer les causes de ces tendances.
- Aucun des éléments mesurés n'étaient présents dans des concentrations qui pourraient avoir des effets toxiques chez le caribou.

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## Objectives

To determine levels of and temporal trends in contaminants in Arctic caribou and moose in order to:

- Provide information to Northerners regarding contaminants in these traditional foods, so that:
  - ◆ They may be better able to make informed choices about food consumption. This includes providing information for health assessments and/or advisories as required.
  - ◆ Wildlife managers can assess possible health effects of contaminants on Arctic moose and caribou populations.
- Further understand the fate and effects of contaminant deposition and transport to the Canadian Arctic.

species for monitoring contaminants in the terrestrial Arctic ecosystem. Two barren-ground caribou herds, one from the eastern Arctic and one from the western Arctic, have been designated for annual sampling, and five additional caribou herds and two moose populations have been designated for sampling every five years.

## Activities in 2008/2009

Tissue samples were collected from the Porcupine and Qamanirjuaq caribou herds and moose from the Yukon. Sampling information was also collected for each animal, including gender, date and location of collection.

Yukon moose and Porcupine caribou were sampled by local hunters as part of the ongoing Yukon Hunter Survey Program. Samples from the Qamanirjuaq caribou herd were taken by a local hunter under the supervision of the local regional biologist.

All samples were prepared for analysis in Whitehorse, YT by the program coordinator and analyzed at the National Laboratory for

## Introduction

Moose and caribou provide an important food resource for Northerners across the Arctic, and have been designated in the NCP blueprint as key

Environmental Testing (Environment Canada) using the inductively coupled plasma technique with mass spectroscopy, and for total mercury using cold vapour atomic absorption spectroscopy, under the supervision of Dr. Derek Muir. Remaining liver and muscle samples were archived at  $-50^{\circ}\text{C}$  at the INAC facility in Whitehorse, YT. Moose and caribou teeth were aged by the project leader and a Yukon Environment technician in Whitehorse using the tooth cementum technique.

## Results and Discussion

Laboratory analyses of samples collected from 2008/9 are expected to be available by fall, 2009. Results for samples collected 2005-08 are presented here (Table 1). Although 31 elements were measured, only results for 7 elements of concern were analyzed in detail (arsenic, cadmium, copper, lead, mercury, selenium and zinc). Where possible, data collected from previous years (under this program and by GNWT) were used along with the current data to explore temporal trends. Renal cadmium, mercury, selenium and zinc increased with age in at least some caribou herds, so age was used as a covariable in analyses. Season of collection also affected some element concentrations;

mercury was higher in spring-collected animals while copper was higher in fall-collected animals. It is important to take this into account when comparing results among herds.

Copper, selenium and zinc are required in trace amounts for the normal functioning of the animal and although selenium and zinc were found in adequate concentrations in all caribou herds measured, copper levels are considered marginal. There is no evidence that any of these elements reached toxic levels in any of the animals measured. The toxic elements (arsenic, cadmium, lead and mercury) were found in measurable amounts, but never higher than is considered 'normal to high' for domestic cattle (Puls 1994). None of these elements approached levels that would be expected to cause toxic effects in the caribou.

Renal cadmium concentrations in caribou showed a general geographical trend, increasing from east to west (Figure 1). Terrestrial lichens from the Yukon averaged 0.08 mg/g (dry weight) (unpublished data) as compared with an average of 0.171 mg/g (dry weight) in northern Quebec (Crête et al. 1992). This suggests that the long-range transport of cadmium that is then absorbed by Arctic lichens

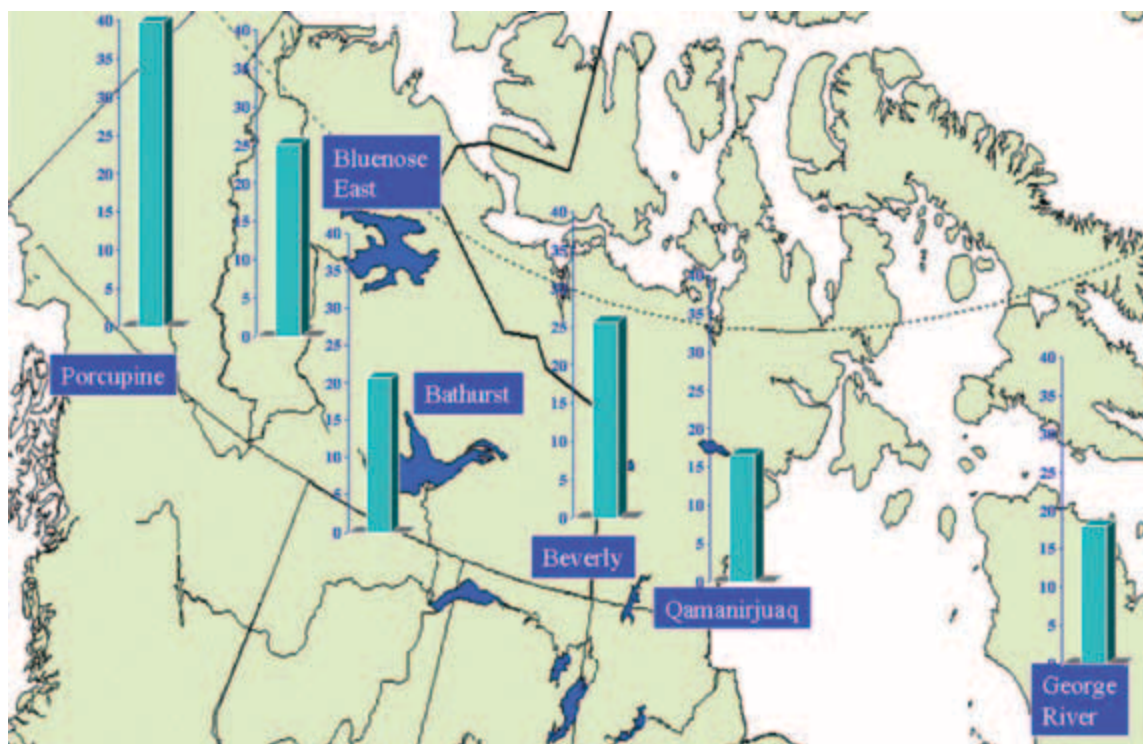


Figure 1. Average renal cadmium concentrations ( $\mu\text{g/g}$  dry weight) in 4-6 year old caribou.

**Table 1. Renal element concentrations (µg/g dry weight) in caribou herds collected in 2005-8 (Mean ± standard deviation).**

Herd	Season	Sex	Year	N	Age	Arsenic	Cadmium	Copper	Lead	Mercury	Selenium	Zinc
Bathurst	Fall/ Winter	F	2006	25	N/A	0.22 ± 0.08	29.3 ± 18.1	16.2 ± 3.6	0.29 ± 0.11	5.30 ± 1.82	3.63 ± 0.89	86.7 ± 18.6
			2007	5	3.0	0.12 ± 0.01	20.5 ± 19.4	24.0 ± 0.8	0.16 ± 0.08	1.40 ± 0.49	3.52 ± 0.44	111.7 ± 6.6
		M	2005	22	N/A	0.08 ± 0.03	18.4 ± 8.8	21.6 ± 3.1	0.14 ± 0.14	1.77 ± 0.55	3.47 ± 0.51	103.0 ± 21.8
			2007	11	2.7	0.14 ± 0.06	19.4 ± 25.9	24.6 ± 4.3	0.16 ± 0.05	1.41 ± 0.97	3.64 ± 0.67	114.1 ± 15.8
Beverly	Spring/ Summer	F	2008	23	N/A	0.08 ± 0.04	38.1 ± 17.9	21.8 ± 1.7	0.26 ± 0.13	4.86 ± 1.12	4.33 ± 0.45	113.5 ± 9.1
			2008	1	N/A	0.07	20.1 ±	22.1	0.33	4.53	4.16	103.4
		M	2008	10	6.3	0.06 ± 0.02	34.3 ± 23.1	20.5 ± 2.9	0.31 ± 0.08	5.71 ± 1.46	4.34 ± 0.52	105.1 ± 8.8
			2008	1	4.0	0.10	30.8	20.3	0.28	7.22	4.53	105.5
Bluenose East	Spring/ Summer	F	2005	5	6.8	0.03 ± 0.01	46.7 ± 32.5	18.0 ± 1.4	0.23 ± 0.06	4.49 ± 2.02	3.52 ± 0.42	97.5 ± 8.0
			2006	15	4.6	0.04 ± 0.01	22.9 ± 10.7	17.4 ± 2.2	0.40 ± 0.78	4.67 ± 0.97	4.15 ± 1.03	101.5 ± 24.2
		M	2008	15	4.9	0.03 ± 0.01	16.8 ± 9.7	22.1 ± 1.8	0.47 ± 0.13	5.15 ± 1.31	5.30 ± 0.56	104.1 ± 7.4
			2008	6	2.7	0.03 ± 0.01	10.7 ± 4.2	23.4 ± 3.7	0.37 ± 0.08	4.40 ± 0.82	4.60 ± 0.39	106.9 ± 5.8
Porcupine	Fall/ Winter	F	2005	4	7.3	0.05 ± 0.03	56.4 ± 51.1	22.5 ± 2.0	0.19 ± 0.06	3.28 ± 1.17	4.93 ± 0.47	127.4 ± 23.9
			2006	3	4.0	0.07 ± 0.00	41.4	22.9	0.14 ± 0.06	3.21 ± 1.72	4.16 ± 0.35	129.0 ± 13.8
		M	2007	1	7.0	0.04	24.2	17.6	0.11	1.25	3.97	103.5
			2005	14	3.8	0.05 ± 0.04	23.1 ± 14.8	23.1 ± 2.4	0.15 ± 0.04	1.81 ± 0.33	4.48 ± 0.63	121.9 ± 18.0
Qaminuriaq	Fall/ Winter	F	2006	7	7.3	0.03 ± 0.02	18.7 ± 13.9	26.3 ± 2.0	0.58 ± 0.81	3.37 ± 0.96	3.58 ± 0.54	104.1 ± 8.5
			2007	10	5.1	0.04 ± 0.01	24.0 ± 15.7	25.1 ± 8.9	0.44 ± 0.15	5.57 ± 2.33	4.09 ± 1.37	110.1 ± 30.5
		M	2006	14	5.8	0.01 ± 0.01	14.0 ± 8.9	25.8 ± 1.8	0.34 ± 0.24	2.58 ± 0.86	3.60 ± 0.46	112.3 ± 14.0
			2007	8	4.0	0.03 ± 0.01	11.5 ± 8.2	20.8 ± 2.5	0.39 ± 0.21	4.23 ± 1.62	3.62 ± 0.58	94.2 ± 10.3

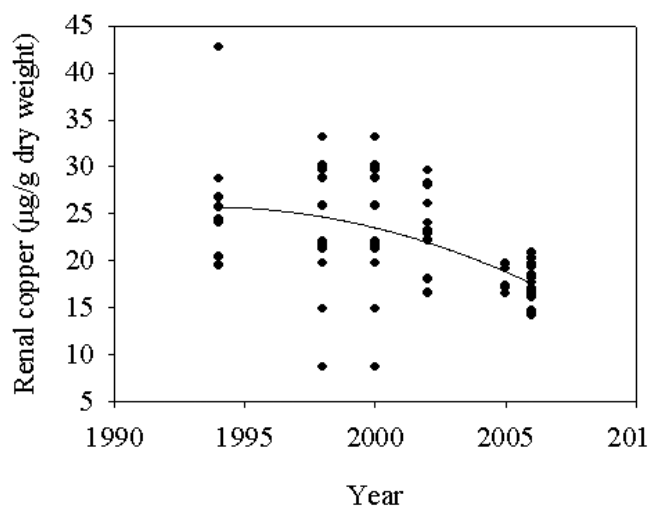


Figure 2. Renal copper concentrations in the Bluenose East caribou herd.

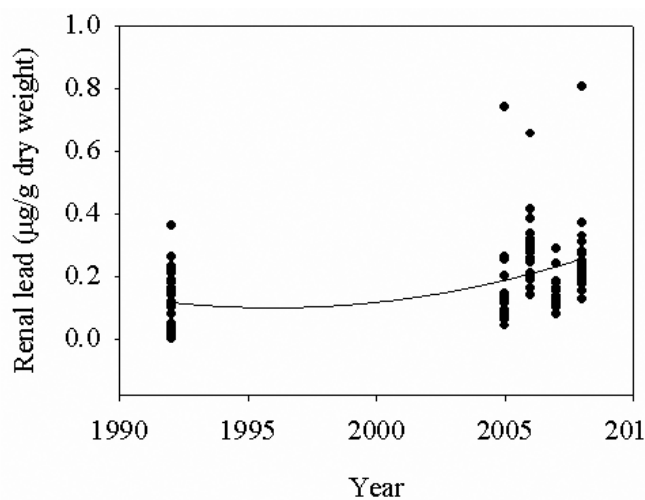


Figure 3. Renal lead concentrations in the Bathurst caribou herd.

and consumed by caribou is not the cause of this trend. It likely reflects higher levels of naturally occurring cadmium in the soil in the west, which is taken up by cadmium hyperaccumulators like willows and passed on to caribou foraging on the willows.

Although most caribou herds measured had low levels of copper when compared with the level of 10-25 ppm (wet weight) that was considered 'marginal' for moose (Flynn and Franzman 1987), the combination of low and declining copper concentrations in the Bluenose East herd (Figure 2) may be a particular concern for the health of the animals in this herd. The average concentration of copper in the Bluenose East herd (4.4 ppm wet weight) is only slightly higher than the 3.72 ppm (wet weight) thought indicative of copper deficiency causing death in moose from Alaska (O'Hara et al. 2001). Copper deficiency can cause reduced fertility and low conception and ovulation rates in cattle (Puls 1994), so even if concentrations are not low enough to cause mortality, they may be low enough to be having an effect on reproduction and therefore population growth/decline. Further monitoring of this herd is recommended.

Renal lead concentrations were generally low, but are increasing over time in the Bathurst herd (Figure 3). It is not clear why this increase is occurring, particularly since it is unique to this herd. Caribou forage samples from the Bathurst

range will be collected in the summer of 2009 and analyzed for lead to try to determine the origin of this contaminant.

Analyzing temporal trends using the current data is difficult for many of the caribou herds because annual sampling has not often been conducted. The Porcupine caribou herd has been sampled annually since 1994, making it the ideal candidate for temporal trend analysis. Renal mercury concentrations are increasing significantly over time in female Porcupine (both spring and fall-collected) and although concentrations in male Porcupine caribou are also increasing over time (Figure 4), the relationship is not quite significant ( $p=0.055$ ).

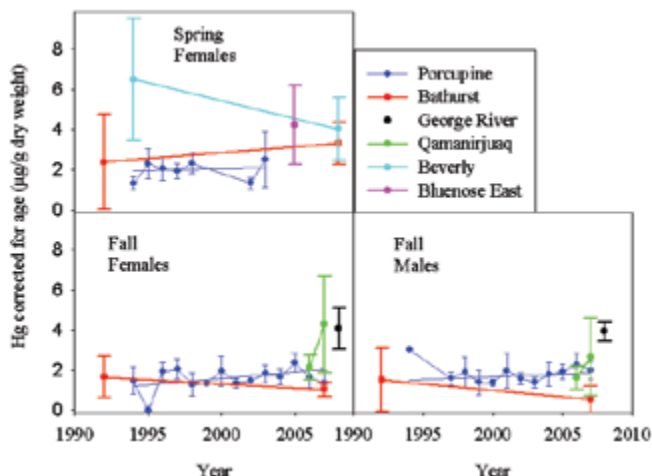


Figure 4. Temporal trend of age-corrected renal Hg in fall-collected Porcupine caribou.



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A recent NCP study on mercury in caribou forage concluded that female Porcupine caribou consume proportionally more mercury than males simply because they consume proportionally more forage. This makes them more sensitive environmental indicators of changes in available environmental mercury than male caribou.

Renal mercury concentrations in spring-collected female Bathurst caribou are also increasing over time, but are decreasing in fall-collected males and females, and in all animals in the Beverly herd. Although these are statistically significant relationships ( $p < 0.05$ ), they are driven largely by relatively old data ( $> 15$  years) with low sample numbers and the comparisons may be confounded by differences in tissue preparation or analysis. Continuing monitoring of these herds using standardized techniques should give a more reliable picture of temporal trends. The Qamanirjuaq caribou herd also shows an increase in renal mercury for males and females (Figure 4) but it is over only two years. Again, continuing to monitor this herd on an annual basis will indicate whether this is a true temporal increase or simply annual variation. Although average renal mercury was slightly lower in George River caribou collected in 2007 than those collected in 1994-96 (0.56 ppm wet weight; [Robillard et al. 2002] as compared with 0.49 ppm wet weight from this study), since age corrections were unable to be applied to the earlier data, a valid comparison is not possible.

## Project Completion Date

This project is ongoing.

## Acknowledgements

Many thanks to Alpine Aviation, Mitch Campbell, Frank Nutarasungnik and Yukon Environment staff, especially Tena Fox, Krista Funk and Angela Milani. I would like to acknowledge the efforts of all hunters who submitted samples to this program – without them, this work would not be possible. This project was funded by the Northern Contaminants Program, Department of Northern Affairs and administered by the Yukon Conservation Society.

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# Source apportionment of mercury in Davis Strait beluga

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## **Abstract**

It is well established that current mercury (Hg) levels in some traditional Arctic country foods such as seals and beluga are very high in terms of human health exposure. But how much higher are current levels in country foods compared to the 19<sup>th</sup> Century or earlier, before pollution was wide-spread? To answer this question for Davis Strait beluga (*Delphinapterus leucas*), this project determined the long-term Hg trend in the beluga, using an established approach involving historical era and modern teeth originally collected along the West Greenland coastline and stored in Copenhagen Zoological Museum (1854), National Environmental research Institute (1985) and Greenland Institute of Natural Resources (1993 and 2000). Modern teeth included samples collected in 1985, 1993 and 2000 while historical samples came mainly from 1854 but included individual beluga harvested up to 1905.

Tooth Hg was significantly correlated with animal age in all time periods. But its accumulation rate with age was 2.3- to 3.3-times more rapid in modern beluga than in historical era teeth. There was some year-to-year variation in the modern tooth Hg-age

## **Résumé**

Il est bien établi que les niveaux de mercure (Hg) actuels dans certains aliments traditionnels de l'Arctique, tels que les phoques et les bélugas, sont très élevés pour ce qui est des dangers qu'ils présentent pour la santé humaine. Mais dans quelle proportion les niveaux actuels dans ces aliments sont-ils supérieurs à ceux du 19<sup>e</sup> siècle ou avant, soit avant que la pollution ne devienne largement répandue ? Pour répondre à cette question pour le béluga du détroit de Davis (*Delphinapterus leucas*), il fallait déterminer la tendance à long terme des concentrations de mercure chez le béluga, à l'aide d'une méthode établie fondée sur des dents de la période historique et des dents de la période moderne initialement ramassées le long de la côte Ouest du Groenland et conservées au Musée zoologique de Copenhague (1854), au National Environmental Research Institute (1985) et au Greenland Institute of Natural Resources (1993 et 2000). Les dents de la période moderne comprenaient des échantillons ramassés en 1985, en 1993 et en 2000 tandis que les échantillons historiques dataient principalement de 1854 mais comprenaient également des échantillons de bélugas individuels recueillis jusqu'en 1905.

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regressions, but these fluctuations were minor compared to the long-term increase. The calculated anthropogenic contribution to tooth Hg in modern 40 yr old beluga was 85%, assuming the 19<sup>th</sup> Century data approximate the natural background level. To more confidentially interpret the Hg data, we also measured the stable carbon and nitrogen isotopic composition of the teeth, to infer the animals' average lifetime dietary habits (trophic level and feeding location). Although some significant isotopic changes occurred in different years, none of the changes affected the overall conclusion that anthropogenic inputs to the Arctic during the 20<sup>th</sup> Century significantly increased the body burden of Hg in modern beluga in Davis Strait.

Les concentrations de mercure dans les dents présentaient une forte corrélation avec l'âge quelle que soit la période. Cependant, le taux d'accumulation avec l'âge était de 2,3 à 3,3 fois plus rapide chez les bélugas modernes. Certaines variations annuelles ont été constatées dans les régressions mercure-âge dans les dents de la période moderne, toutefois ces variations étaient mineures comparativement aux augmentations à long terme. La contribution anthropogénique aux concentrations de mercure calculée pour les dents des bélugas modernes âgés de 40 ans était de 85 %, en supposant que les données du 19<sup>e</sup> siècle s'approchaient du niveau de référence naturel. Afin de mieux interpréter les données concernant le mercure, nous avons également mesuré la composition de carbone stable et la composition isotopique de l'azote de la dent afin de déduire les habitudes alimentaires au cours de la durée de vie moyenne de l'animal (niveau trophique et lieu d'alimentation). Malgré d'importants changements isotopiques constatés au cours des différentes années, aucun de ces changements ne modifiait la conclusion générale que les intrants anthropogéniques dans l'Arctique au 20<sup>e</sup> siècle augmentent considérablement les concentrations de mercure dans le corps des bélugas de la période moderne dans le détroit de Davis.

## Key Messages

- Mercury levels in modern Davis Strait beluga accumulate at significantly faster rates with age than during the mid-19<sup>th</sup> and early 20<sup>th</sup> Centuries, according to analyses of their teeth.
- This increase in accumulation rate is not due to changes in beluga feeding habits over time; the most likely explanation is that it reflects the effect of anthropogenic pollution over the last century.

## Messages clés

- Les analyses des dents montrent que les niveaux de mercure chez les bélugas du détroit de Davis de la période moderne s'accumulent beaucoup plus rapidement qu'au milieu du 19<sup>e</sup> et au début du 20<sup>e</sup> siècle.
- Cette augmentation du taux d'accumulation n'est pas attribuable à des changements des habitudes alimentaires des bélugas au cours du temps; l'explication la plus plausible est qu'elle reflète les effets de la pollution anthropogénique au cours du siècle dernier.

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## Objectives

1. Retrieve teeth of 250 beluga from the High Arctic (Davis Strait) and Cumberland Sound stocks, spanning the period 1854 to 2005, from museum and DFO collections.
2. Determine animal age, total Hg concentrations, and stable carbon and nitrogen isotope

composition in tooth cementum, to give average life-time measures of Hg exposure and feeding habits.

3. Report on tooth Hg – age regressions as a measure of Hg accumulation rates by the beluga living in different time periods.

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## Introduction

Source apportionment of Hg in the Arctic (i.e., discerning local, natural inputs from long-range transported air pollution (LRTAP)) is recognized by NCP as an important element in understanding why Hg levels in Arctic marine biota are relatively high and increasing in many instances. However, we lack a complete picture of how much anthropogenic Hg is present in Arctic biota, especially in the marine biota that comprise a large portion of traditional country foods. Answering this question requires that the long-term changes in biotic Hg have been determined (back to the 19<sup>th</sup> Century or earlier), using biological hard tissues such as teeth, hair and feathers. But there are relatively few existing long-term datasets from which one can determine the LRTAP contribution. A review of the literature identified fewer than 15 such studies globally (Outridge, 2005), with less than 8 for Arctic animals and humans. These previous studies (on beluga, seabirds, humans, seals, polar bear) found that anthropogenic Hg comprised at least 80% of total modern Hg (Dietz et al., 2006a; Dietz et al., 2006b; Hansen, 1981; Outridge et al., 2005; Outridge et al., 2002). But there are likely to be significant regional variations in the proportion of anthropogenic Hg (e.g. for beluga (Outridge et al., 2005)), and inter-species differences within regions (e.g. among raptors, (Dietz et al., 2006b)). There may be also species demographic and feeding behaviour changes which can complicate interpretation of long-term trends (Outridge 2005), and these have not been considered by all of the published work to date. Thus, additional datasets are required to add to the present incomplete picture.

In this study, the long-term Hg trend was determined in teeth taken from the beluga stock which summers in Canadian waters and winters in Davis Strait off West Greenland. It is believed to comprise one (representing about 15%) of two High Arctic beluga stocks, the other wintering in the North Water polynya (Heide-Jørgensen et al., 2003). Unlike the North Water stock, the Davis Strait stock's annual migration into and out of Canadian and Greenland waters means that both Nunavut and Greenland hunters have harvested the population in historical and modern times (NAMMCO, 2004). Mercury levels in the teeth of modern beluga are strongly correlated with Hg concentrations in internal organs, muktuk and

muscle (Outridge et al. 2000), and so temporal changes in tooth Hg probably reflect soft tissue Hg trends. Age data and stable isotope determinations on the teeth were also carried out or retrieved from existing records to assist in interpreting (or eliminating) alternative causes of tooth Hg changes, such as varying age structure or feeding behaviour of the population.

## Activities in 2008/2009

Historical samples of teeth from beluga (N=27) harvested along the West Greenland coast between 1854 and 1907 were loaned from the Danish Zoological Museum, Copenhagen. Of these, 18 were recorded as collected in 1854, 5 in the period 1868 to 1905, and 4 were not specifically dated by their records but were known to have been harvested sometime before the later decades of the 19<sup>th</sup> Century. Modern samples from West Greenland collected in 1985 (N=38), 1993 (N=40) and 2000 (N=30) were contributed by the Danish Department of Arctic Environment (DMU), Roskilde, and the Greenland Institute for Natural Resources, Nuuk. Because only one beluga stock occurs along the entire length of West Greenland (Heide-Jørgensen et al., 2003), beluga harvested by different communities can confidently be assigned to the same population.

The teeth were carefully cleaned of any adhering mounting material and biological tissue using a hand-held Dremel® grinding tool, and a complete cross-section of cementum (the outer layer of dental material in beluga teeth) removed from each tooth with a cutting tool. The sections typically weighed in the range of 0.3 to 1 g dry wt. This section was immersed in 50% ethanol, followed by immersion in 10% nitric acid and repeated rinses with distilled deionized water, before being air-dried in a HEPA filtered-air work bench. A 20-40 mg subsample of material for stable isotope analysis was removed using the Dremel tool equipped with a stainless steel rotary grinder. Teeth from 1985 and 1993 had previously been aged, but the historical and 2000 samples had not. Therefore, the historical and 2000 year teeth were aged by the same consultant (Age Dynamics, C. Lockyer) who had aged the 1993 samples; a subset of the 1985 teeth (which had been previously aged by a DMU technician) were re-aged as part of this study to ensure comparability in age estimates.

The remaining tooth was aged using standard techniques that enumerate dentine and cementum growth layers; ages were calculated assuming that one growth layer corresponded to one calendar year (Stewart et al. 2006).

Tooth total Hg concentrations were determined by Department of Fisheries and Oceans laboratories, Winnipeg, using cold-vapour AAS techniques described by Outridge et al. (2005). Standard reference materials (TORT-2 and CRM2976, N=4 each) and in-house tooth reference materials (NIST Bone Ash combined with measured quantities of DOLT-2 Liver SRM, giving nominal concentrations of 0.02 and 0.20  $\mu\text{g/g}$  DW; N=5 each) were also analysed within the sample batches, indicating an accuracy better than 5% of certified or nominal values, and a precision of <5% relative standard deviation (RSD). Stable carbon ( $\delta^{13}\text{C}$ ) and nitrogen ( $\delta^{15}\text{N}$ ) isotope determinations were carried out at the Hatch Laboratories, University of Ottawa. 1–2 mg subsamples of tooth powder and standard materials were weighed into tin capsules and loaded into an elemental analyser interfaced to an isotope ratio mass spectrometer (IRMS). The samples were flash combusted at 1800°C, and the resulted gas products carried by helium through columns of oxidizing/reducing chemicals optimised for  $\text{CO}_2$  and  $\text{N}_2$ . The gases were separated by a “purge and trap” adsorption column and sent to the IRMS. All  $\delta^{15}\text{N}$  data were reported as ‰ vs. AIR and normalized to internal standards calibrated to the International Standard Materials IAEA-N1 (+0.4‰), IAEA-N2 (+20.3‰), USGS-40 (-4.52‰) and USGS-41 (47.57‰). All  $\delta^{13}\text{C}$  data were reported as ‰ vs. Pee Dee Belemnite, and normalized to internal standards calibrated to International Standards IAEA-CH-6 (-10.4‰), NBS-22 (-29.91‰), USGS-40 (-26.24‰) and USGS-41 (37.76‰). Internal blind checks on accuracy and precision were performed using an in-house glutamic acid material (N=10), and externally-submitted walrus tooth powder (N=20), which gave values within 0.5% of the long-term average values and within-batch precisions of 0.6% RSD for isotope ratios and 1.4% RSD for C and N concentration data.

## Activities in 2009/2010

A secondary part of this project is to compare Davis Strait and Cumberland Sound (Baffin Island)

beluga in terms of their tooth Hg–age and stable isotope compositions. Therefore, similar analyses are being carried out on the teeth of Cumberland Sound beluga landed at Pangnirtung in 1984, 1997 and 2002. Owing to delays in retrieving the teeth from DFO archives, Hg and stable isotope analyses of these samples (N=80) are not yet completed. To ensure comparability between the Cumberland Sound and Davis Strait age estimates, C. Lockyer is re-ageing 15 Cumberland Sound teeth which were previously aged by DFO; these determinations will also be completed in 2009–10.

## Results

Figure 1 show tooth Hg results for West Greenland beluga as a function of age, for animals harvested in 1854–1905 (“19<sup>th</sup> Century”), 1985, 1993 and 2000. All time periods exhibited significant tooth Hg – age regressions (Table 1). For the 19<sup>th</sup> Century samples this finding supports the idea that the teeth were well preserved and had not been significantly contaminated by Hg during storage, because contamination should result in tooth Hg data independent of animal age. The 5 teeth known to have been collected during 1868 to 1905 period were not statistical outliers from the regression for 1854 beluga alone, and so were included in the 19<sup>th</sup> Century group. The slopes of the tooth Hg – age regression equations on transformed data (Table 1) were significantly higher in all three modern years than in the historical samples, equivalent to 2.3 to 3.3 times more rapid accumulation in modern animals. Beluga harvested in 1993 displayed a lower rate of accumulation than animals caught in 1985 and 2000 (slope of 0.486 vs. 0.659 and 0.681 respectively, with transformed data) mainly because of the absence of individuals in 1993 with tooth [Hg] over 0.10  $\mu\text{g/g}$  DW which were found in the 1985 and 2000 harvests.

Given that these equations were derived from log and square-root transformed data, the untransformed tooth Hg – age relationships tend to be exponential (see Fig. 1). The historical to modern differences therefore increase from younger to older animals, corresponding to almost an order of magnitude difference in average tooth Hg concentrations in older beluga. For example, geometric mean ( $\pm 2$  S.E.) tooth Hg concentrations in 40 yr old beluga were  $0.018 \pm 0.022$   $\mu\text{g/g}$  DW in the 19<sup>th</sup> Century and  $0.121 \pm 0.010$   $\mu\text{g/g}$  DW in the year 2000.

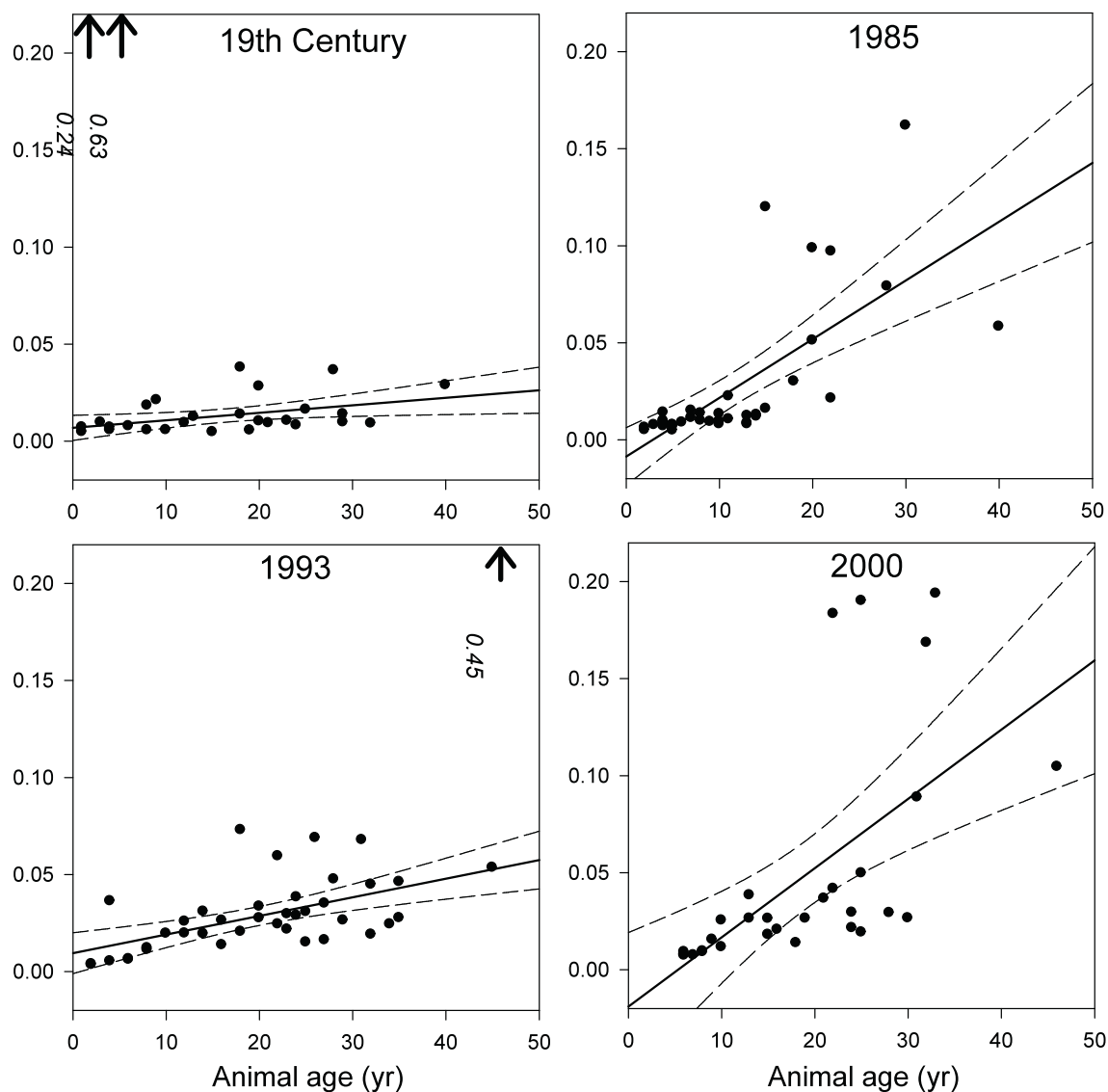


Fig. 1 Tooth mercury concentrations in West Greenland beluga as a function of animal age for four time periods. (Solid lines represent linear regressions on the untransformed data shown, as plotted by SigmaPlot 10.0 software; dashed lines denote 95% confidence intervals. See Table 1 for regression equations on  $\log_n$  and square-root transformed data. Arrows indicate Hg values that were excluded from the regressions because they were statistical outliers to the general relationships)

**Table 1. Tooth mercury vs age regressions for beluga harvested in Davis Strait off West Greenland from 1854 to 2000. (Data were transformed as recommended by Sokal and Rohlf (1981): natural log for Hg concentrations ( $\mu\text{g/g DW}$ ), and square-root for ages)**

Time Period	Regression ( $\pm$ S.E. of co-efficients and intercepts)	Df	$r^2$ value	$P$ value
1854-1905	$\text{Ln Tooth Hg} = 0.208 (\pm 0.074) * \sqrt{\text{Age}} - 5.314 (\pm 0.300)$	26	0.24	0.010
1985	$\text{Ln Tooth Hg} = 0.659 (\pm 0.079) * \sqrt{\text{Age}} - 6.312 (\pm 0.269)$	37	0.66	<0.001
1993	$\text{Ln Tooth Hg} = 0.486 (\pm 0.070) * \sqrt{\text{Age}} - 5.815 (\pm 0.315)$	39	0.56	<0.001
2000	$\text{Ln Tooth Hg} = 0.681 (\pm 0.099) * \sqrt{\text{Age}} - 6.420 (\pm 0.431)$	29	0.63	<0.001



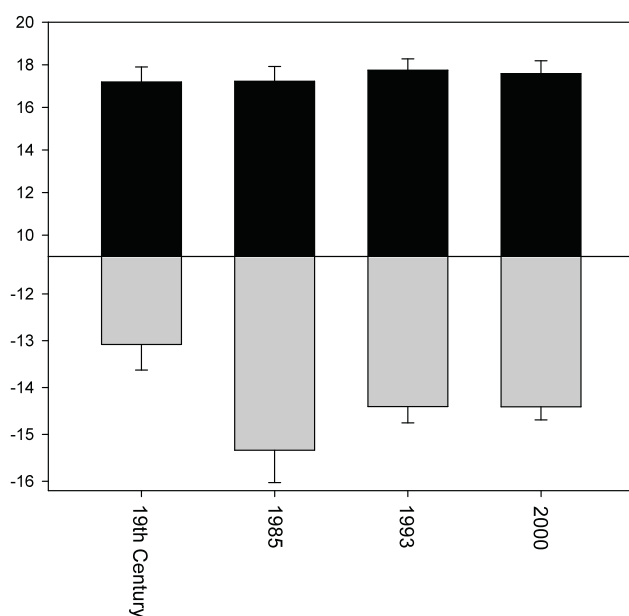


Figure 2. Mean ( $\pm$  S.D.) stable carbon and nitrogen isotope values in West Greenland beluga tooth cementum from the four study time periods.

Average  $\delta^{15}\text{N}$  values were significantly higher in 1993 than in the 19<sup>th</sup> Century and 1985 animals, but only by 0.5-0.6 ‰ (Fig. 2); 1985, 2000 and 19<sup>th</sup> Century data were not significantly different from each other.  $\delta^{13}\text{C}$  values were significantly lower in 1985 than in any other year, and the other modern samples (1993 and 2000) were also significantly lower than in the 19<sup>th</sup> Century (Fig. 2). Carbon concentrations and C:N values were significantly higher in 1985 teeth than in other time periods, and these samples were also different from other samples in that they had been mounted in modeling wax (Table 2). The 19<sup>th</sup> Century samples, many of which had been mounted in

an unknown type of “cement” in the museum, exhibited no sign of C (or N) contamination, because their C/N ratios were identical to 1993 and 2000 year samples. It is unlikely that extraneous C contamination would have an identical C and N content to beluga teeth.

## Discussion and Conclusions

The results of this study on Davis Strait beluga are similar to earlier work on populations in the Beaufort Sea (Outridge et al. 2002, in review) and around Somerset Island in the Central Canadian Arctic (Outridge et al. 2005). In all three studies, significant increases in tooth Hg concentrations occurred that exceeded the pre-industrial or 19<sup>th</sup> Century levels by up to an order of magnitude, with the difference increasing with animal age. This age-dependant pattern is explained by the compounding effects of bioaccumulation and biomagnification exerting a stronger effect on Hg body burdens in older animals than in younger ones (Bernhard and Andreae 1984). Geographically, the Somerset Island study by Outridge et al. (2005) is closest to the present study. Compared to that work, the present study has several advantages including:

- More precise control on the specific High Arctic beluga stock included among the samples (the former study probably included animals from both stocks);
- the historical tooth samples were collected from recently-killed animals, cleaned and subsequently kept in museum dry storage, whereas (Outridge et al. (2005) used teeth collected from archeological sites where weathering and diagenesis of samples may become an issue); and,

Table 2. Stable carbon and nitrogen isotope ratios and concentrations in West Greenland beluga teeth

	$\delta^{13}\text{C}$ Mean	$\delta^{13}\text{C}$ S.D.	$\delta^{15}\text{N}$ Mean	$\delta^{15}\text{N}$ S.D.	C Mean (% DW)	C (%) S.D.	N Mean (% DW)	N (%) S.D.	C/N Mean	C/N S.D.
19 <sup>th</sup> Century	-13.08	0.54	17.20	0.70	12.60	0.64	4.12	0.23	3.06	0.06
1985	-15.34	0.69	17.22	0.70	13.43	0.78	4.12	0.23	3.26	0.12
1993	-14.41	0.35	17.75	0.53	12.11	0.53	3.96	0.19	3.06	0.04
2000	-14.41	0.28	17.58	0.60	12.12	0.47	3.96	0.15	3.06	0.05

- a longer time trend going back to 1854, with intervening samples from the 1870 to 1905, together with a more comprehensive sampling of recent decades (1985, 1993 and 2000) which should better document any inter-annual variation in the harvest.

One difference with the earlier beluga studies is that the previous age estimates which assumed two dental growth layers were deposited per year are now known to be incorrect, and that only one layer per year is laid down (Stewart et al. 2006). Therefore, the slopes and intercepts of Hg – age regression equations derived here cannot be directly compared with earlier published regressions; those equations must be recalculated with the new age estimates before direct comparison can occur. However, the results for specific ages can be compared. For the Davis Strait population, the geometric mean tooth Hg concentration in 40 yr olds in 2000 was 6.7 times higher than in the 19<sup>th</sup> Century. Accordingly, the calculated proportion of anthropogenic Hg in 40 yr olds in 2000 was 85%, assuming that the 19<sup>th</sup> Century samples approximate naturally-occurring Hg levels in the population. Similar findings were reported for Somerset Island beluga in the 1990s (5.9 times increase over late 19<sup>th</sup> Century, Outridge et al. 2005) and Beaufort Sea beluga in 1993 (9.1 times above 15<sup>th</sup>-17<sup>th</sup> Centuries, Outridge et al. 2002 and in review).

The stable isotopic results indicated that the evidence for a significant anthropogenic input, based on the long-term Hg data, was not affected by significant changes in beluga feeding behaviour over time. Although 1993 animals had slightly higher  $\delta^{15}\text{N}$  values than other years, which might bias their Hg values towards higher levels, the regression for 1993 was in fact somewhat lower than in 1985 and 2000 although still significantly higher than the 19<sup>th</sup> Century animals. Belugas are known to live up to 60 years of age or more, and their tooth cementum cross-sections reflect incremental material laid down over their lives. Although teeth cross-sections therefore integrate inter-annual variation of Hg exposure throughout the life of each individual, and are thus more representative of lifetime average exposure than soft tissue data, it is possible that significant within-population differences in Hg body burdens occur, such as those reported by Loseto et al. (2006)

among the Beaufort beluga. The inclusion or exclusion of these individuals in certain annual harvests could cause the inter-annual differences in tooth Hg – age regressions observed in the present study. However, these recent inter-annual or decadal fluctuations in Davis Strait beluga are minor in comparison to the long-term increase of tooth Hg.

Interpretation of the  $\delta^{13}\text{C}$  data is complicated by evidence, in the form of C concentration and C/N ratio data, that the modeling wax used to originally mount the 1985 teeth had irredeemably contaminated the teeth with small amounts of C with depleted isotopic values, despite thorough attempts at cleaning the samples. A similar finding was reported by Stewart et al (2006) in beluga teeth mounted in epoxy resin. Leaving the 1985 data aside, the 1993 and 2000 teeth were never exposed to mounting wax or other synthetic organic material, and their C/N ratios were similar to each other and to the 19<sup>th</sup> Century sample, suggesting uncontaminated samples in all 3 groups. C/N ratios are employed to judge the degree of preservation and organic contamination of ancient biological materials (DeNiro 1985). The most likely explanation for the decline of  $\delta^{13}\text{C}$  in 1993 and 2000 relative to the 19<sup>th</sup> Century is the dissolution of anthropogenic  $\text{CO}_2$  in the Arctic Ocean (see Quay et al. 1992). While a shift of beluga to deeper, pelagic feeding locations in recent years is a possible alternative explanation, the same decline of  $\sim 1.5\text{‰}$  has been observed in other northern marine biota in different regions and was attributed to a  $\text{CO}_2$  effect (Newsome et al. 2007). Even if the Davis Strait  $\delta^{13}\text{C}$  data were evidence of more recent pelagic feeding, pelagic fish are thought to contain lower methyl-Hg concentrations than in-shore and benthic prey (Stern and Macdonald 2005), which would tend to reduce the modern Hg levels relative to the 19<sup>th</sup> Century. Therefore, the conclusion that anthropogenic Hg comprises 85% of total Hg in modern 40 yr old beluga is a robust and conservative finding.

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# Atmospheric deposition and release of methylmercury in glacially-fed catchments of Auyuittuq National Park, Baffin Island

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## Abstract

Atmospheric deposition and release of mercury (THg) and monomethyl-mercury (MeHg) was measured in the snowpack, firn, and in lakes and streams of the glacierized catchment around the 6000 km<sup>2</sup> Penny ice cap in Auyuittuq National Park, southern Baffin Island. The mean levels of THg and MeHg in snow and firn were  $0.692 \pm 0.478$  ng/L and  $0.159 \pm 0.163$  ng/L, respectively. Variations between freshly fallen snow, aged snow and firn were relatively small, and within the observed range of sample-to-sample variability at the scale of sampling. No compelling evidence of surface Hg enhancement in the winter snowpack possibly associated with AMDEs was found. We estimated net accumulation rates for THg and MeHg on Penny ice cap to be  $\sim 0.25$  and  $0.06 \mu\text{g m}^{-2} \text{yr}^{-1}$ , respectively. Seasonal thaw-freeze cycles in the snowpack may limit post-depositional evasion of Hg from the firn on the ice cap. While the levels of THg in meltwater-fed streams and lakes compare with those measured in snow and firn, the MeHg levels in surface waters were more than 10 times lower than in snow and firn,

## Résumé

Les retombées atmosphériques et les rejets de mercure (THg) et de mercure monométhylé (MeHg) ont été mesurés dans le manteau neigeux, dans le névé ainsi que dans les lacs et cours d'eau du glacier d'amoncellement de neige de la calotte glaciaire Penny de 6 000 km<sup>2</sup>, dans le parc national Auyuittuq, au sud de l'île de Baffin. Les niveaux moyens de THg et de MeHg dans la neige et dans le névé étaient de  $0,692 \pm 0,478$  ng/L et de  $0,159 \pm 0,163$  ng/L, respectivement. Les variations entre la neige fraîche, la neige ancienne et le névé étaient relativement faibles, ainsi que dans la plage observée de la variation d'un échantillon à l'autre de notre échantillonnage. Nous n'avons trouvé aucune preuve convaincante d'une augmentation du Hg de surface dans le manteau neigeux d'hiver qui pourrait être associée aux phénomènes d'épuisement du mercure atmosphérique (AMDE). Nous avons estimé les taux nets d'accumulation du THg et du MeHg de la calotte glaciaire Penny à  $\sim 0,25$  et à  $0,06 \mu\text{g m}^{-2} \text{an}^{-1}$ , respectivement. Les cycles saisonniers de gel et de dégel du manteau neigeux peuvent limiter l'évasion du Hg après sédimentation du névé de la calotte glaciaire.

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suggesting rapid loss of MeHg from the snowpack prior to or during spring freshet, or from the meltwater itself by as yet undetermined processes.

## Key Messages

- Direct atmospheric deposition of mercury (THg) and monomethyl-mercury (MeHg) occurs in the Penny ice cap catchment.
- The measured concentrations in snow and firn (aged snow) are comparable to those measured in other Arctic glaciers. Mean values are  $\sim 0.7$  ng/L for THg and  $\sim 0.2$  ng/L for MeHg.
- The MeHg accounts on average for  $\sim 20$  % of THg.
- The partial summer melt and refreezing of the snowpack creates physical barriers (ice layers) in the snowpack that probably limit evasion of Hg, and promote its storage in the firn (aged snow).
- While the THg in the snowpack may contribute much of the Hg found in lakes and streams, it appears that MeHg is lost from the snowpack early in the melt season or is rapidly removed from meltwater after its release, such that the observed levels in these waters are much lower than those found in the snowpack.

Alors que les niveaux de THg dans les cours d'eau et dans les lacs alimentés par la fonte des neiges se comparent à ceux mesurés dans la neige et dans le névé, les niveaux de MeHg dans les eaux de surface étaient 10 fois plus bas que dans la neige et dans le névé, ce qui permet de croire à une perte rapide du MeHg dans le manteau neigeux avant ou pendant la crue nivale du printemps ou dans l'eau de fonte elle-même selon un processus encore indéterminé.

## Messages clés

- Des retombées atmosphériques de mercure (THg) et de mercure monométhylé se produisent dans la calotte glaciaire Penny.
- Les concentrations mesurées dans la neige et dans le névé (neige ancienne) sont comparables à celles mesurées dans d'autres glaciers de l'Arctique. Les valeurs moyennes sont de  $\sim 0,7$  ng/L pour le THg et de  $\sim 0,2$  ng/L pour le MeHg.
- En moyenne, le MeHg représente  $\sim 20$  % du THg.
- Le dégel estival partiel et le gel du manteau neigeux créent des obstacles physiques (couches de glace) dans le manteau neigeux qui limitent probablement l'évasion du Hg et favorisent son accumulation dans le névé (neige ancienne).
- Alors que le THg dans le manteau neigeux peut contribuer à la plupart du Hg que l'on trouve dans les lacs et dans les cours d'eau, il semblerait que le MeHg s'échappe du manteau neigeux lors de la fonte ou s'élimine rapidement de l'eau de fonte, ce qui fait que les niveaux observés dans ces eaux sont nettement inférieurs à ceux que l'on trouve dans le manteau neigeux.

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## Objectives

The primary objectives of this study were to:

- (1) Quantify the atmospheric input of total and monomethyl mercury (MeHg) in snow, glacial meltwaters, stream and lake waters of the Penny ice cap catchment in Auyuittuq National Park;
- (2) Investigate the internal storage and release of Hg and MeHg in glacial firn and ice; and
- (3) Investigate the relationship between atmospheric deposition of MeHg and marine aerosols (e.g., sulfate, bromine) deposited in the eastern Arctic under present and past environmental conditions (e.g., reduced sea-ice cover) using ice cores.



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Objectives (1) and (2) were accomplished, while objective (3) will be addressed as part of the follow-up (year 2) of this study.

## Introduction

Arctic snowpacks are believed to act as temporary sinks for atmospheric Hg which may be released during spring snowmelt into aquatic ecosystems. While much attention has been devoted to the photo-chemical processes that transfer inorganic Hg between air and snow, the sources and fates of toxic monomethyl-mercury (MeHg) in polar snow are still poorly understood. In particular, there are few quantitative estimates of direct atmospheric inputs of MeHg to Arctic snowpacks. This project aims to improve our understanding of the sources and pathways that deliver Hg, and in particular MeHg, to the aquatic ecosystem in a glacierized catchment on southern Baffin Island. The study area is Auyuittuq National Park on Cumberland Peninsula. In its center is the 6000 km<sup>2</sup> Penny ice cap, which feeds meltwater to lakes and streams draining into Cumberland Sound or Baffin Bay. We sought to quantify the atmospheric deposition and net accumulation of Hg and MeHg in Penny ice cap, and its release by summer melt. By comparing levels of total mercury (THg) and MeHg in snow, firn and glacial ice with levels measured in meltwater-fed streams and lakes, we attempted to quantify the contribution of atmospheric Hg input in snow to the aquatic environment. In previous work, Loseto et al. (2004) found that most of the mercury other than MeHg was lost in the spring freshet of streams in the Resolute (Cornwallis Island) area. In unpublished work from the Resolute Area we found little photodegradation of MeHg in snow but it was rapidly degraded in lake water samples in the presence of dissolved organic carbon. We therefore sought to establish if the same process(es) were effective in the Penny ice cap catchment. A secondary goal of our study was to compare concentrations and fluxes of Hg deposited on Penny ice cap with measurements made elsewhere in the Arctic (e.g., St-Louis *et al.*, 2005) in order to evaluate regional differences and infer dominant Hg source areas and atmospheric transport pathways. This goal complements a major International Polar Year (IPY) research initiative led by GSC, which aims to document spatial and

temporal trends of climate and air pollution in the Arctic using snow and ice cores. Our proposal for 2008-09 was financially and logistically leveraged by both NCP and IPY.

## Activities in 2007/2008

Surface and sub-surface snow, firn and ice samples were collected on Penny ice cap in April 2007 and April 2008 (Fig. 1). A total of ~140 samples were obtained (many in duplicate), with volumes varying between ~1 and 2.5 L (liquid). Surface snow samples were taken along a series of transects to define local spatial variability in Hg deposition, while sub-surface samples were taken from snowpits and shallow firn cores at the summit of the ice cap (~1830 m asl). One series of surface snow samples was collected immediately following a storm that deposited 20 cm of fresh snow on Penny ice cap in mid-April 1997. The accumulation period spanned by the sub-surface samples is estimated to be 10-12 years. The stratigraphy, density and texture of the snow and firn strata were recorded in detail, and updated hourly recordings of air temperature and snow height surface changes were downloaded from the autostation installed on the ice cap in April 2007. All samples were returned frozen to Ottawa where they were processed and analyzed for [THg] and [MeHg] following stringently clean protocols and standard methods. Summer field work was conducted in early July 2008 with helicopter support. A total of 16 sites were sampled, which included streams, ponds and lakes fed by snowpack or glacial meltwater (Fig. 2). Measurements were made at each site for pH (acidity), specific conductivity, and temperature. Samples for THg and MeHg were taken in duplicate at each site. These were returned to Ottawa and analyzed by the same methods as the frozen samples, using the standard EPA method 1631e involving oxidation of Hg to Hg(II) by BrCl, followed by analysis by CV-AAS. For the purpose of this study, "total mercury" (THg) refers to that portion of Hg in water which can be quantified by this protocol. For quality assurance, a sub-set of snow and firn samples were also split and analyzed for [THg] in laboratories at the University of Ottawa Biosciences Complex and at the GSC.



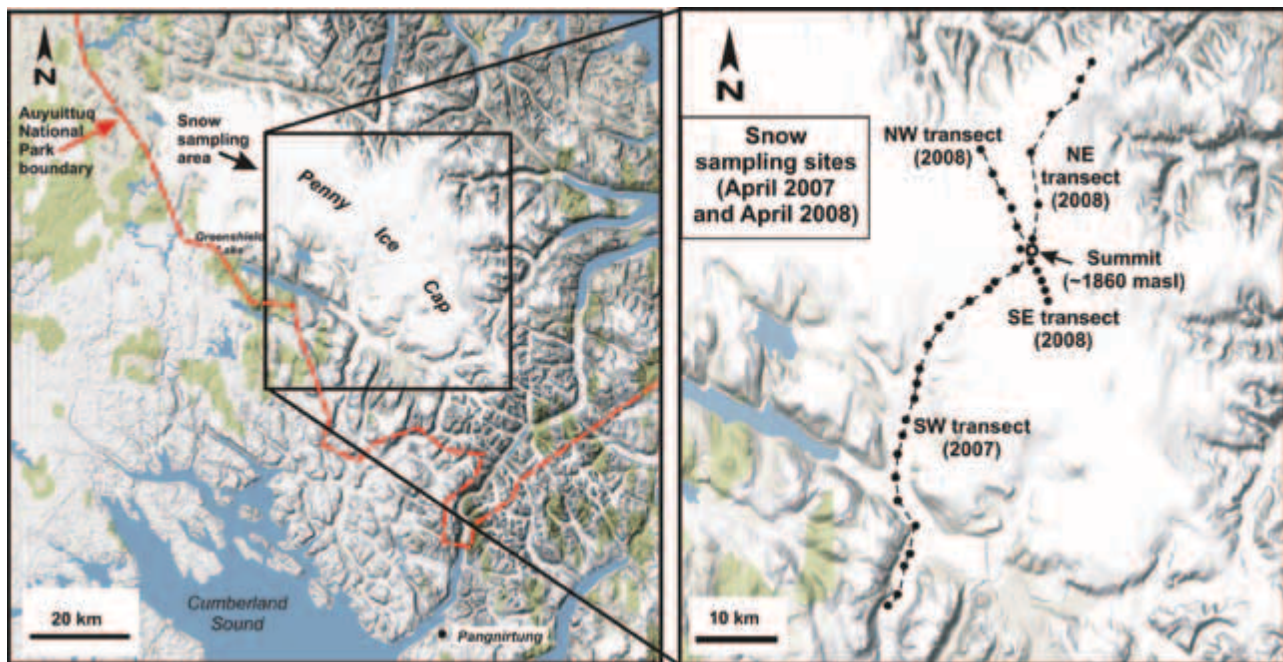


Figure 1. Location of Snow and Firn sampling sites on Penny ice cap, Auyuittuq National Park, Baffin Island

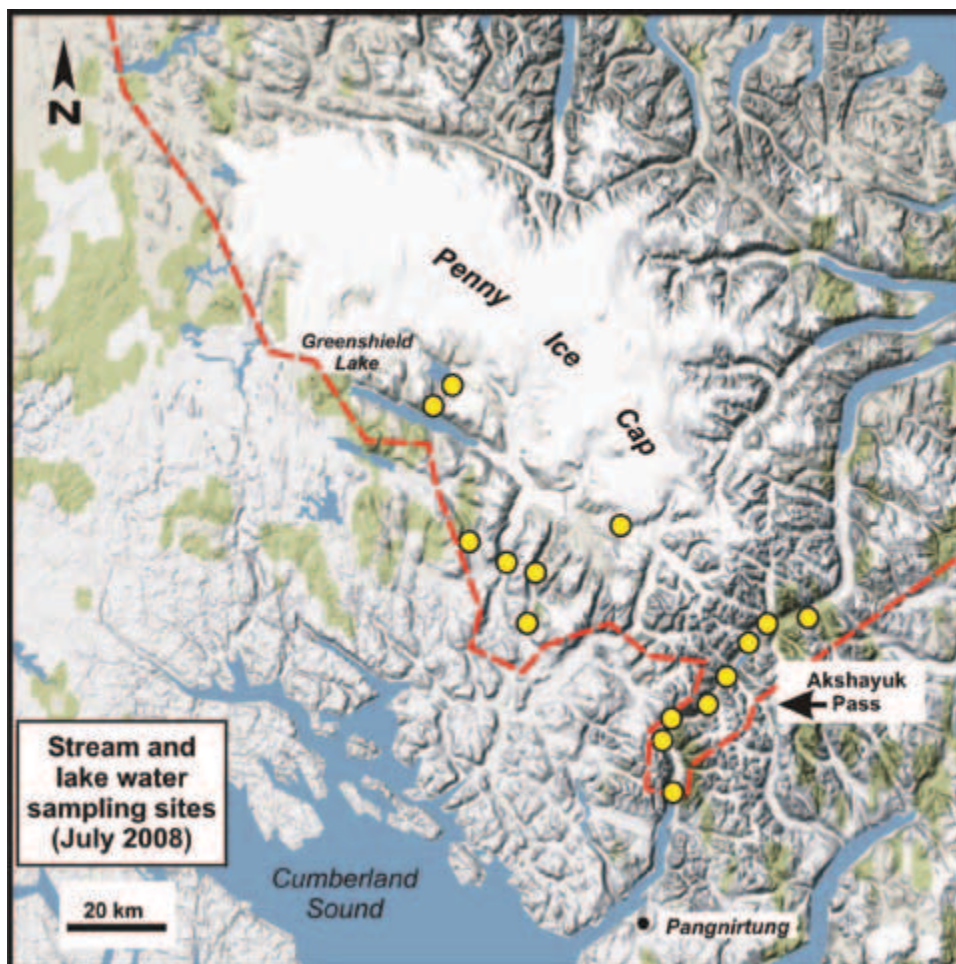


Figure 2. Location of stream and lake water sampling sites in Auyuittuq National Park

## Results

Results are summarized below in Table 1 (snow, firn and ice) and 2 (lake and stream waters).

## Discussion and Conclusions

(What follows is a very preliminary discussion of results) The average [THg] and [MeHg] measured in snow and firn on Penny ice cap representing 1-12 years of accumulation are 0.692 ng/L and 0.159 ng/L, respectively, with MeHg representing ~20 % of the THg measured. On average, the lowest [THg] was that of freshly fallen snow (0.526 ng/L), while the highest [THg] was in discrete ice layers found in the snowpack or firn (1.000 ng/L). Aged surface (winter) snow had a mean [THg] of 0.772 ng/L, only slightly higher than in deeper winter snow strata (0.544 ng/L). The mean [THg] in firn was higher still (0.854 ng/L). For [MeHg], mean concentrations ranked from lowest in aged winter snow (0.083-0.084 ng/L),

followed by firn and ice layers (0.107 to 0.131 ng/L), and the highest mean values were in freshly fallen snow (0.161 ng/L). For surface snow ( $Z \leq 10$  cm) collected within the same year (2007 or 2008), the [THg] and [MeHg] levels in individual samples varied by 20-30 % and 27-45 %, respectively, relative to mean values. In freshly fallen snow the sample-to-sample variability was 29 % of the mean for [THg], and 45 % of the mean for [MeHg], over a linear sampling distance of ~60 km and an altitude range of ~1500 m. Hence the spatial variability of THg and particularly that of MeHg is quite large in surface snow. This is probably due to the heterogeneity of atmospheric deposition, combined with “stratigraphic noise” induced by snow drifting and post-depositional modification (e.g., evasion following photoreduction). The slight differences found between the mean concentrations of THg and MeHg in freshly fallen snow, aged surface snow, and subsurface winter snow are therefore well within the range of expected natural

**Table 1. Total mercury (THg) and monomethyl-mercury (MeHg) concentrations in snow, firn and ice on Penny ice cap (sampled April 2007 and 2008).**

Total mercury [THg] (ng/L) (D.L. = 0.200 ng/L)						
	n	Min	Max	Average	Std Dev	Median
Fresh snow ( $Z \leq 10$ cm)	30	0.333	1.041	0.526	0.153	0.525
Aged snow ( $Z \leq 10$ cm)	20	0.263	2.880	0.772	0.624	0.565
Aged snow ( $Z > 10$ cm)	16	0.216	1.243	0.544	0.284	0.459
Firn (incl. ice layers)	73	0.203	3.477	0.854	0.629	0.661
Ice layers in snow and firn	20	0.364	2.058	1.000	0.400	0.892
All snow, firn and ice samples	141	0.203	3.477	0.692	0.478	0.562
Monomethyl-mercury [MeHg] (ng/L) (D.L. = 0.020 ng/L)						
	n	Min	Max	Average	Std Dev	Median
Fresh snow ( $Z \leq 10$ cm)	30	0.059	0.335	0.161	0.073	0.142
Aged snow ( $Z \leq 10$ cm)	18	0.044	0.129	0.083	0.020	0.076
Aged snow ( $Z > 10$ cm)	16	<0.020	0.178	0.084	0.048	0.086
Firn (incl. ice layers)	68	<0.020	0.460	0.107	0.072	0.093
Ice layers in snow and firn	20	0.031	0.379	0.131	0.102	0.121
All snow, firn and ice samples	137	0.021	0.868	0.159	0.163	0.104

**Table 2. Total mercury (THg) and monomethyl-mercury (MeHg) concentrations in snow, firn and ice on Penny ice cap (sampled July 2008).**

		Total mercury [THg] (ng/L) (D.L. = 0.200 ng/L)				
	n	Min	Max	Average	Std Dev	Median
Akshayuk Pass lakes + streams						
Unfiltered	14	0.695	2.038	1.349	0.516	1.324
Filtered	8	0.085	1.029	0.424	0.363	0.279
Ice-contact lakes & streams						
Low sediment load	5	0.278	1.403	0.796	0.500	0.740
Turbid	9	2.403	6.853	4.439	1.759	3.911
All water samples - July 2008	36	0.085	6.853	1.839	1.164	1.819
		Monomethyl-mercury [MeHg] (ng/L) (D.L. = 0.020 ng/L)				
	n	Min	Max	Average	Std Dev	Median
Akshayuk Pass lakes + streams						
Unfiltered	14	0.000	0.024	0.009	0.008	0.009
Filtered	8	0.000	0.020	0.010	0.012	0.007
Ice-contact lakes & streams						
Low sediment load	5	0.000	0.025	0.010	0.010	0.007
Turbid	9	0.000	0.020	0.007	0.008	0.005
All water samples	36	0.000	0.025	0.009	0.008	

variability. Thus we did not find compelling evidence of a surface enhancement in [THg] (or [MeHg]) in the winter snowpack on Penny ice cap that could be attributed to Atmospheric Mercury Depletion Events as was reported elsewhere (Steffen et al., 2008, Poissant et al., 2008; and references therein). If such events took place in the southeastern Baffin region between polar sunrise (early February at this latitude) and the time of our sampling (early to mid-April), the resulting Hg enhancements in the winter snowpack were either short-lived (i.e., the Hg in surface snow was re-emitted to the air) or they were undetectable at the scale at which we sampled. The mean [THg] figures reported here for the Penny ice cap snowpack are comparable with published and unpublished measurements made at Canadian High Arctic glacier sites (St-Louis et al., 2005; Zheng, 2008, pers. comm.) and in central Greenland (Boutron et al., 1998; Mann et al., 2005). Mean values of [THg] in snow or recent firn reported for these sites vary within a narrow

range of ~0.4 to 1.0 ng/L, and are typically close to an overall average of ~0.6 ng/L. However net Hg accumulation rates in snow may vary between these sites as a function of the wet/dry deposition ratios and snow+ice accumulation rates at each site. On Penny ice cap the estimated net accumulated rate is 0.25  $\mu\text{g m}^{-2} \text{yr}^{-1}$  for THg, which is considerably less than modern atmospheric deposition rates in the Arctic inferred from lake sediments and peat bogs (Outridge *et al.* 2008 and references therein). The estimated net accumulation rate for MeHg is 0.06  $\mu\text{g m}^{-2} \text{yr}^{-1}$ . There are indications that Hg may be more efficiently stored in Penny ice cap (i.e., lesser re-emission to the atmosphere) than in colder glaciers owing to entrapment in firn during summer thaw-refreezing cycles. If this is the case, it may be possible to determine accumulation rates of Hg on the ice cap under past climate regimes using existing archived or new ice cores. Net Hg storage presently only occurs in the area above the ice cap's equilibrium line, which lies somewhere between 1500 and



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1750 m asl. Atmospheric Hg (and MeHg) deposited in snow at lower elevations is lost to melt and runoff during summer, and enters streams and lakes draining towards Cumberland Sound or Baffin Bay. Measurements of [THg] in selected lakes and streams sampled in July 2008 revealed [THg] in water that were only slightly higher than in the Penny ice cap snow cap, typically ranging between 0.4 and 1.4 ng/L, but higher in unfiltered or turbid (sediment-rich) waters where a geogenic Hg source may contribute part of the total [THg] measured. In contrast, the [MeHg] levels in streams and lakes were surprisingly low, ranging from <0.020 ng/L to 0.025 ng/L, and averaging 0.009 ng/L, i.e. more than 10 times lower than the mean levels measured in the Penny ice cap snowpack. If the MeHg levels in the latter are representative of snow cover elsewhere in the study catchment, it is remarkable that the lakes and streams, which are primarily fed by snow and ice meltwater, should have [MeHg] so much lower than in the snowpack. This suggests that either the MeHg present in the winter snowpack is mostly lost in the spring freshet (prior to the time we sampled in July), or that MeHg is lost from meltwater in a relatively short time (days to weeks) after it is released from the snowpack, by processes which are yet to be identified.

## Expected Project Completion Date

The second phase of our project is scheduled for completion in late March, 2010.

## Acknowledgments

We wish to thank the Polar Continental Shelf Project for aircraft support during field operations, and Parks Canada staff in Pangnirtung for invaluable logistical assistance. Steven Akeeagok, Daniel Kilabuk and Christophe Kinnard were energetic and enthusiastic helpers in the field. Emmanuel Yumvihoze (Univ. of Ottawa) and Pierre Pelchat (GSC) assisted with the laboratory work. This project was supported by INAC-NCP, NRCan, NSERC, and the International Polar Year initiative.

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# Characterizing contaminant-related health effects in Beluga whales (*Delphinapterus leucas*) from the western Canadian Arctic.

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## Abstract

Beluga whales (*Delphinapterus leucas*) in the Arctic may be vulnerable to the combined effects of contaminants and a changing climate. That said, very little is known about beluga health. We have been pioneering new methods to measure the health of beluga whales in close collaboration with Inuvialuit community members from Tuktoyaktuk and with other researchers working in the area. Our beluga whale health research continues, with investments in sampling, methods development, contaminant analysis, genomics analysis and preliminary results interpretation well underway. A changing climate and ice regime may have dramatic impacts on contaminant pathways, food web productivity, and beluga feeding ecology. If these two major stressors affect beluga health, reduce stock abundance or diminished quality of beluga whales may impact upon community harvests. Further efforts that have engaged the community members of Tuktoyaktuk and students have helped to enhance outreach as

## Résumé

Les bélugas (*Delphinapterus leucas*) de l'Arctique sont sans doute vulnérables aux effets combinés des contaminants et du changement climatique. Cela dit, nos connaissances sur la santé des bélugas sont très limitées. Nous avons mis au point de nouvelles méthodes pour évaluer la santé des bélugas en étroite collaboration avec les membres des collectivités inuvialuit de Tuktoyaktuk ainsi qu'avec d'autres chercheurs qui travaillent dans la région. Nous poursuivons nos recherches sur la santé des bélugas et investissons dans l'échantillonnage, la mise au point de méthodes, les analyses de contaminants et les analyses génomiques, et les interprétations des résultats préliminaires sont bien en cours. Le changement climatique et du régime des glaces risque d'avoir d'importantes conséquences sur les voies de passage des contaminants, la productivité du réseau trophique et l'écologie de l'alimentation du béluga. Si ces deux importants facteurs de stress affectent la santé des bélugas, la diminution de l'abondance des stocks ou de la qualité des bélugas

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well as the scientific viability of this project. This project represents an emerging collaboration between the Northern Contaminants Programme (NCP-INAC) and the Department of Fisheries and Oceans Canada (*Ecosystem Research Initiative and National Headquarters*)

## Key Messages

- a second year of field sampling has been successfully completed, with the participation of team members, collaborators and community members;
- 24 beluga whales were sampled at Hendrickson island in the Beaufort Sea;
- new beluga whale-specific genomic techniques were developed in collaboration with the University of Victoria, with a total of 15 gene primers now available;
- 24 beluga whale blubber samples were analyzed for priority contaminants, including PCBs and PBDEs at the DFO Laboratory for Aquatic Chemical Analysis in Sidney BC, and metabolites at NWRC;
- preliminary results suggest that some gene expression endpoints correlate negatively with PCBs, although outstanding age information may refine this observation;
- the concentration of vitamin A correlated positively with PCBs, but thyroid hormone concentrations did not;
- PCB concentrations correlated with body length (as in 2007), this being attributable to a combination of age (data outstanding) and feeding ecology (stable isotopes);
- Additional sampling in 2009-10 and further laboratory investigations will provide insight into the roles of feeding ecology, condition, and condition on the health of beluga whales.

risque d'avoir une incidence sur les récoltes des collectivités. Des efforts supplémentaires auxquels ont participé les membres de la collectivité de Tuktoyaktuk ainsi que des étudiants ont aidé à renforcer le rayonnement et la viabilité scientifique de ce projet. Ce projet est le fruit d'une collaboration naissante entre le Programme de lutte contre les contaminants du Nord (PLCN – AINC) et le ministère des Pêches et des Océans (*Projet de recherche sur l'écosystème et administration centrale*).

## Messages clés

- Une deuxième année d'échantillonnage sur le terrain a été terminée avec succès, avec la participation des membres de l'équipe, des collaborateurs et des membres de la collectivité.
- Des échantillons ont été prélevés sur 24 bélugas autour de l'île Hendrickson dans la mer de Beaufort.
- De nouvelles techniques génomiques adaptées aux bélugas ont été mises au point en collaboration avec l'université de Victoria, avec un total de 15 amorces de gènes disponibles.
- Les échantillons de lard de 24 bélugas ont été analysés pour détecter la présence de contaminants prioritaires, y compris les BPC et les PBDE dans le laboratoire d'expertise en analyses chimiques aquatiques du MPO, à Sidney, C.-B., et de métabolites au CNRF.
- Les résultats préliminaires montrent que certains effets d'expression génétique présentent une corrélation négative avec les BPC, cependant des renseignements sur l'âge pourraient affiner cette observation.
- La concentration de vitamine A présente une corrélation positive avec les BPC, mais pas les concentrations d'hormones thyroïdiennes.
- Les concentrations de BPC présentent une corrélation avec la longueur du corps (tout comme en 2007), ce qui est attribuable à une combinaison de l'âge (données en suspens) et des relations trophiques (isotopes stables).
- L'échantillonnage supplémentaire qui sera effectué en 2009-2010 ainsi que des recherches en laboratoire supplémentaires offriront de nouvelles connaissances sur les rôles et les conditions des relations trophiques et les conditions sur la santé des bélugas.



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## Objectives

Characterize health risks associated with dietary intake, accumulation and metabolism of contaminants in beluga whales.

## Introduction

Beluga whales are at the top of the Arctic food web, rendering them vulnerable to contamination by a variety of persistent environmental contaminants that are transported from distant sources. Our research has demonstrated that complex environmental mixtures of POPs are affecting the health of free-ranging pinnipeds, and present a tangible risk to cetaceans, in several areas of southern Canada (Ross 2006; Mos *et al.* 2006; Tabuchi *et al.* 2006). Concerns about contaminants in the Canadian Arctic emanate, in part, from studies which demonstrate possible health effects in subsistence-oriented humans, polar bears and beluga whales (Dallaire *et al.* 2004; White *et al.* 1994; Braathen *et al.* 2004). Technical and logistical challenges have generally precluded health assessments in Arctic marine mammals, but new and emerging technologies provide an opportunity to build on past efforts and shed light on an important facet of Arctic contamination (Stern *et al.* 2005; Lockhart *et al.* 2005; Muir *et al.* 1999).

A complicating juxtaposition in the way of climate change may add a layer of stress to beluga whales, by altering contaminant pathways, or by reducing the condition of beluga whales (Macdonald 2005). Despite the wealth of information about pathways and fate of different classes of environmental contaminants over time and space in the Arctic little is known about adverse health effects associated with these contaminants in vulnerable species, including beluga whales.

Changes in Arctic sea ice cover, temperature profiles, food web productivity, and beluga distribution and feeding ecology may profoundly change the course of contaminant fate in the Arctic environment, as well as the condition and health of beluga. Climate change has the potential to confound our understanding of mechanistic linkages between contaminant exposure and health effects, but may also have serious implications for the health of beluga whale populations. Our programme is designed to characterize condition and

nutritional status of belugas in order to generate insight into the relationship between climate, contaminants and health of beluga.

## Activities 2008/2009

This was an intensive year that incorporated a large field camp in the summer of 2008, and a series of efforts in the laboratory aimed at developing, validating and applying health assessment techniques to beluga whales.

We partnered with other beluga experts at Hendrickson Island to conduct a holistic sampling program. In addition to our NCP funded program were projects designed to examine mercury and new compounds (Stern and Tomy). We worked closely with both programs to ensure the best sampling strategies that avoided duplication of efforts. In addition to our research team, the following individuals and programs participated in the Hendrickson Island effort: L. Chan and S. Ostertag carried out a neurotoxic study (funded by ArcticNet), and S. Raverty and O. Nielsen examined beluga for disease and parasites (CFL/IPY, S. Ferguson). In addition to the science teams, participants from the community included youth and elders. Frank and Nellie Pokiak along with their family sampled at Hendrickson Island. The FJMC-funded beluga whale monitor participated in sampling. Two youth participated in the science program (Nasivvik Funded, Ostertag/Loseto). A young family assisted with camp logistics (Loseto/Ross FJMC funded). Camp was set up June 30 2009 and shut down July 22 2009.

## Results

### *Morphometrics*

We collected samples from 24 beluga whales for health assessments and contaminant analyses in collaboration with the Tuktoyaktuk community beluga hunt. Of the 24 whales harvested and sampled 19 were male (mean length = 420 cm;  $\sigma = 27$  cm), 4 were female (mean length = 363 cm;  $\sigma = 16.8$ ), one of which had a fetus that was full term (160 cm). Blubber thickness averaged 8 cm ( $\sigma = 2.8$ ), with males averaging 8.7 cm ( $\sigma = 2.7$ ) and females 7.4 cm ( $\sigma = 2.9$ ). Age will be determined by G. Stern (FWI/DFO)

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## Genomics

Blubber, skin, liver and muscle samples were taken from the 24 Hendrickson Island beluga whales, preserved in RNA later and stored at -20°C until total RNA isolation. Blubber samples were divided into inner, middle, and outer blubber in order to investigate any possible variations in gene expression within the blubber layer.

Briefly, total RNA in those samples was isolated using the single-step RNA isolation method based on guanidine isothiocyanate / phenol / chloroform extraction with Trizol (Invitrogen Canada Inc., Toronto, Ontario, Canada) as a reagent. Total cDNA was produced using Superscript II RNase H- reverse transcriptase (Invitrogen Canada Inc.). At present, RNA from all the 24 samples of blubber, skin, liver and muscle has been successfully extracted.

In order to assess the health of this beluga whale population, a total of 15 target genes were developed so far, including three housekeeping genes (ribosomal protein L8, Glyceraldehyde-3-phosphate dehydrogenase (GAPDH) and cytoplasmic B-actin). The expression of those specific genes was investigated using quantitative real-time polymerase chain reaction (qRT-PCR) and relative quantification (based on the best housekeeping gene):

- Ribosomal protein L8 (L8)
- Glyceraldehyde-3-phosphate Dehydrogenase (GAPDH)
- Cytoplasmic Beta Actin (CBA)
- Thyroid Receptor Alpha (TR-a)
- Thyroid Receptor Beta (TR-b)
- Estrogen Receptor Alpha (ER-a)
- Peroxisome Proliferator-Activated Receptor Gamma (PPAR-g)
- Retinoid X Receptor Alpha (RXR-a)
- Adiponectin
- Leptin
- Insulin-Like Growth Factor 1 (ILGF1)
- Vitamin D Receptor (VDR)
- Metallothionein I (MT1)
- Heat Shock Protein 70-1 (HSP70-1)

- Glucocorticoid Receptor (GR)
- Aryl Hydrocarbon Receptor (AhR)

Housekeeping genes are used to evaluate the quality of the data. Using the “crossing point method” (Pfaffl *et al.*, 2004), L8 was found to be the most stable gene amongst the three housekeeping genes and was therefore chosen as a normalizer gene. L8 values were also used to identify outliers in the dataset.

Preliminary results are available for the three housekeeping genes as well as three of the toxicology related genes (Aryl hydrocarbon receptor (AhR), glucocorticoid receptor (GR) and metallothionein (MT1)) in the outer blubber layer and muscle samples.

While AhR, GR, and MT1 expressions were not influenced by such factors as body length and blubber thickness ( $p > 0.05$ ), it is presently unclear whether age may have influenced these endocrine endpoints; age data are not yet available. The present study reveals a significant negative correlation between the expressions of AhR and GR in the outer blubber and PCB concentrations (see Figure 1 a, b). Biological effects of dioxin-like PCBs are believed to be mediated essentially via the aryl hydrocarbon receptor. A negative correlation between dioxin and AhR expression has been observed in laboratory rats (Roman *et al.*, 1998; Pollenz *et al.*, 1998), as well as a negative trend for GR was reported in arctic char after exposure to PCB metabolites (Aluru *et al.*, 2004). Further research over the coming months will enable us to better characterize the factors underlying these observations.

## PCBs and PBDEs

The PCB (206 congeners) and PBDE (23 congeners) concentrations in 2008 whales were slightly lower than those observed in 2007 (only males reported). PCBs averaged 3541 ng/g lw  $\pm$  1788 (in 2007 3638 ng/g lw  $\pm$  1409) and PBDEs averaged 17.9 ng/g lw  $\pm$  7.6 (in 2007 20.6 ng/g lw  $\pm$  9.8). *Note, in the 2007 synopsis report data presented in wet weight.* Similar to 2007, concentrations of PCBs and PBDEs showed a significant positive relationship ( $r^2 = 0.6$ ;  $p < 0.0001$ ). PCB and PBDE relationships with length were weaker in 2008 (PCBs:  $r^2 = 0.25$ ;  $p = 0.025$ ; PBDEs:  $r^2 = 0.002$ ;

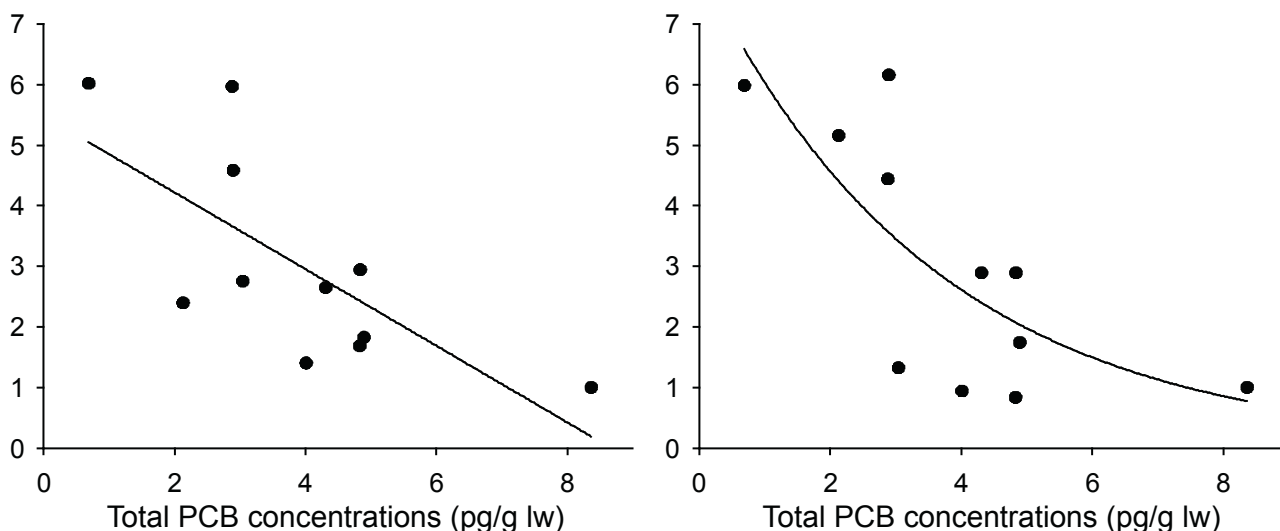


Figure 1: The expressions of AhR (1a, left) and GR (1b, right) decrease with increasing PCB concentrations in beluga whales ( $r^2 = 0.62$ ,  $p = 0.003$ ; and  $r^2 = 0.43$ ,  $p = 0.027$ ; respectively).

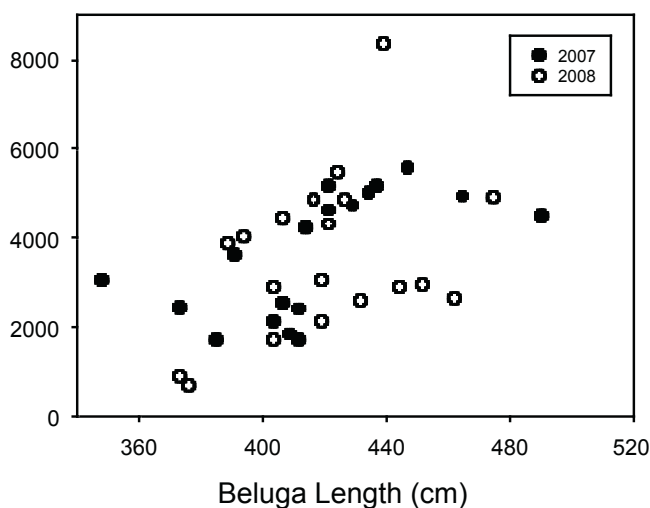


Figure 2: Total PCB concentrations correlated with total body length in belugas, likely due to a combination of age and feeding ecology.

$p = 0.4$ ) than those observed in 2007 (PCBs:  $r^2 = 0.4$ ,  $p = .002$ ; PBDEs  $r^2 = 0.3$ ,  $p = 0.002$ ) (see Figure 2).

An opportunity to sample both mother and a fetus demonstrates the transplacental transfer of PCBs and PBDEs. PCBs doubled in concentration in the fetus relative to the mother, whereas the reverse was true for PBDEs. These observations, coupled with an evaluation of pattern changes from mother to fetus, reflect the physico-chemical barrier at

the level of the placenta. The PCB concentration in the mother (208.5 ng/g lw) was less than half of that in the fetus (557.8 ng/g lw) that was dominated by heavier PCB congeners (hepta- to deca- PCBs). The reverse trend was observed for PBDEs whereby the mother had double the concentration (2.1 ng/g lw) than the fetus (1.03 ng/g lw). The fetus PBDE profile was dominated by tetra congeners with little contribution from heavier congeners.

#### PCB and PBDE Metabolites

The hydroxy- and methyl-sulfone-polychlorinated biphenyls (OH- and MeSO<sub>2</sub>-PCB) fell below the detection limit to be quantifiable in 17 samples. However the hydroxy-polybrominated diphenyl ethers were measurable in 15 of the 17 samples, averaging 1.54 ng/g ww (+/- 1.3 ng/g ww). This appears unusual, given the much higher concentrations of parent PCB compounds relative to PBDEs. No trends were observed with length of beluga or the two size groups observed for the 2007 belugas. Overall the low levels are consistent with the findings reported for these contaminants in beluga from western Hudson Bay and the St. Lawrence estuary (McKinney *et al.* 2006). Although MeSO<sub>2</sub>-PCBs were measured in blood here, they are known to be largely sequestered in the liver with blood levels being very low as shown in polar bears (Gebbink *et al.* 2008a, 2008b).

## Diet and Condition

### Stable Isotopes

Mean stable isotope values were similar to 2007, but relationships with beluga length were much weaker in 2008 (Table 1). The only significant trend with length was observed for liver  $\delta^{13}\text{C}$  that suggests larger males feeding in the offshore and smaller whales in the nearshore, similar to previous reports (Loseto et. al., 2008).

### Lipid Analysis

Blubber and liver samples were extracted for lipid class and fatty acids analysis.

Lipid classes analysis of blubber revealed a consistent domination of triglycerides, the major energy storage lipid, with trace amounts of phospholipids. Fatty acid analysis on blubber and liver are underway at FWI.

### Vitamin A

Vitamin A was significantly higher in 2008 belugas relative to 2007 ( $p = 0.004$ , only males evaluated). In 2007 the positive trend observed with length and PCBs supported dietary sources to be a driving factor of trends rather than a toxic effect on hormonal endpoints. The 2008 results continue to show a positive trend with PCBs; however there was no relationship with length (see Figure 3). Results for liver and serum vitamin A are also presented and related to length and PCB concentrations. Vitamin A in serum positively related to liver ( $r = 0.26$ ;  $p = 0.04$ ) and blubber ( $r = 0.5$ ;  $p = 0.04$ ), however liver and blubber were not significantly related.

**Table 1. Mean carbon and nitrogen stable isotopes (per mil) for beluga liver and muscle samples. Correlations determined for beluga length trends for carbon and nitrogen isotopic values in beluga liver and muscle.**

	$\delta^{15}\text{N}$	Std Dev	Length Trend	$\delta^{13}\text{C}$	Std Dev	Length Trend
2007						
liver	17.71	0.76	$r = 0.5$ ; $p=0.04$	-20.2	0.57	$r = -0.6$ ; $p=0.004$
muscle	16.54	0.63	$r = 0.1$ ; $p=0.6$	-18.44	0.38	$r = -0.4$ ; $p=0.2$
2008						
liver	16.85	1.20	$r = 0.05$ ; $p=0.8$	-18.76	0.93	$r = -0.5$ ; $p=0.02$
muscle	16.85	1.07	$r = 0.3$ ; $p=0.1$	-18.00	0.90	$r = 0.3$ ; $p=0.1$

### Vitamin A reported for males only.

	Vitamin A	Std Dev	Length Trend	PCB trend	n
2007					
Blubber (ng/g)	24.6	9.9	$r = 0.7$ ; $p=0.015$	$r = 0.7$ ; $p=0.001$	18
2008					
Blubber (ng/g)	32.30	11.60	$r = -0.1$ ; $p=0.9$	$r = 0.44$ ; $p=0.06$	18
Liver (ug/g)	552.06	394.04	$r = 0.1$ ; $p=0.9$	$r = 0.2$ ; $p=0.4$	16
Serum (ug/dl)	43.33	12.91	$r = 0.2$ ; $p=0.9$	$r = 0.5$ ; $p=0.04$	15

Fetus Vitamin A blubber: 3.3 ng/g

Females Vitamin A blubber: 28.5 ng/g

Females Vitamin A serum: 25 ug/dl

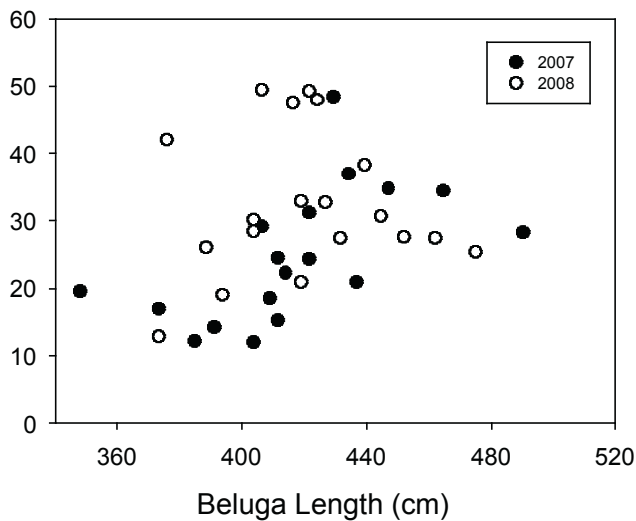


Figure 3: Vitamin A in males. Trend with body length was only significant in 2007.

### Thyroid Hormones

Thyroid hormones were measured in serum. Free circulating T3 and T4 were determined along with total T3 and T4 in serum samples. All were positively related to one another ( $r > 0.8$ ,  $p < 0.001$ ). Thyroid levels revealed no significant relationships with beluga length or contaminants. Both free and circulating thyroids had significant positive trends with vitamin A in serum and blubber (see

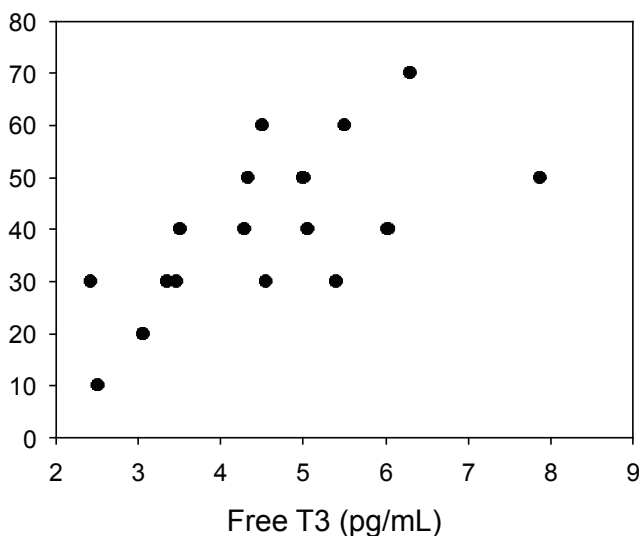


Figure 4: The dietary hormone Vitamin A correlated with thyroid hormones in circulation, underscoring the tight physiological regulation of these co-transported compounds.

Figure 4). These early results might suggest that contaminant concentrations are not high enough to disrupt thyroid hormone homeostasis, but a small sample size, and considerable confounding influences (age, sex, and condition) may have masked our preliminary assessment.

### Cholesterol

Both vitamin E and A are fat soluble vitamins but play different roles in maintenance of health. Vitamin A – retinoids are important for vision and metabolism, gene transcription, and vitamin E is known for its prevention of lipid oxidation and propagation of free radicals. The positive relationship between cholesterol and the lipophilic vitamin E observed in our belugas (see Figure 5) has been observed in humans (Cham *et al.*, 1998). We will further investigate these trends with other hormones and toxic endpoints, with the ultimate aim of characterizing the health of free-ranging belugas.

### Conclusion

Our preliminary results demonstrate the value of genomics, nutrient analysis and endocrine measurements in characterizing the health of free-ranging beluga whales. Additional sampling and analysis will shed light on the nature of factors

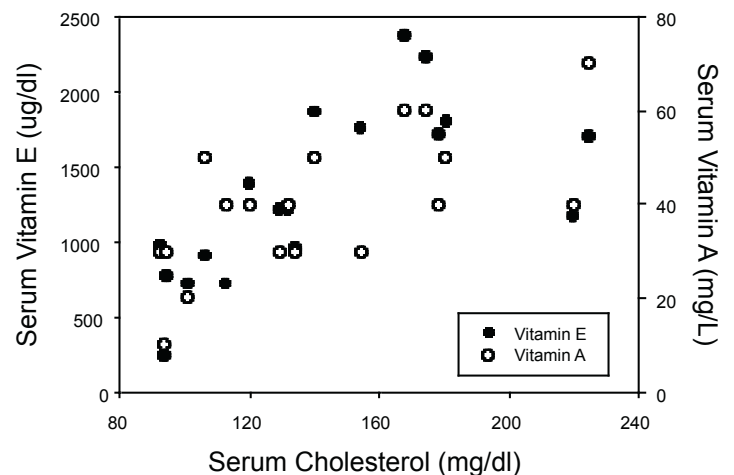


Figure 5: Vitamin E correlated with cholesterol in beluga serum, with both endpoints providing insight into the condition of the sampled animals.

underlying variability in these observations. While such natural factors as age and sex underlie some of the variation, preliminary results suggest that differences in feeding ecology and contaminant concentrations also play a role.

## Expected Project Completion Date

**Phase 1:** Pilot study 2007-08: Field sampling and initial methods development.

**Phase 2:** Research effort 2008-09: second year of sampling, dedicated methods development and initial round of analyses. Partnership with IPY and ArcticNET.

**Phase 3:** Research effort 2009-10 (underway): Complete development and validation of methods; conduct a 3<sup>rd</sup> year of sampling; complete health measurements on existing samples; model climate scenarios; begin dedicated publishing.

**Phase 4:** Reporting out efforts 2010-11: complete publications in international scientific literature; carry out dedicated outreach efforts in multiple communities.

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# Trophodynamics of some BFRs, PFCs, current use pesticides and legacy persistent organic pollutants in a marine food web from Cumberland Sound

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## Abstract

This project is aimed at understanding the critical initial processes involved in the bioaccumulation and trophic transfer of some chemicals of emerging concern in an eastern Arctic marine food web from Cumberland Sound (Nunavut). The compounds of interest include the fluorine-based surfactants, the bromine-based flame retardants, a suite of organochlorines (OCs) including PCBs and current use pesticides. Particular emphasis was placed on understanding the trophodynamics of target compounds in the lower food web. The samples that make up part of the food web include: beluga, narwhal, ringed and harp seals, Greenland shark, turbot, char, thorny skate, herring, sculpin, capelin, mixed zooplankton, brown amphipod, scallop and snails. Stable isotopes ( $\delta^{15}\text{N}$  and  $\delta^{13}\text{C}$ ) and fatty acid analyses are ongoing with the hope that it

## Résumé

Ce projet vise à comprendre les processus critiques initiaux participant à la bioaccumulation et au transfert trophique de certains produits chimiques qui constituent une préoccupation émergente dans le réseau alimentaire marin de l'est de l'Arctique de la baie de Cumberland (Nunavut). Les composés qui nous intéressent comprennent les surfactants à base de fluor, les ignifugeants bromés, un ensemble de composés organochlorés (CO), y compris les BPC et les pesticides d'usage courant. Une emphase particulière a été accordée à la compréhension de la trophodynamique des composés cibles dans le bas du réseau trophique. Les échantillons qui composent une partie du réseau trophique comprennent : le béluga, le narval, les phoques annelés et du Groenland, le requin du Groenland, le turbot, l'omble chevalier, la raie épineuse, le hareng, le

will help construct the relative trophic status of biota. In the top trophic level (TL) animals, total ( $\Sigma$ ) concentrations (geometric mean) of the perfluorinated carboxylic acids (PFCA: sum of  $C_8$ - $C_{12}$ ) in liver ranged from 43.1 ng/g (ww) in beluga to 5.2 ng/g (ww) in harp seal. In general, PFUA ( $C_9$ ) was the dominant PFCA in these animals except in Greenland shark where PFDoDa ( $C_{12}$ ) dominated. In the two pinnipeds species,  $\Sigma_5$ PFCA was smaller than that of perfluorooctane sulfonate (PFOS); the converse was true for the cetaceans and Greenland shark. PFOS concentrations in cetaceans were *ca.* 2 and 3× greater than that of the PFOS neutral precursor, perfluorooctane sulfonamide (PFOSA). Concentrations of PFOSA in ringed and harp seals were *ca.* 3 ng/g (ww), while PFOS concentrations were 11.1 and 10.5 ng/g (ww), respectively. In the lower TL animals, the polychlorinated biphenyls (PCBs) were the dominant OC measured. Wet weight tissue-based concentrations of  $\Sigma$ PCBs (sum of *mono* to *deca*-congeners) ranged from 18.5 ng/g in turbot to 1.7 ng/g in sculpin. Toxaphene was the second most abundant OC; the rank order of  $\Sigma$ toxaphene (sum of 35 congeners) in the lower TL animals was turbot > herring > char > capelin > thorny skate  $\approx$  scallop > sculpin. DDT, chlordane and hexachlorocyclohexane were all detectable in the lower TL animals. Endosulfan I and II, endosulfan sulfate, endrin, pentachloronitrobenzene and dacthal were also measurable in lower TL animals in our food web. With a few exceptions, the concentrations of these pesticides were in the 100 pg/g (ww) range. Chlorothalonil was undetectable in the lower TL animals.

chabot, le capelan, le zooplancton mixte, l'amphipode brun, les pétoncles et les bigorneaux. Les analyses des isotopes stables ( $\delta^{15}\text{N}$  et  $\delta^{13}\text{C}$ ) et des acides gras se poursuivent dans l'espoir qu'elles aideront à comprendre l'état trophique relatif du biote. Parmi les animaux de niveau trophique supérieur, les concentrations totales ( $\Sigma$ ) (moyenne géométrique) et les acides carboxyliques perfluorés (PFCA : somme de  $C_8$ - $C_{12}$ ) dans le foie variaient de 43,1 ng/g (ww) chez le béluga à 5,2 ng/g (ww) chez le phoque du Groenland. En général, le PFCA dominant chez ces animaux était le PFUA ( $C_9$ ), à l'exception du requin du Groenland chez qui dominait le PFDoDa ( $C_{12}$ ). Dans deux espèces de pinnipèdes, les concentrations totales  $\Sigma_5$ PFCA étaient inférieures aux perfluorooctanesulfonates (PFOS); le contraire était vrai chez les cétacés et le requin du Groenland. Les concentrations de PFOS chez les cétacés étaient environ 2 et 3 fois supérieures par rapport au précurseur neutre de PFOS, le perfluorooctane sulfonamide (PFOSA). Les concentrations de PFOSA chez les phoques annelés et du Groenland s'élevaient à environ 3 ng/g (ww), tandis que les concentrations de PFOS s'élevaient à 11,1 et à 10,5 ng/g (ww), respectivement. Parmi les animaux de niveau trophique inférieur, les diphényles polychlorés (BPC) constituaient le CO mesuré dominant. Les concentrations totales dans les tissus à poids frais de  $\Sigma$ BPC (somme des *mono* et *déca*-congénères) variaient de 18,5 ng/g chez le turbot à 1,7 ng/g chez le chabot. La toxaphène constituait le deuxième CO le plus abondant; l'ordre de classement du total des toxaphènes (somme de 35 congénères) parmi les animaux de niveau trophique inférieur était le suivant : turbot > hareng > omble chevalier > capelan > raie épineuse  $\approx$  pétoncles > chabot. Le DDT, le chlordane et l'hexachlorocyclohexane pouvaient tous être détectés chez les animaux de niveau trophique inférieur. L'endosulfan I et II, le sulfate d'endosulfan, l'endrine, le pentachloronitrobenzène et le Dacthal étaient également mesurables chez les animaux de niveau inférieur dans notre réseau trophique. Hormis quelques exceptions, les concentrations de ces pesticides étaient de l'ordre de 100 pg/g (ww). Le chlorothalonil n'était pas détectable chez les animaux de niveau trophique inférieur.

## Key Messages

- In beluga, narwhal and Greenland shark,  $\Sigma_5$ PFCA were greater than that of PFOS and PFOSA;

## Messages clés

- Chez le béluga, le narval et le phoque du Groenland, les concentrations totales  $\Sigma_5$ PFCA

- 
- In pinnipeds, concentrations of PFOS were greater than that of its neutral precursor, PFOSA; the converse was true for cetaceans;
  - Polychlorinated biphenyls was the dominant organochlorine measured in the lower trophic level animals: wet weight tissue based concentrations ranged from 18.5 in turbot to 2.1 ng/g in thorny skate;
  - With the exception of chlorothalonil, all the chemicals that make up our suite of current use pesticides (endosulfane I and II, endosulfan sulfate, endrin, pentachloronitrobenzene and dacthal) were detectable in the lower trophic level animals.
- étaient supérieures aux concentrations de PFOS et de PFOSA.
  - Chez les pinnipèdes, les concentrations de PFOS étaient supérieures à celles de son précurseur neutre, le PFOSA; le contraire se vérifiait chez les cétacés.
  - Les diphényles polychlorés constituaient le composé organochloré dominant mesuré chez les animaux de niveau trophique inférieur : les concentrations dans les tissus à poids frais variaient entre 18,5 ng/g chez le turbot et 2,1 ng/g chez la raie épineuse.
  - À l'exception du chlorothalonil, tous les produits chimiques qui composent notre éventail de pesticides d'usage courant (endosulfan I et II, sulfate d'endosulfan, endrine, pentachloronitrobenzène et Dacthal) pouvaient être décelés chez les animaux de niveau trophique inférieur.
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## Objectives

- To determine the levels of new and emerging contaminants including suite of per- and poly-fluorinated compounds (PFCs), bromine- and chlorine-based flame retardants, current use pesticides and legacy POPs in an archived marine food web from Cumberland Sound;
- Using stable isotopes ( $\delta^{15}\text{N}$  and  $\delta^{13}\text{C}$ ) and fatty acid profiling as tools to aid our understanding of the mechanisms and processes of bioaccumulation of these contaminants particularly at lower trophic levels and transfer to upper trophic level species.

## Introduction

There are few studies that have examined the trophodynamics of new and emerging compounds and current use pesticides (CUPs) in Arctic marine food webs. The difficulty in sampling in remote regions is likely one factor for this paucity. Wolkers *et al.* (2005) reported on the congener-specific accumulation of brominated diphenyl ethers (BDEs) in two Norwegian Arctic food chains; the first food chain consisted of polar cod-ringed seal-polar bear and the other polar cod-beluga whale(1). In another study, Sømro *et al.* (2006) investigated the biomagnification of BDEs and

hexabromocyclododecane (HBCD) in a Norwegian Arctic marine food chain consisting of four invertebrates, polar cod, ringed seals and polar bears(2). Morris *et al.* (2007) assessed the distribution of BDEs, HBCD and CUPs in a Canadian Arctic food chain consisting of water, zooplankton, phytoplankton, arctic cod and ringed seals from Barrow Strait (Nunavut)(3). Using  $\delta^{15}\text{N}$  to assess relative trophic status of biota, Tomy *et al.* recently reported on the extent of trophic transfer of the some BFRs and PFCs in components of food webs from the eastern and western Canadian Arctic(4, 5).

It could be argued that one of the gaps in Arctic food web studies examined to date is the lack of critical components of the food web. In particular, emphasis on accumulation at the lower level of the food web is not well characterized yet important because bioaccumulation processes occurring at this level tend to drive contaminant concentrations at higher trophic levels.

The aim of this study is to examine the behavior of target compounds in a more thorough and well characterized food web from Cumberland Sound (Nunavut). Stable isotopes ( $\delta^{15}\text{N}$  and  $\delta^{13}\text{C}$ ) and fatty acid profiling are being used to construct the relative trophic structure and establish trophic links of biota and to complement contaminant analyses.

## Activities in 2008/2009

### *Samples investigated:*

The animals investigated in this study were collected from the Cumberland Sound include: beluga ( $n=6$ ), narwhal ( $n=7$ ), ringed ( $n=6$ ) and harp ( $n=4$ ) seals, Greenland shark ( $n=11$ ), char ( $n=5$ ), turbot ( $n=8$ ), thorny skate ( $n=6$ ), herring ( $n=1$ ), sculpin ( $n=9$ ), capelin ( $n=5$ ), scallops ( $n=7$ ), snails ( $n=2$ ), brown amphipod ( $n=1$ ) and mixed zooplankton ( $n=6$ ). Animals were collected in 2007 by various PIs on the project and stored at  $-30^{\circ}\text{C}$ . Good laboratory practice was adopted when handling sample tissues during field collections, storage and sub-sampling.

### *Chemical analysis:*

Analytical methods adopted for our study have already been published in the peer-review literature. All chemicals of interest except for the fatty acids were analyzed in the toxic aquatic chemical laboratory at Fisheries & Oceans Canada, Winnipeg. Fatty acids and stable isotope of nitrogen were analyzed at the Great Lakes Institute for Environmental Research. The hydrophobic chemicals like BDEs, HBCD, and CUPs were analyzed in the blubber of the marine mammals and in the muscle tissue of the other animals as described previously. BDEs were analyzed by gas chromatography with electron capture negative ion (ECNI) low resolution mass spectrometry. Quantification of BDE congeners were done using external standard solutions. Total BDE congeners are based on the sum of -47, -85, -99, -100, -153 and -154. Isomers of HBCD were analyzed by liquid chromatography tandem mass spectrometry in the negative electrospray ionization mode. Total HBCD is based on the sum of the  $\alpha$ - and  $\gamma$ -diastereoisomers. SCCPs were analyzed by high resolution mass spectrometry. The suite of CUPs which include, chlorothalonil, dacthal, endosulfan I and endosulfan sulfate were analyzed by GC and detection based on electron capture. Quantification of CUPs was done by the external standard method. Fluorinated compounds were extracted and analyzed in the liver of the marine mammals and in whole-homogenates of the invertebrates as described in Tomy *et al.* (6) When liver samples were unavailable, muscle tissues was analyzed. Compounds investigated include perfluorooctanoate ( $\text{C}_8$ : PFOA), perfluorononanoate ( $\text{C}_9$ : PFNA), perfluorodecanoate ( $\text{C}_{10}$ : PFDA),

perfluoroundecanoate ( $\text{C}_{11}$ : PFUA), perfluorodecanoate ( $\text{C}_{12}$ : PFDoDA), perfluorooctane sulfonate (PFOS) and perfluorooctane sulfonamide (PFOSA). Total perfluorinated carboxylate concentrations are based on the sum of  $\text{C}_8$ - $\text{C}_{12}$ .

### *Quality assurance:*

Certified reference materials for BDEs in beluga blubber from the National Institute of Standards and Technology (SRM 1945) and Wellington Certified Reference material for fish tissue (WMF-01) were used for each batch of 20 samples. The agreement between our measured BDE concentrations and the accepted SRM-1945 and WMF-01 values were very good. No reference materials are yet available for HBCD, CUPs or for the fluorinated compounds. Our laboratory also participates in the NCP Quality Assurance Program.

### *Statistical analyses:*

For calculating arithmetic and geometric means, non-detect concentrations were replaced with half the method detection limit. Statistical treatment of the data was done using SigmaStat. The Q-test was used to remove outliers in the data-set.

## Results

Concentrations of chemicals examined in this study are presented in Tables 1 and 2.

### *PFCs in top trophic level (TL) animals.*

The rank order of  $\Sigma_5$ PFCAs were beluga  $\approx$  Greenland shark > narwhal > ringed seal > harp seal. In general, PFUA concentrations were greater than any other PFCA in these TL animals except in Greenland shark where PFDoDa was the dominant PFCA. Interestingly, PFOS concentrations were greater than  $\Sigma_5$ PFCAs concentrations in pinnipeds; the converse was true for the cetaceans and Greenland shark. PFOS concentrations exceeded those of its neutral-precursor, PFOSA, in whales and Greenland shark. In ringed and harp seals, liver based PFOSA concentrations were *ca.* 3 ng/g (wet weight) which was 3 $\times$  smaller than PFOS concentrations. On the other hand, PFOS concentrations in narwhal and beluga were *ca.* 2 and 3 $\times$  greater than PFOSA, respectively. We hypothesize that this discrepancy, which has been observed by other researchers, is likely due to the differences in the metabolic capabilities between pinnipeds and cetaceans (6, 7).



**Table 1. Liver-based concentrations (ng/g, wet weight) of PFCs in top trophic level animals from CumberSound<sup>a</sup>.**

Species	n		PFCA	PFOS	PFOSA
Beluga	6	GM	43.08	9.43	26.03
		min	38.07	7.92	18.21
		max	47.60	11.40	40.87
Narwhal	7	GM	22.21	8.87	17.17
		min	11.71	6.24	7.52
		max	50.78	14.05	37.95
Harp seal	4	GM	5.28	10.54	3.02
		min	2.93	6.99	1.54
		max	12.78	16.63	6.27
Ringed seal	6	GM	7.50	11.13	3.07
		min	2.18	6.67	2.25
		max	23.40	29.15	6.27
Greenland shark	10	GM	39.59	3.21	20.10
		min	17.76	nd	nd
		max	110.79	4.97	44.10

<sup>a</sup> GM = geometric mean, min = minimum value, max = maximum value.

OCs in lower tropic level animals. PCBs were the most dominant OCs in lower TL animals. The rank order of  $\Sigma$ PCBs (sum of *mono-* to *deca*-congeners) in the animals was turbot > herring  $\approx$  capelin > char > scallop > thorny skate  $\approx$  sculpin. Toxaphene was the second most dominant OC with  $\Sigma$ toxaphene (sum of 35 congeners): concentrations in herring (9.91 ng/g, ww) similar to that of  $\Sigma$ PCBs (12.07 ng/g). In turbot,  $\Sigma$ toxaphene concentrations (12.23 ng/g) were only *ca.* 1.5 $\times$  smaller than  $\Sigma$ PCBs (18.54 ng/g). Concentrations of  $\Sigma$ DDT (sum of *o,p* and *p,p*-DDT and DDE- and DDD-metabolites) were greatest in turbot (9.13 ng/g), followed by herring (3.99 ng/g) and char (2.61 ng/g). Smaller concentrations of  $\Sigma$ DDT were measured in capelin (0.93 ng/g), scallop (0.26 ng/g), sculpin (0.14 ng/g) and thorny skate (0.32 ng/g). Chlordane was also detectable in all the lower TL animals analyzed: concentrations of  $\Sigma$ chlordanes (sum of 6 compounds) ranged from 7.92 ng/g in turbot to 0.09 ng/g in sculpin. Concentrations of  $\Sigma$ chlorobenzenes (sum of *tetra-* to *hexa-*) and  $\Sigma$ hexachlorocyclohexanes (sum of  $\alpha$ -,  $\beta$ -, and

**Table 2. Lipid content (%) and tissue-based wet weight concentrations (ng/g) of a suite of organochlorine compounds in lower trophic level animals from CumberSound.**

	% lipids	$\Sigma$ CBZ <sup>1</sup>	$\Sigma$ HCH <sup>2</sup>	$\Sigma$ CHLOR <sup>3</sup>	$\Sigma$ DDT	$\Sigma$ PCBs	$\Sigma$ Tox <sup>4</sup>	Dieldrin	Endo1 <sup>5</sup>	Endo2 <sup>6</sup>	ES <sup>7</sup>	Endrin	PCNB <sup>8</sup>	Dacthal
Capelin (n=4)	2.79	1.60	0.45	1.76	0.93	10.72	2.79	2.72	0.08	0.01	0.13	0.41	0.15	0.05
Herring (n=1)	2.15	0.97	0.58	2.78	3.99	12.07	9.91	0.17	0.08	0.02	0.05	0.12	0.07	0.28
Scallop (n=4)	0.18	0.26	0.08	0.11	0.26	4.13	0.14	0.05	0.02	0.03	0.14	0.03	0.07	0.04
Sculpin (n=4)	0.23	0.04	0.09	0.09	0.14	1.66	0.07	0.03	0.01	0.01	0.05	0.01	0.04	0.01
T. Skate (n=4)	0.10	0.12	0.09	0.12	0.32	2.10	0.15	0.15	0.03	0.01	0.37	0.07	0.03	0.01
Turbot (n=2)	11.46	1.78	2.38	7.92	9.13	18.54	12.23	3.26	0.02	0.02	0.04	1.25	0.08	0.02
Char (n=2)	2.79	1.12	0.94	2.68	2.61	7.47	3.90	0.94	0.02	0.01	0.01	0.55	0.05	0.01

<sup>1</sup> CBZ = chlorobenzenes; <sup>2</sup> HCH = hexachlorocyclohexanes; <sup>3</sup> CHLOR = chlordanes; <sup>4</sup> Tox = toxaphenes; <sup>5</sup> Endo1 = endosulfan I;

<sup>6</sup> Endo2 = endosulfan II; <sup>7</sup> ES = endosulfan sulfate; <sup>8</sup> PCNB = pentachloronitrobenzene



$\gamma$ -isomers) were greatest in turbot, 1.78 and 2.38 ng/g, respectively, relative to the other lower TL animals.

With the exception of chlorothalonil, all the other CUPs examined in this study were detectable in the lower TL animals. In general, concentrations of endosulfan I and II were below 100 pg/g (ww) in all the animals. Endosulfan sulfate concentrations ranged from 370 pg/g (ww) in thorny skate to 10 pg/g in char. The rank order of endrin in the lower TL animals was turbot >> char > capelin > herring > thorny skate > scallop > scuplin. Pentachloronitrobenzene (PCNB) concentrations were greatest in capelin (150 pg/g, ww); concentrations of PCNB in the other animals ranged from 30 pg/g (ww) in thorny skate to 80 ng/g (ww) in turbot. Dacthal concentrations were similar in scuplin, thorny skate, turbot and char (10-20 pg/g, ww) and much smaller than that measured in herring (280 pg/g, ww)

## Activities That Are Ongoing

There are still gaps in our dataset that we hope to fill over the next few months. Stable isotope and fatty acid analysis on the samples are on-going. All the samples ( $n=84$ ) have been analyzed for BFRs (BDEs and HBCD) but interpretation of chromatograms still needs to be done.

Chromatograms of the marine mammals for PCBs and CUPs and lower trophic levels animals for PFCs all need to be interpreted.

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# Bioaccumulation of Perfluorinated Compounds in the Vegetation-Caribou-Wolf Food Chain

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## Abstract

This project studied the bioaccumulation of perfluorinated compounds in a northern food-chain: vegetation-caribou-wolves. These compounds are emerging contaminants of concern in the Arctic, and they have been found in relatively high concentrations in caribou from the Canadian north.

Traditional knowledge collected through elder interviews for this project shows the strength of the vegetation-caribou-wolf food chain, and provided a wealth of information about the inter-relatedness of caribou and wolves. Stable isotope analysis of vegetation, caribou and wolf tissues showed that caribou had been feeding mainly on lichen and wolf mainly on caribou.

Our results show that highest PFC concentrations were found in the liver of wolves. The liver concentrations of caribous showed a similar pattern

## Résumé

Ce projet visait à étudier la bioaccumulation de composés perfluorés (CPF) dans la chaîne alimentaire du Nord : végétation-caribou-loup. Ces composés sont des contaminants qui commencent à devenir une préoccupation dans l'Arctique et ils ont été trouvés dans des concentrations relativement élevées chez les caribous du Nord canadien.

Les connaissances traditionnelles recueillies en interviewant les aînés dans le cadre de ce projet révèlent la force de la chaîne alimentaire végétation-caribou-loup et ont permis d'obtenir de précieux renseignements sur l'interdépendance entre les caribous et les loups. Une analyse des isotopes stables de la végétation et des tissus des caribous et des loups montre que les caribous se nourrissent essentiellement de lichens tandis que les loups se nourrissent essentiellement de caribous.

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of PFCs with approximately 2-3 times lower concentrations. In vegetation relatively high levels were found in willow. Lichen had higher amounts than the other two plant species, willow and cotton grass. Biomagnification of the PFCs was seen in the lichen-caribou-wolf food chain.

## Key Messages

- Perfluorinated compounds (PFCs) are accumulating in terrestrial food chains, as demonstrated in the vegetation-caribou-wolf chain.
- Lichens and willows contain much higher concentrations of PFCs than grasses and sedges.
- Traditional knowledge strengthens the choice of vegetation-caribou-wolf as a model for northern terrestrial food webs.

Nos résultats montrent que les concentrations les plus élevées de CPF se trouvent dans le foie des loups. Les concentrations dans le foie des caribous révélaient la présence de CPF mais à des concentrations environ deux à trois fois inférieures. Dans la végétation, des concentrations relativement élevées ont été relevées dans les saules. Les lichens contenaient des quantités supérieures aux deux autres espèces de plantes, soit les saules et la linaigrette. La bioamplification des CPF a été observée dans la chaîne alimentaire lichen-caribou-loup.

## Messages clés

- Les composés perfluorés (CFP) s'accumulent dans les chaînes alimentaires terrestres, comme le montre la chaîne alimentaire végétation-caribou-loup.
- Les lichens et les saules contiennent des concentrations de CFP nettement plus élevées que les herbes et les laïches.
- Les connaissances traditionnelles renforcent le choix de la chaîne alimentaire végétation-caribou-loup comme modèle pour les réseaux trophiques terrestres du Nord.

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## Objectives

- To determine whether perfluorinated compounds bioaccumulate using the vegetation-caribou-wolf food chain as a model.
- To determine whether the diet of the Porcupine caribou herd has changed over the past decade, which could be an indicator of climate change.
- To compare the ability of stable isotope analysis and Quantitative Fatty Acid Signature Analysis to predict the diet of a terrestrial ungulate.
- To incorporate traditional knowledge into the interpretation, discussion, and future planning.

## Introduction

Perfluorinated compounds (PFCs) are a family of industrial chemicals used in wide variety of applications such as soil and stain repellents, paper coatings and fire fighting foams. Since the early millennium it is known that perfluorooctane

sulfonate (PFOS), perfluorooctanoate (PFOA) and other members of the family are widely distributed in the environment as well as in humans [Giesy et al. 2002]. There is still considerable debate on how they reach remote regions such as the Arctic. Two mechanisms of distribution have been proposed: Aquatic transport by oceanic currents and atmospheric transport of precursor substances, which are subsequently degraded to perfluoroalkyl acids as well as the acids substances themselves.

Studies on bioaccumulation potentials have mainly focused on marine food webs where the concentrations in top predators were high due to the long food chains. However, for marine environments it is uncertain whether the aquatic and/or atmospheric sources are most relevant. Only a few Arctic terrestrial mammals have been studied. Surprisingly high PFOS concentrations were found in Arctic fox (*Vulpes lagopus*) livers [Martin et al. 2004], probably due to the opportunistic diet that includes marine mammals. Even northern caribou

(*Rangifer tarandus*) were found to have measurable concentrations of PFOS in liver and muscle [Tittlemier et al. 2006; Ostertag et al. 2008].

In this study a terrestrial food web in northern Canada that included vegetation (plants and lichens), barren ground caribou (*Rangifer tarandus groenlandicus*) and wolves (*Canis lupus*) was studied. The lichens reflect direct atmospheric input of PFCs, because they lack roots and receive their nutrients from the atmosphere. Caribou is adapted to eat lichens, which constitute a large part of its diet. Wolves represent one of the top predators in the ecosystem, and their staple prey is caribou. This setup gives the unique opportunity to examine the importance of atmospheric transport alone as well as evidence for food web biomagnification.

The vegetation–caribou–wolf chain is a simple yet very good model to test bioaccumulation in terrestrial food webs. It is well-separated in trophic levels. The vegetation, in this context, contains both plants and lichens; while plants obtain most of their nutrients through root systems, lichens absorb their nutrients from air. Hence the vegetation in this model enables to differentiate between direct atmospheric sources and accumulation from soils. More than half of the caribou winter diet comprises of lichen, while more than half of its summer diet is comprised of sedges. Hence caribou is uniquely suited to model bioaccumulation from different vegetation types. Finally, northern wolves’

diet consists mainly of caribou. Traditional knowledge collected for this study further shows that wolves eat preferential parts of caribou, specifically liver, which exposes wolves to higher levels of contaminants such as PFCs which are associated with proteins.

## Activities in 2008/2009

### Sample Collection

Lichens, cotton grass, aquatic sedge, and willow leaf samples were taken in summer 2008 at different locations in the northern Yukon. Table 1 lists the vegetation samples that were collected. Caribou samples were collected in 2006 and 2007 from the Porcupine herd in the Yukon and from the Bathurst herd in Northwest Territories and consisted of muscle, kidney and liver. Wolf liver and muscle samples were obtained from animals collected in the same region as the caribou. All caribou and wolf sampling was done by local hunters and trappers as part of traditional hunts.

The plant and lichen samples were freeze dried and homogenized within the collection bags. 1 g of vegetation was accurately weighed into a 50 ml polypropylene centrifuge tube and extracted by shaking with methanol. The tube was centrifuged at 4000 rpm for 5 min and decanted into a clean polypropylene centrifuge tube and the residue was re-extracted twice. Each sample was spiked

**Table 1: Species and type of vegetation collected in the northern Yukon (summer 2008)**

Species	type	Location				
		Chapman Lake	Eagle Plains	North Slope	Old Crow	Rock River
<i>Carex aquatilis</i>	Sedge	X		X		
<i>Cladina mitis</i>	Sedge	X				
<i>Cladina rangiferina</i>	Lichen		X		X	X
<i>Eriophorum angustifolium</i>	Sedge			X		
<i>Eriophorum vaginatum</i>	Sedge	X	X	X		X
<i>Flavocetraria cucullata</i>	Lichen	X			X	X
<i>Flavocetraria nivalis</i>	Lichen		X	X		
<i>Salix glauca</i>	Shrub			X		
<i>Salix pulchra</i>	Shrub	X	X		X	
Soil	Soil	X	X	X	X	X

with recovery standards containing  $^{13}\text{C}$ -PFOA,  $^{13}\text{C}$ -PFNA and  $^{13}\text{C}$ -PFOS. This process was repeated twice more, combining the extracts. The 20 ml extract was then concentrated to 1 ml and applied to a 100 mg carbon solid phase extraction cartridge. The sample was eluted with methanol and the eluant was then taken to dryness and reconstituted in 1 ml of 50/50 methanol/water

The liver and muscle samples (0.2–0.3 g) were analysed according to the method of Hansen et al. (2001) using methyl tert-butyl ether (MTBE) and tetrabutylammonium (TBA) hydrogen sulfate.

Analysis of perfluorinated acids was conducted by liquid chromatography (LC) with negative ion electrospray tandem mass spectrometry (LC-MS/MS) using an Agilent 1100 pumping system coupled to a mass spectrometer (API 4000, Applied Biosystems- MDS Sciex). The final concentration in biota was reported per g of wet tissue and for plant samples, per gram of dry plant matter.

Method detection limits ( $3 \times \text{SD}$  of blank values) for 1 g samples ranged from 0.001 ng/g for PFNA to 0.04 ng/g for PFOA.

#### Stable isotope analysis

Stable isotope analysis for C and N was conducted for vegetation, caribou and wolf samples. Briefly, the ratio of  $^{13}\text{C}$  to  $^{12}\text{C}$  as compared with standards is measured in parts per thousand (‰) and denoted  $\delta^{13}\text{C}$ .  $\delta^{13}\text{C}$  varies among photosynthetic organisms,

and that in turn allows for tracing food sources [Ben David et al 2001]. The ratio of  $^{15}\text{N}$  and  $^{14}\text{N}$  as compared to standards is measured in ‰ and denoted  $\delta^{14}\text{N}$ .  $\delta^{14}\text{N}$  is correlated to the trophic level of the organism.

## Results

Highest PFC concentrations were found in the liver of wolves, perfluorononanoate (PFNA) had the highest concentration (mean 6.8 ng/g wet weight), followed by perfluorodecanoate (PFDA, mean 3.1 ng/g ww) and perfluoroundecanoate (PFUnA, mean 3.4 ng/g ww) (Table 2). PFOS concentrations were lower than these three compounds. Low amounts of perfluorododecanoate (PFDoA) and perfluorotridecanoate (PFTriA) were also observed in wolf liver with average concentrations in caribou tissues < 0.60 ng/g. The PFC concentrations in caribou liver showed a similar pattern with approximately 3 times lower PFNA and about 2 times lower PFDA and PFUnA values. The PFC contents in the caribou muscle were much lower, with average concentrations < 0.1 ng/g.

Low levels of PFCs were observed in vegetation and the pattern of PFCs differed widely between the plants and the lichen. Willow had the highest concentrations of PFOS and PFOA while lichen had highest concentrations of PFNA, PFDA and PFUnA. Only minor amounts of all PFCs were found in the other two plant species, sedge and cotton grass. Lichen showed a pattern of higher

**Table 2. Concentrations (Mean  $\pm$  SD) of perfluoroalkyl acids in vegetation, wolf and caribou samples<sup>1</sup>**

Vegetation	Herd/area	N	PFOS	PFOA	PFNA	PFDA	PFUnA	PFDoA
Cottongrass	Porcupine	10	<mdl	0.02 $\pm$ 0.04	<mdl	<mdl	<mdl	0.01 $\pm$ 0.02
Lichen	Porcupine	20	0.02 $\pm$ 0.03	0.03 $\pm$ 0.03	0.10 $\pm$ 0.09	0.05 $\pm$ 0.10	0.06 $\pm$ 0.05	0.02 $\pm$ 0.03
Sedge	Porcupine	3	<mdl	<mdl	<mdl	<mdl	<mdl	0.01 $\pm$ 0.00
Willow	Porcupine	7	0.11 $\pm$ 0.18	0.31 $\pm$ 0.38	0.09 $\pm$ 0.09	0.02 $\pm$ 0.04	0.00 $\pm$ 0.01	0.01 $\pm$ 0.01
Wolf liver	Bathurst	7	2.7 $\pm$ 0.76	0.09 $\pm$ 0.09	6.8 $\pm$ 3.1	3.7 $\pm$ 0.92	3.4 $\pm$ 1.0	0.60 $\pm$ 0.18
Caribou liver	Porcupine	10	0.67 $\pm$ 0.40	<mdl	2.2 $\pm$ 0.73	1.9 $\pm$ 0.34	1.7 $\pm$ 0.32	<mdl
Caribou muscle	Porcupine	7	0.03 $\pm$ 0.05	<mdl	0.04 $\pm$ 0.01	0.02 $\pm$ 0.01	0.03 $\pm$ 0.01	<mdl
Caribou muscle	Bathurst	7	0.08 $\pm$ 0.04	<mdl	0.07 $\pm$ 0.02	0.04 $\pm$ 0.02	0.08 $\pm$ 0.02	0.01 $\pm$ 0.00

<sup>1</sup> Arithmetic mean concentrations and standard deviations (SD). Dry weight basis for vegetation and fresh (wet) weight basis for muscle and caribou. MDL's ranged from 0.001 ng/g for PFNA to 0.04 ng/g for PFOA in plants and were generally 0.01 ng/g in caribou and wolf samples



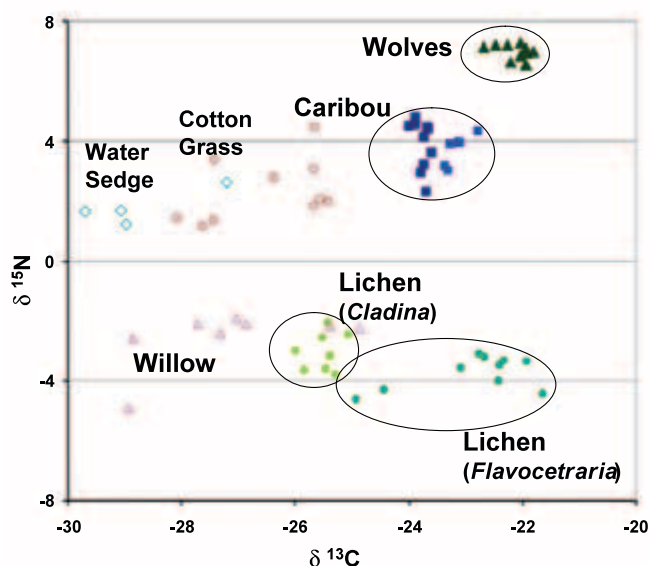


Figure 1. Plot of Nitrogen and carbon stable isotopes data in vegetation, caribou and wolf samples analysis

concentrations of odd chain length PFCAs (PFNA, PFUnA, and PFTrA) while willow did not show this pattern.

Results of carbon and nitrogen stable isotope analyses of vegetation and well as caribou and wolf muscle samples are shown in Figure 1. The lichen, caribou and wolf have similar  $\delta^{13}\text{C}$  implying that the caribou were feeding mainly on lichen before they were sampled and that wolves were feeding mainly on caribou. However, there is a large difference of  $\delta^{15}\text{N}$  between caribou and lichen (7-8 ‰) which implies that there may be other food sources since  $\delta^{15}\text{N}$  generally increases by a factor of 3.4-3.8 ‰ from prey to predator (Jardine et al. 2006).

## Discussion and Conclusions

Not having root systems, lichens absorb nutrients and water from precipitation the atmosphere. Therefore, PFC levels in lichen are representative of direct input from the atmosphere. In winter, about two-thirds of caribou diet is lichens, whereas in summer their diet is more diverse [Thompson and McCourt 1981, Boertje 1984].

It can be seen that PFCs mainly enrich in the liver of caribou whereas the accumulation in the muscular tissue is low. The analyte pattern found in caribou only shows PFCAs with chain length PFNA to PFUnA, whereas the plants show detectable

amounts of PFOA and PFDoA. The same was found for the wolf liver. The wolf diet in northern Canada consists of caribou and moose, and lesser quantities of smaller mammals. Considering concentrations in lichen, caribou (liver) and wolf liver it is clear that PFOS, PFNA, PFDA and PFUnA are biomagnifying in the food web. Further calculations to convert PFCs results in caribou and wolf to a whole body basis also confirm that these compounds biomagnify (results not shown).

The traditional knowledge (TK) interviews provide an excellent rationale for choosing the vegetation-caribou-wolf food chain as a model for northern terrestrial food-webs. The interviews tell us about the tight relationship between the wolves and the caribou: *"They don't hunt for them, they live with them"* [Percy Henry, Dawson]. Regularly, caribou constitutes the main food source for wolves in the research area, an estimated 80%. Some of the interviewees also indicated that the wolves eat preferential parts of the caribou, and leave the rest for scavengers. The preferred parts are tongue and liver; other parts will be eaten depending on the supply. The lead hunter starts with the tongue, then cuts the belly open and eats the liver (or part of) before the other wolves can join [Joel Peter, Freddy Frost, Old Crow]. The preference to liver would expose wolves to a high level of PFC contaminants from caribou. The TK also raises the question if the lead hunter will be exposed to higher levels of contaminants than other pack members, as it would seem that the lead hunter eats more liver than the other wolves. This might have implications if concentrations reached thresholds for possible effects. However, Environment Canada has estimated a No Effects concentration of 40 ng/g (wet wt) for liver of mammals (Environment Canada 2004) and therefore wolf livers are, on average, 15-times lower than this threshold. Also concentrations in wolf liver are 10 to 1000 times lower than found in livers of many arctic mammals including polar bears, mink, and arctic fox (Houde et al. 2006).

This study has shown that the atmospheric distribution of PFCs leads to measurable concentrations in vegetation in a remote region such as northern Canada. As a result, herbivorous mammals such as caribou and subsequently their predators such as the wolf accumulate PFCAs and PFOS. The generally low concentrations in



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comparison to marine mammals can be explained by the lack of the marine input and the shorter terrestrial food chain. While concentrations are low the fact that these chemicals increase in concentration from vegetation to caribou is a concern. This may also be the case for many other chemicals which have a combination of properties enabling widespread distribution and bioaccumulation (Kelly et al. 2008). This pathway, which is ultimately important for human exposure in the Arctic (Ostertag et al. 2009) needs further investigation.

## Expected Project Completion Date

July 31<sup>st</sup> 2009

## Acknowledgments

The researchers acknowledge with gratitude the contribution of the elders who contributed traditional knowledge: Robert Alexie Sr. and Woody Elias from Fort McPherson, NT, Percy Henry and Julia Morberg from Dawson, NT, Freddy Frost, Stan Njootli and Joel Peter from Old Crow, YT. Megan Williams from the Vuntut Gwitchin First Nations in Old Crow, YT, Jody Beaumont from Tr'ondek Hwech'in First nations in Dawson, YT facilitated the traditional knowledge interviews in their communities. Madaline deRepentigny was a co-interviewer with Alestine Andre in Dawson, YT. Mary Jane Moses interviewed three elders in Old Crow, YT.

Tamara Hansen from Inuvik, NT and Myranda Charlie from Old Crow, YT, participated in the sample collections for this project. Caribou serum was donated by Dorothy Cooley from Environment Yukon. This work would not be possible without the kind support of INAC-NCP.

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# Temporal trend studies of the atmospheric Hg deposition with ice-core/snow in Canadian High Arctic

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David Bass, student from University of Ottawa, field help and lab assistant.

## Abstract

A 10.34-meter pit was dug and continuous sampling of the pit was performed on Mt. Oxford, Nunavut, Canada in April-May, 2008. From bottom of the pit, a 4.2-meter core was drilled to extend the pit record. First age estimation of the pit samples is ~30 years and the ice core extension (with an overlay of 62 cm with the pit), 10-12 years, which makes the record of ~42 years. Resolution for the full record is ranging from 3 to 6 samples per year. All samples were taken, protected, preserved and decontaminated following strict procedures and technique developed at Geological Survey of Canada (GSC). Analyses of samples were completed and partial results have been presented in the 2008 Annual workshop held in Yellowknife.

## Résumé

Un puits de 10,34 mètres a été creusé et des échantillons ont été prélevés continuellement au mont Oxford, au Nunavut, Canada, en avril-mai 2008. Une carotte de 4,2 mètres a été forée à partir du puits de la fosse afin de prolonger les enregistrements à partir du puits. La première estimation de l'âge des échantillons prélevés dans le puits est d'environ 30 ans et de 10 à 12 ans pour le prolongement de la carotte de glace (avec un recouvrement de 62 cm avec le puits), soit un enregistrement d'environ 42 ans. La résolution pour l'enregistrement complet varie de 3 à 6 échantillons par année. Tous les échantillons ont été prélevés, protégés, préservés et décontaminés suivant des procédures et des techniques rigoureuses mises au point par la Commission géologique du

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Canada (CGC). Les analyses des échantillons sont terminées et les résultats partiels ont été présentés lors de l'atelier annuel de 2008 tenu à Yellowknife.

## Key Messages

- Temporal variation of total Hg in ice/firn profile is comparable to records/results from direct atmospheric measurements and caribou samples.
- This study further confirms that Ice cores and snow/firn profiles are one of the most prominent natural media and therefore, it is possible to reconstruct pre-recorded atmospheric Hg information from ice and firn.

## Messages clés

- Les variations temporelles de mercure total dans les profils de glace et de névé sont comparables aux enregistrements ou résultats provenant des mesures atmosphériques directes et des échantillons prélevés sur les caribous.
- Cette étude confirme une fois de plus que les échantillons de glace et les profils de neige et de névé constituent l'un des meilleurs médias naturels et permettent par conséquent de reconstituer l'information atmosphérique préenregistrée sur le mercure à partir de la glace et du névé.

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## Objectives

To reconstruct a temporal variation of total Hg from ice and firn/snow in Canadian High Arctic and to evaluate if the reconstructed archive can be comparable to other records such as direct atmospheric measurements (in order to reconstruct atmospheric information before direct atmospheric monitoring). Also, to evaluate if present sample preservation and storage are proper for Hg quantification in snow/firn and ice samples.

## Introduction

Ice caps preserve atmospheric depositions, including both natural and anthropogenic contributions. Because ice is virtually frozen water, chemical, physical, or biological change/processing (diagenesis) is limited. Therefore, reconstruction of archives from ice reflects concurrent atmospheric depositions. Due to mercury depletion events (MEDs) (Schroeder, W.H. et al., 2003, Steffen et al., 2007) and its easy conversion between its different forms, Hg in ice/snow samples is harder, compared to other trace metals, to be precisely quantified. Many studies have been carried out in Arctic environment. However, there are so far no high resolution records that extend back to 40 years.

In order to confirm that samples for total Hg study can be properly preserved and stored in a low temperature walk-in freezer before analyses,

comparison studies are also needed. To do so, two methods should be used, one in the north and the other in base labs. This is important because contaminated samples provide false information and mislead conclusions. This study aims to compare results quantified by a portable Zeeman RA-915+ Hg analyzer (LOD=0.5 pg/g) in Resolute Bay laboratory immediately after returning from the ice cap and Tekran 2600 in laboratories at GSC in Ottawa (LOD=<0.05 pg/g).

## Activities in 2008/2009

- Field trip: Field sampling was carried out in April and May, 2008. Two person team for this work (also team up with another colleague for pollen studies in ice/snow). A total of 10.34 meter pit was dug and sampled.
- Sample analyses were completed before this fiscal year (before end of March of 2009);
- Near on site measurements (in the labs at Resolute PCSP base) were completed (May, 2008) as planned.
- Air Hg measurements in Resolute area were carried out in May, 2008 and presented at the 2008 NCP annual workshop.
- Total Hg distribution in current snow at the Churchill Northern Study Center was carried out for comparison purpose (March 2008).

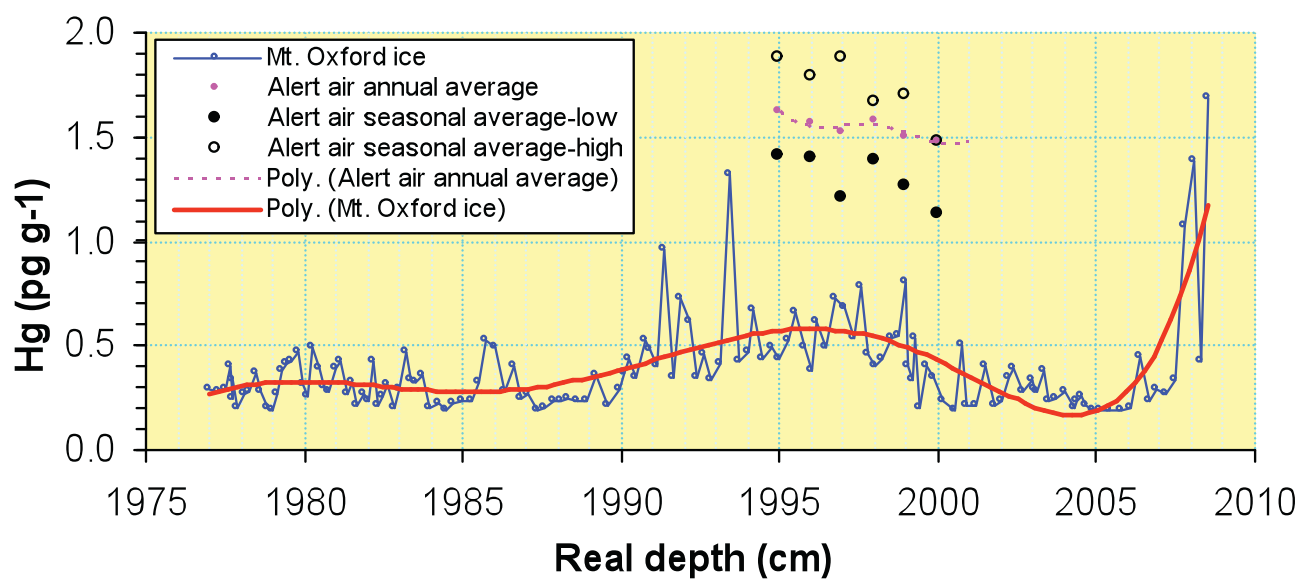


Figure 1. Reconstruction of total Hg with snow/firn retrieved from Mt. Oxford, Nunavut, Canada.

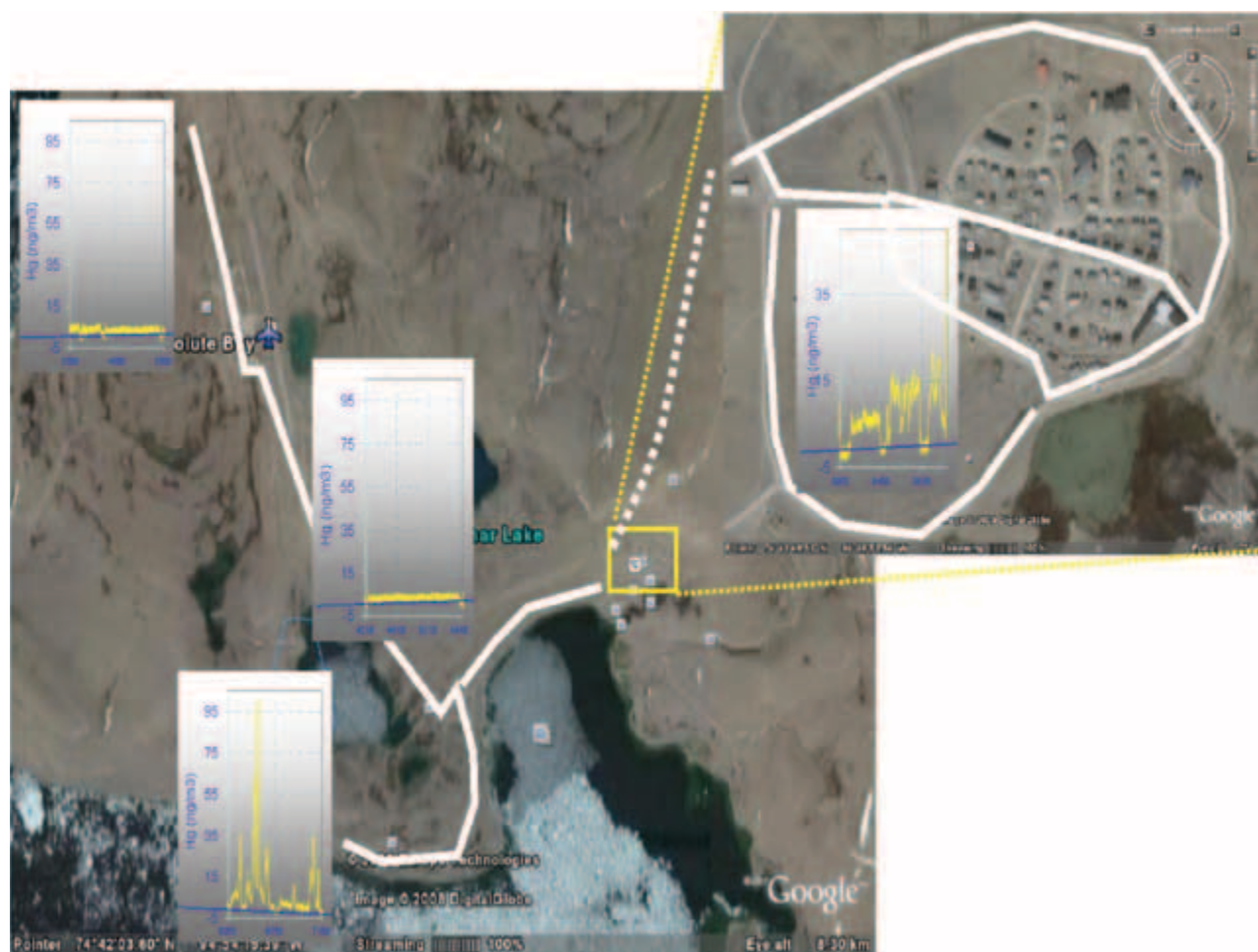


Figure 2. Gaseous Hg measurement in Resolute area in May 2008.



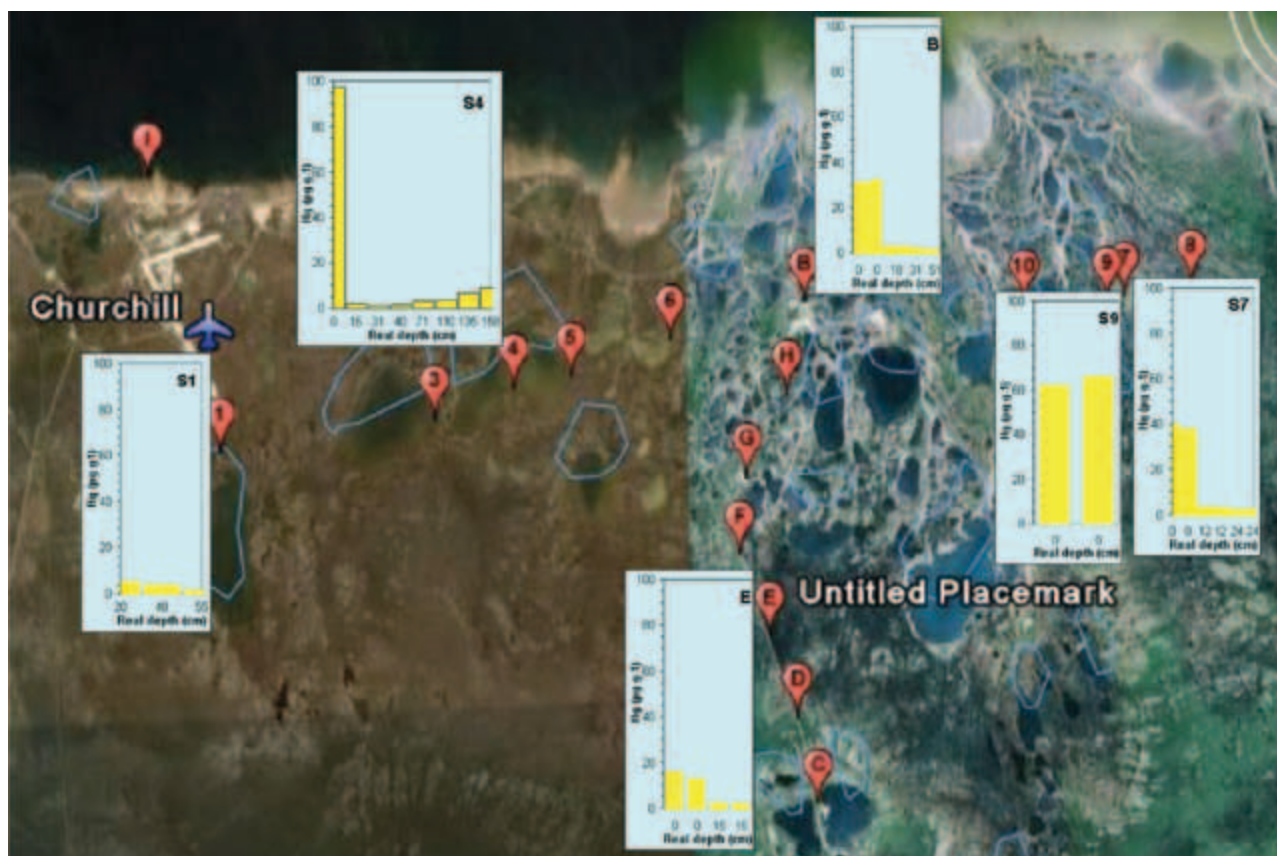


Figure 3. Total Hg in current year snow layers in Churchill area in March 2008.

## Results

Primary results from the pit samples (from 1973 to 2008, Figure 1) show that a higher deposition of total Hg was found between 1991 and 1999 (Average of 0.56 pg/g, N=32), and the lowest were found between 2004-2005 (Average of 0.22 pg/g, N=9), 1987-1989 (Average of 0.25 pg/g, N=11) and 1984 (Average of 0.21 pg/g, N=5). The highest (Average of 1.69 pg/g, N=4), however, was found in the top 35 cm snow/firn layers (2008 May to 2007 late summer). This is most likely because of the higher porous structure of the top now/firn layers, which can more easily communicate with outside atmosphere. Further detailed interpretation of the data set will be pursued when sample analyses are completed.

Atmospheric measurements of Hg in Resolute area showed higher in village and dumping site area but at background level in other areas (Figure 2).

Total Hg in current snow at Churchill area showed 2-4 times higher than those in glaciers (Figure 3).

Comparison of two methods by two different instruments shows they are comparable and preservation/shipping procedures are reliable. However, sample container types can be critical for storage of snow/ice samples for Hg quantification based on the preliminary results from this study (results are being processed now).

## Discussion and Conclusions

Results from this study show correlation with those from Caribou completed by Dr. Mary Gamberg (personal communication), Yukon. This could be due to both samples (snow/firn and caribou) receive Hg mainly from atmospheric sources (for glaciers, almost 100% of deposition of Hg is atmospheric).

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Results achieved in this study are going to compare with records of direct atmospheric monitoring at Alert, Nunavut, Canada, carried out by Environment Canada (Steffen A. 2008). When they are comparable fully, it is justified to extend atmospheric mercury deposition record back to hundreds, even thousands of years with ice media. Currently, longer ice core drilling (up to 1000 years) is aimed at Agassiz Ice Cap in the coming field seasons.

## Expected Project Completion Date

Study of total Hg in snow/ice samples taken from Mt. Oxford has been completed. Results will be published whenever suitable and possible. However, continuation of longer total Hg archive reconstruction and comparison of ice/snow records with direct atmospheric measurements will go 3 to 5 years more, including a possible new deeper drilling (down to bed rock) at Agassiz Ice Cap.

## Acknowledgments

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# Mercury in Caribou Forage

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## **Abstract**

Results from the Arctic Caribou and Moose Contaminant Monitoring Program indicate an increasing temporal trend in renal mercury in Porcupine caribou, particularly in females. In order to better understand the dynamics of mercury within these caribou populations and be better able to predict if mercury may become a toxicological issue, we have investigated the question of why mercury levels are higher in female caribou than in males. Samples of caribou forage were collected from the winter and summer range of the Porcupine caribou and analyzed for total and methyl mercury. The results were used with an energetics model developed for the Porcupine caribou herd to estimate mercury intake by cows, bulls and non-reproducing cows. Results suggest that although cows may ingest slightly more MeHg than bulls on the North Slope (summer range), the more likely reason for the higher concentrations of Hg seen in cows is due to their smaller body size and their proportionally higher intake of forage. One remaining question is how much Hg is ingested from mushrooms in the fall, and whether both cows and bulls ingest mushrooms at the same rate.

## **Résumé**

Les résultats du Programme de surveillance des contaminants chez les caribous et les orignaux arctiques indiquent une augmentation de la tendance temporelle du taux de mercure dans les reins chez les caribous de la Porcupine, en particulier chez les femelles. Afin de mieux comprendre la dynamique du mercure au sein de ces populations de caribous et d'être mieux en mesure de prédire si le mercure risque de devenir un problème toxicologique, nous avons tenté de déterminer pourquoi les niveaux de mercure sont plus élevés chez les femelles que chez les mâles. Des échantillons de fourrage du caribou ont été recueillis dans les aires d'hivernage et d'été des caribous de la Porcupine et analysés afin de mesurer la quantité totale de mercure et de méthylmercure. Les résultats ont été utilisés avec un modèle énergétique élaboré pour le troupeau de caribous de Porcupine afin d'estimer l'ingestion de mercure par les femelles, les mâles et les femelles non reproductrices. Les résultats laissent penser que même si les femelles ingèrent peut-être légèrement plus de méthylmercure (MeHg) que les mâles sur le versant nord (aire d'été), les concentrations supérieures de mercure (Hg) constatées chez les femelles s'expliquent probablement par le fait que leur corps est de plus petite taille et qu'elles ingèrent proportionnellement des quantités de fourrage supérieures. Une question qui subsiste consiste à savoir combien de

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mercure est ingéré par la consommation de champignons l'automne et si les mâles et les femelles ingèrent les mêmes taux de champignons.

## Key Message

- Female caribou have higher concentrations of renal mercury than males, most likely because they are smaller and eat proportionally more food (and therefore more mercury).

## Message clé

- Les femelles caribous présentent des concentrations supérieures de mercure dans les reins que les mâles, probablement en raison de leur plus petite taille et du fait qu'elles consomment proportionnellement davantage de nourriture (et par conséquent davantage de mercure).

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## Objectives

- To determine why female Porcupine caribou have higher renal mercury concentrations than males. This will allow a better understanding of mercury dynamics within the caribou population, enabling more accurate predictions of whether mercury is likely to become a health issue for the caribou or consumers of the caribou.
- To further understand the fate and effects of mercury deposition in the Canadian Arctic.

## Introduction

Caribou provide an important food resource for Northerners across the Arctic. Many Arctic caribou herds, including the Porcupine, are currently in decline and this is causing considerable concern among wildlife managers as well as those people of the north using these herds as a food source (Russell et al. 2002; Gunn et al. 2005). Results from the Arctic Caribou and Moose Contaminant Monitoring Program indicate an increasing temporal trend in renal mercury in Porcupine caribou, particularly in females. Although mercury levels are not yet at the point where toxicological effects are thought to occur, if concentrations continue to increase, there may come a point where they will (if they have not already) begin to negatively impact the health of the caribou population, or those consuming the caribou.

In order to better understand the dynamics of mercury within these caribou populations and be better able to predict if mercury may become a toxicological issue, we have investigated the

question of why mercury levels are higher in female caribou than in males.

There is potential for the mercury content of the forage consumed during the calving season to be different between the genders. In early April, female Porcupine caribou travel north to the Arctic Coast while the males remain in the more southern wintering grounds. By early June, the females reach the coast and spend about six weeks there before the males join them and they begin their journey back south (Russell et al. 1992). During the calving period, therefore, although males and females are consuming relatively similar species of forage, they are doing so in very different geographical areas. The Arctic Coast may be subject to an increased mercury load from mercury depletion events in the spring, or simply the proximity of the ocean, which acts as a sink and a transport pathway for mercury (Steffen et al. 2007). If this is the case, caribou forage in this area should have higher levels of mercury than similar species in the more southern wintering areas where the males are foraging at that time, which would explain the higher levels of mercury seen in female caribou.

In order to further explore these hypotheses, a mercury intake model was developed for the Porcupine caribou utilizing an existing forage intake model used by Environment Canada to estimate nutrient intake by the Porcupine caribou.

## Activities in 2008/2009

Vegetation was collected in late June, 2008 from Chapman Lake (southern Dempster Highway) by the PI, two employees of the Aurora Research

Station and five volunteers from the Tr'ondek Hwech'in First Nation in Dawson. Vegetation was collected at the same time from the North Slope by Don Reid, Wildlife Conservation Society.

Vegetation samples were kept frozen until sorted (by species) and analyzed. Total and methyl mercury were analyzed by Quicksilver Labs, Colorado, using a High Pressure Liquid Chromatography Speciation System. Quality assurance from this laboratory was within acceptable parameters.

Total mercury (THg) and methyl mercury (MeHg) intakes were modeled for the Porcupine caribou herd using an energetics model (Russell et al 2004). The model tracks the intake, digestion and allocation of fat and protein for an individual caribou throughout the year. Because not all plant groups considered in the model were analyzed in this study, the following assumptions were made:

- Evergreen shrubs, forbs, *Equisetum* had THg and MeHg concentrations equivalent to willows.
- *Eriophorum* heads and standing dead had THg and MeHg concentrations equivalent to entire *Eriophorum* plants.
- Mosses and mushrooms had THg and MeHg concentrations equivalent to lichens.

In the model, the caribou are 'presented' with a changing complex of plant communities throughout their annual cycle. In late winter, cows begin to move north from the taiga to their tundra calving

grounds. During this period the bulls and non-pregnant cows stay back on the winter range and follow up to 3 weeks later. During spring green-up, coincident with the calving periods cows remain primarily on the coastal plain while bulls are much more mobile and are able to better track newly emerging plant communities. We modeled the intake for bulls, cows and the hypothetical situation where cows remained with the bulls using mercury measured in vegetation from the winter range (Chapman Lake) and the summer range (North Slope). Note that a different activity budget was not used for bulls during the rut. We know that bulls drastically reduce feeding for a 3 week period in October during the rut although separate activity budgets have not been collected. We can assume that this period, coincident with some of the highest intake of Hg for cows, would be a period of low intake of Hg in bulls.

## Results and Discussion

Statistical comparisons of vegetation mercury concentrations between locations on a species basis were difficult because only two species were able to be collected from both locations (Table 1). As a result, the species were grouped according to plant type (Figure 1). Although the average concentration of MeHg is higher in all plant groups from the North Slope than from Chapman Lake, the only statistical difference is for willows ( $p = 0.027$  using a Kruskal-Wallis one-way ANOVA). THg

**Table 1. Total (THg) and Methyl (MeHg) Mercury in vegetation collected from Chapman Lake and the North Slope of the Yukon in June, 2008.**

Species	Chapman Lake			North Slope		
	N	THg (ng/g dry wt)	MeHg (ng/g dry wt)	N	THg (ng/g dry wt)	MeHg (ng/g dry wt)
<i>Carex aquatilis</i>	5	5.62 ± 2.33	0.25 ± 0.10	3	5.36 ± 1.33	0.60 ± 0.51
<i>Cladina mitis</i>	5	107.33 ± 123.41	2.07 ± 2.63			
<i>Eriophorum angustifolium</i>				4	5.27 ± 1.34	0.30 ± 0.14
<i>Eriophorum vaginatum</i>	5	11.15 ± 10.87	0.20 ± 0.21	4	10.97 ± 3.81	0.31 ± 0.17
<i>Flavocetraria cucullata</i>	5	71.77 ± 86.29	1.79 ± 1.96			
<i>Flavocetraria nivalis</i>				1	48.86 ± 4.65	2.53 ± 0.02
<i>Salix glauca</i>				4	5.24 ± 1.57	1.10 ± 0.54
<i>Salix pulchra</i>	5	5.55 ± 1.40	0.31 ± 0.25			

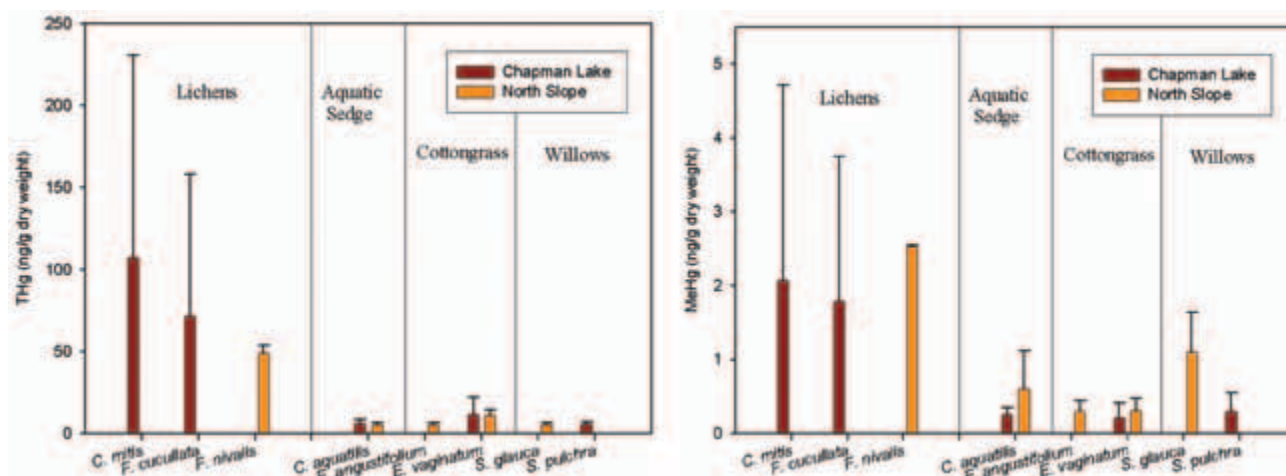


Figure 1. Total and Methyl mercury in vegetation collected from Chapman Lake and the North Slope of the Yukon in June, 2008.

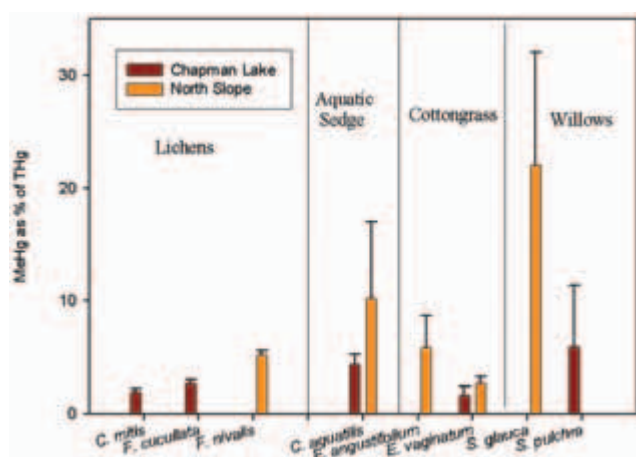


Figure 2. The proportion of THg found as MeHg in vegetation collected from Chapman Lake and the North Slope of the Yukon in June, 2008.

was not significantly different between locations in any of the plant groups (Figure 1), but the proportion of THg found as MeHg was significantly higher in vegetation from the North Slope for all plant groups except sedges where it approached significance ( $p=0.053$ ) (Figure 2).

Average THg and MeHg intakes by Porcupine caribou over the course of a calendar year are shown in Figure 3. In the winter both genders are eating mainly lichen which have relatively high concentrations of Hg. In March there is an increase in Hg intake as the snow melts and the quantity of lichens consumed increases (as the caribou have

to spend less time cratering through snow for the lichens and can spend more time eating). In April/May the caribou start eating newly emerging vegetation (deciduous shrubs and forbs) which are lower in mercury than lichens, so mercury intake declines rapidly. During June and July THg remains low while MeHg fluctuates, likely due to consumption of willows which, on the North Slope, contain a higher proportion of MeHg. As the summer wanes the diet shifts back to lichens and the mercury intake increases accordingly. The peak in mercury intake seen in mid-September is due to a brief period of mushroom intake. In this model we have assumed that mushrooms are equivalent to lichens in terms of mercury content, but in fact they may actually be higher. Kalač et al (2004) report THg in mushrooms from unpolluted areas ranging from 1-20 mg/kg (or 20,000 ng/g) which is considerably higher than lichens measured in this study.

The model used to calculate mercury intake used vegetation data from the appropriate location (ie. North Slope or Chapman Lake), depending where the caribou were on that Julian day (on average). Cows with bulls were assumed to be in the same locations and eating the same diet as bulls. Therefore, any difference in Hg intake between cows with bulls and cows would be due to location. Figures 3 and 4 shows very little difference among the three groups for total intake of Hg over the year, although there is a slight increase in MeHg in cows in mid-June, likely due to consumption of

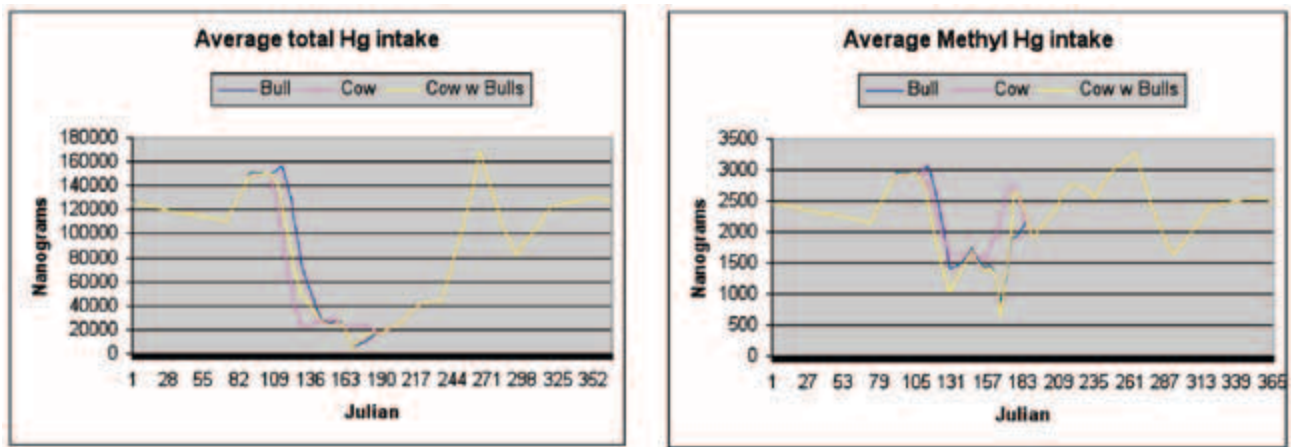


Figure 3. Average THg and MeHg intake by Porcupine caribou over one calendar year.

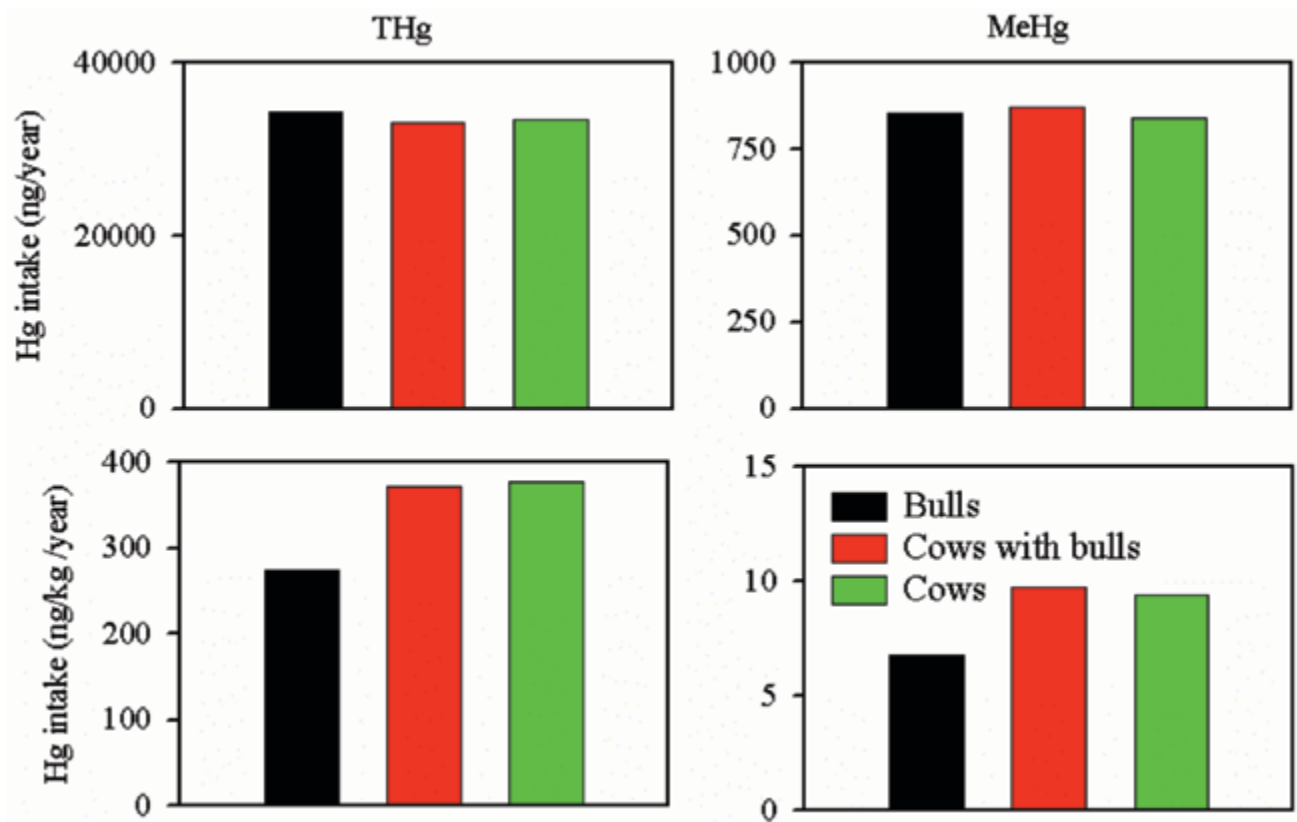


Figure 4. Hg intake by Porcupine caribou over a calendar year, and Hg intake over a calendar year as a proportion of body weight.

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willows on the North Slope. However, if the Hg intake is expressed as a proportion of body weight, bulls clearly ingest less Hg (proportionally).

Using the vegetation data from this study in the energetics model for the Porcupine caribou suggests that although cows may ingest slightly more MeHg than bulls on the North Slope, the more likely reason for the higher concentrations of Hg seen in cows is due to their smaller body size and their proportionally higher intake of forage. One remaining question is how much Hg is ingested from mushrooms in the fall, and whether both cows and bulls ingest mushrooms at the same rate.

## Project Completion Date

This project has been completed.

## Acknowledgements

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# PCBs in Arctic Ocean Seawater

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## Abstract

The objective of this project was to provide accurate measurements of PCBs at the extremely low levels present in Arctic Ocean seawater using a large-volume pumping approach. Previous samples frequently suffer from contamination and the uncertainties inherent in such data prevent a proper perspective being developed in biogeochemical cycling of PCBs in the Arctic Ocean. To avoid potential contamination associated with ship-based sampling, seawater samples were collected using the landfast sea ice as a platform during a unique sampling opportunity as the Canadian Coast Guard Ship *Amundsen* completed its over-wintering period in the Amundsen Gulf region (IPY CFL study area). Samples were obtained by deploying *in situ* pump systems (Infiltrax and Seastar) equipped with stainless-steel columns containing XAD-2 resin, which extract dissolved PCBs beneath the landfast ice. The columns were preceded by glass fiber filters to intercept suspended particulates (and associated PCBs). Five seawater samples (181-530 L) were collected over the period 29 April – 16 June, 2008, together with four 'field blanks' (quality control samples). PCB congeners were analyzed at an ultra-clean facility (AXYS Analytical Services Ltd.),

## Résumé

L'objectif de ce projet consistait à fournir des mesures précises des BPC présents à des taux extrêmement faibles dans l'eau de mer de l'océan Arctique à l'aide d'une méthode de pompage à haut débit. Les échantillons recueillis auparavant étaient souvent contaminés et les incertitudes inhérentes à ce type de données empêchent d'obtenir une bonne perspective en matière d'établissement du cycle biogéochimique des BPC dans l'océan Arctique. Afin d'éviter une contamination potentielle lorsque les échantillons sont recueillis par bateau, les échantillons d'eau de mer ont été recueillis en utilisant la glace de rive comme plateforme lors d'une occasion unique de prélever des échantillons alors que le navire de la Garde côtière canadienne *Amundsen* terminait son séjour hivernal dans la région du golfe Amundsen (région de l'étude API-CSC). Les échantillons ont été obtenus en déployant des systèmes de pompes *in situ* (Infiltrax et Seastar) équipées de colonnes en acier inoxydable contenant de la résine XAD-2 permettant d'extraire les BPC dissous sous la glace de rive. Les colonnes étaient munies de filtres en fibre de verre afin d'intercepter les particules en suspension (et les BPC connexes). Cinq échantillons d'eau de mer (181-530 l) ont été recueillis au cours de la période comprise entre le

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which guarantees ultra-low detection limits. Low blank levels ( $<15$  pg absolute) were generally achieved (three out of four field blanks), implying that the clean techniques used for the sampling were largely adequate. After correcting for the blank levels, we estimate concentrations of PCBs in Canadian Arctic Ocean seawater of generally less than 4 pg/L. These values are lower than most previously reported values for Canadian Arctic seawater but similar to recently reported values for the European Arctic. These levels imply a standing stock of PCB in the upper ocean of the Canadian Basin of about 3000 kg.

29 avril et le 16 juin 2008, ainsi que quatre « blancs de terrain » (échantillons de contrôle de la qualité). Les congénères de BPC ont été analysés dans une installation ultra-propre (AXYS Analytical Services Ltd.) qui garantit la détection de concentrations ultra-faibles. Des niveaux de blanc très faibles ( $<15$  pg absolus) ont généralement été obtenues (trois sur quatre des blancs de terrain), ce qui signifie que les techniques propres utilisées sont largement adéquates. Après correction en fonction des niveaux de blanc, nous estimons que les concentrations de BPC dans l'eau de mer de l'Arctique canadien sont généralement inférieures à 4 pg/l. Ces valeurs sont inférieures aux valeurs déclarées auparavant pour l'eau de mer de l'Arctique canadien mais similaires aux valeurs déclarées récemment dans l'Arctique européen. Ces niveaux supposent un stock actuel d'environ 3 000 kg de BPC dans les couches supérieures de l'océan du bassin canadien.

## Key Messages

- A set of five samples of Arctic Ocean seawater, together with four blanks (QA/QC samples), were collected using clean techniques from the sea ice platform in Amundsen Gulf (IPY CFL study area).
- After correcting for low levels of contamination associated with handling, sampling and lab analysis, PCB concentrations in the seawater samples (dissolved fraction) are estimated at generally less than 4 pg/L.

## Messages clés

- Un ensemble de cinq échantillons d'eau de mer de l'océan Arctique, ainsi que de quatre blancs de terrain (échantillons d'assurance et de contrôle de la qualité), ont été prélevés à l'aide de techniques propres à partir de la plateforme de glace d'eau de mer dans le golfe Amundsen (région de l'étude API-CSC).
- Après correction en fonction des faibles niveaux de contaminants liés à la manipulation, à l'échantillonnage et aux analyses en laboratoire, les concentrations de BPC dans les échantillons d'eau de mer (fraction dissoute) sont généralement estimées à moins de 4 pg/l.

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## Objectives

1. Sample Canadian Arctic Ocean seawater beneath the sea ice cleanly and in replicate.
2. Quantify PCBs in the seawater samples at ultra-low detection limits.
3. Determine PCB congener patterns in Arctic surface water and link these to sources or processes using ancillary data sets.
4. Communicate these new data, and what we can infer about previous data and model assumptions, in a timely manner.

## Introduction

Although much has been learned over the past 30 years about traditional contaminants like PCBs and their pathways, processes and effects in the Canadian Arctic, the cycling of PCB within the Arctic Ocean (seawater) remains a major knowledge gap. PCB concentration data for Arctic seawaters are rare, partly because opportunities to collect the samples are rare, and partly because it is challenging to sample and analyze PCBs at the extremely low concentrations that occur in Arctic seawater, without contamination or other biases.

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Ships, for instance, represent significant potential sources of contamination. Data collected during this study and by other studies using clean techniques increasingly imply that some of the data collected in the past – including some in the Canadian Arctic – are wrong.

The goal of this project is to obtain new, trustworthy measurements of dissolved PCB concentrations in Canadian Arctic Ocean seawater, guided by recent developments in our understanding of the challenges associated with this work. Here, we report on a first attempt at this sampling during spring 2008 in the Amundsen Gulf region of the Beaufort Sea (IPY CFL study area). Arctic Ocean seawater samples were collected using clean techniques during the over-wintering of the Canadian Coast Guard Ship *Amundsen* in Amundsen Gulf. In situ pump systems equipped with glass fibre filters to collect suspended particulates (and associated PCBs) and columns containing XAD-2 resin, which extract dissolved PCBs, were deployed beneath the sea ice, thus sampling Arctic Ocean seawater remote from the influence of possible sources of contamination (e.g., the ship). The samples were analyzed at an ultra-clean facility, which guarantees the necessary ultra-low detection limits. The new data obtained provide a first step towards ultimately addressing what remains a major gap in our understanding of the sources, sinks and reservoirs for PCBs in the Arctic Ocean.

## Activities in 2008/2009

### *Pre-sampling preparation and validation of equipment*

Prior to the field work, the Teflon columns intended for use in the sampling were forwarded to the ultra-clean laboratory facility at AXYS Analytical Services Ltd. (AXYS) in Sydney, BC, and subjected to rigorous, sequential solvent cleaning, followed by confirmatory testing to ensure that they met the necessary ultra-clean standards for this project. Unfortunately, despite several attempts to clean the Teflon columns, these columns did not reliably meet the necessary ultra-clean requirements. New stainless-steel columns (ca. 500 mL) were purchased and tested (all passing), then prepared for the field work. Each column was packed with ca. 250 g of XAD resin, which had been independently cleaned and tested, then spiked with 40 µL of field standard

solution containing labelled standards PCB 31, 95, and 153. The prepared columns were packaged individually in vacuum-sealed bags and transported to the CCGS *Amundsen*, where they were stored aboard until use (days to weeks). At FWI, glass fibre filters (GF/F, 142 mm, 0.7 µm nominal pore size) were combusted (460°, 4 hrs), individually packaged in aluminium envelopes and vacuum-sealed in bags and deployed to the field.

### *Sampling*

Unusual sea ice conditions, which allowed a more or less continual westward motion of the ice, precluded the establishment of the landfast ice camp planned for May-June 2008. Consequently, sampling was conducted whenever opportunities arose to temporarily access the sea ice for a sufficient length of time – six occasions during the period of 29 April–16 June 2009 (Table 1). Using clean techniques, in situ pump systems (Infiltrax and Seastar) were deployed beneath the sea ice alone or in pairs (depending on conditions). Columns and filters were installed on the pumps away from the ship (with the exception of Field blanks 1 and 2 and Sample 1). After sampling, columns and filters were placed in ziplocks and tinfoil, respectively, for transport back to the ship, and then returned to vacuum-sealed containers immediately after being taken aboard. Five samples consisting of 181–530 L of pumped seawater were collected, together with four field blanks, which consisted of 1- to 2-L of seawater pumped through the system. Blanks and samples were collected back-to-back when conditions allowed (April 29, June 8, and June 12; Table 1). For deployments lasting longer than about 30 hours, the pumps were temporarily removed from the water to exchange the batteries and then redeployed. In two cases (Samples 4 and 5), the filters were also exchanged at that time because they were visibly coated with material.

### *Lab Analysis*

XAD resin and filters were analyzed in separate batches (two for XAD), with each batch including a laboratory blank and spiked matrix (QA/QC) sample. The blank and spike for the XAD batches were prepared using 30 g of clean XAD resin, while the blank and spike for the filter batches were prepared using a clean wound-glass filter. Analysis was conducted by high-resolution gas

**Table 1. List of seawater samples and field blanks (QA/QC samples) collected in spring 2008.**

Description	Date	Duration (hours)	Sample depth (m)	Volume (L)	Pump type	Lat. (N)	Long. (W)	Bottom depth (m)
Field blank 1	29-Apr-08	0.17	15	1	Infiltrex	70° 44.65	123° 33.10	545
Sample 1	29-Apr-08	42.21	15	392	Infiltrex	70° 44.65	123° 33.10	545
Field blank 2	30-Apr-08	0.17	15	1.1	SeaStar	70° 46.47	123° 54.13	462
Sample 2	09-May-08	41.75	15	249	SeaStar	70° 04.45	125° 09.05	31.5
Field blank 3	08-Jun-08	0.00	0.5	2.4	Infiltrex	69° 49.64	123° 37.86	78.9
Sample 3	08-Jun-08	29.83	15	181	SeaStar	69° 49.64	123° 37.86	78.9
Sample 4	12-Jun-08	59.13	15	494	SeaStar	69° 49.46	123° 37.97	78.9
Field blank 4	12-Jun-08	0.12	15	1.5	SeaStar	69° 49.46	123° 37.97	78.9
Sample 5	16-Jun-08	59.17	15	530	SeaStar	69° 57.39	125° 52.01	107

chromatography/high-resolution mass spectrometry in accordance with AXYS method MLA-010, modified from US EPA method 1668A. Briefly, each sample was spiked with labelled quantification standards and then extracted by Soxhlet using dichloromethane. The extract was split into two equal portions (one for analysis, the other for back-up), and the analysis portion spiked with labelled cleanup standards and then cleaned up using standard chromatography columns. Final extracts were reduced in volume and spiked with labelled recovery (internal) standards prior to analysis on a HRMS coupled to a HRGC, equipped with a SPB-Octyl chromatography column (30 m, 0.25 mm i.d., 0.25 µm film thickness). The spiked samples show generally good recoveries for the PCB congeners (94% ± 7.3%). The laboratory blanks generally contained very low or non-quantifiable amounts of PCBs (20-43 of 159 reported congeners). The majority of the quantified amounts (74%) were less than 5 pg absolute (5 pg per blank), with only a few congeners (generally penta- and hexachlorobiphenyls) at 10-20 pg or greater. PCB7, 11 and 122 were present at relatively high levels in both XAD blanks and consequently excluded from further consideration.

## Results and Discussion

### *Field blanks*

Because concentrations of PCBs in Arctic seawater are extremely low, there is significant risk of contamination during storage, handling and sampling.

Determining 'blank levels', i.e., levels of PCB in field blanks, which have been treated exactly like samples, is therefore crucial for assessing the reliability and resolution of the data (cf. Sobek et al., 2003). The blank levels of individual PCB congeners in XAD Field blanks 1, 2 and 3 (Table 1) were generally (60% of detected values) less than 10 pg absolute and the majority of values (80%) less than 20 pg. Most of the higher values were highly-chlorinated congeners (PCB 138, 153, 180, 187, 206), especially in XAD Field blank 2. In general, the field blank levels are within a factor of two to four of the levels in the lab blanks (Figure 2), and this small difference may be accounted for simply by the roughly eight-fold greater quantity of XAD resin in the field blanks (ca. 250 g), compared to the lab blanks (30 g), and trace amounts associated with the stainless steel columns (generally 1-2 pg; see Figure 3). The low levels in these three field blanks are also consistent with field blanks from previous studies employing rigorous QA/QC and ultra-clean techniques, e.g., ca. 3-8 pg in a study in the Barents Sea and North Pole area (Gustafsson et al., 2005), 8-15 pg in PUF absorbent and GFF filter blanks in a study in the Baltic Sea (Sobek et al., 2003). We therefore conclude that, in general, the clean handling and sampling procedures employed in the study successfully minimized contamination and achieved conditions appropriate for accurately quantifying PCBs in Arctic seawater. However, XAD Field blank 4 is an exception, with clearly

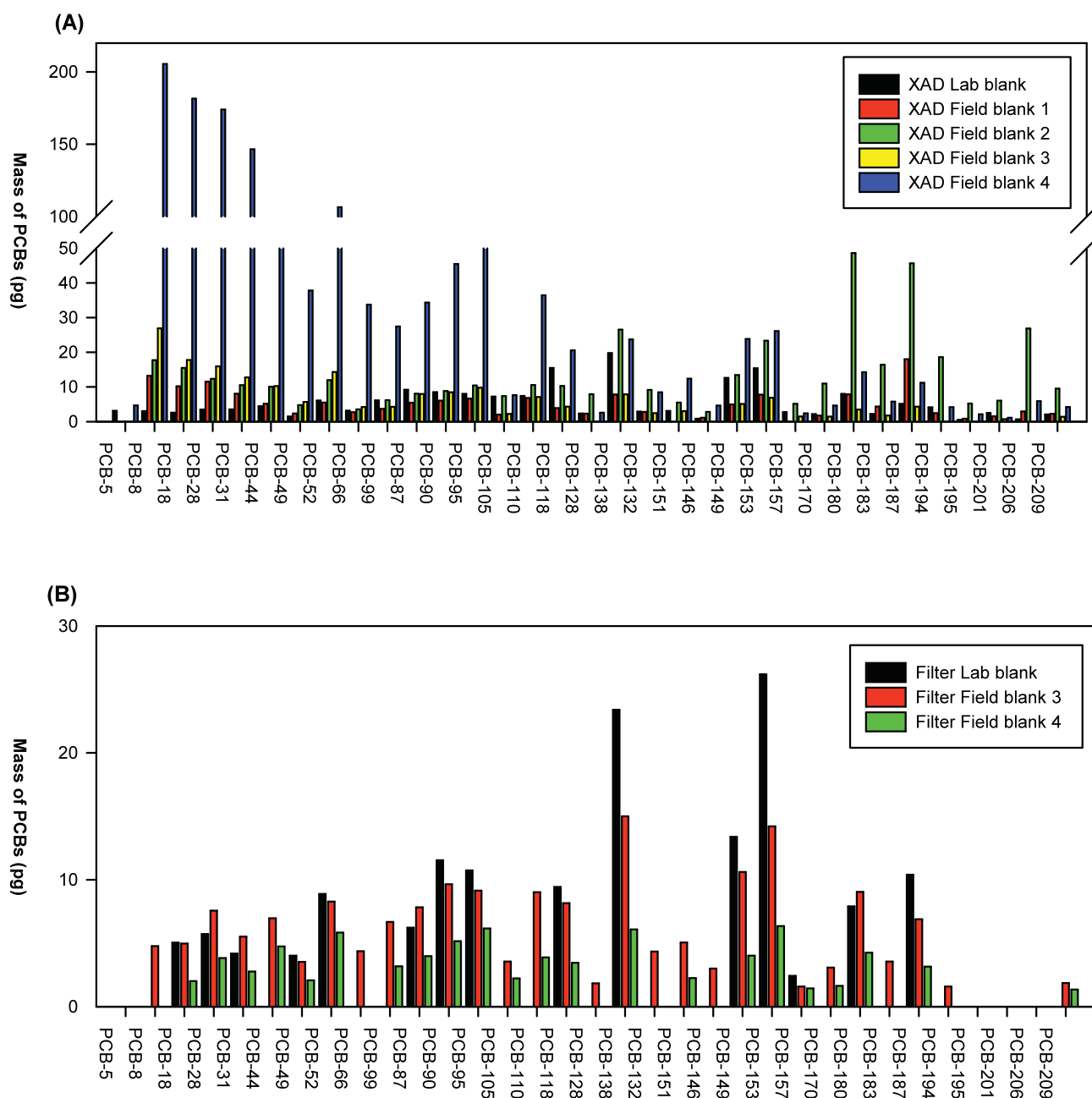


Figure 2. Absolute mass (pg) of PCB congeners in laboratory and field blanks for (a) XAD resin and (b) glass fibre filters.

elevated levels (>30 to ca. 200 pg) of most low- and moderately chlorinated PCB congeners (Figure 2). We do not have an explanation for the elevated levels in this blank, although we note that the congener profile suggests airborne contamination. The associated filter field blank had low levels comparable to those in the other filter blanks (compare scales on Figure 2A and B).

### High-volume samples

The samples contained between 12 and 84 quantifiable PCB congeners, with individual congeners present at, on average, between 8 and 40 pg absolute in the XAD samples, and 4-6 pg absolute in the filter samples. For comparison with blank levels, in Figure 3 we have plotted the absolute amounts of total PCBs (in units of pg) in the samples and blanks against sample volume (L)

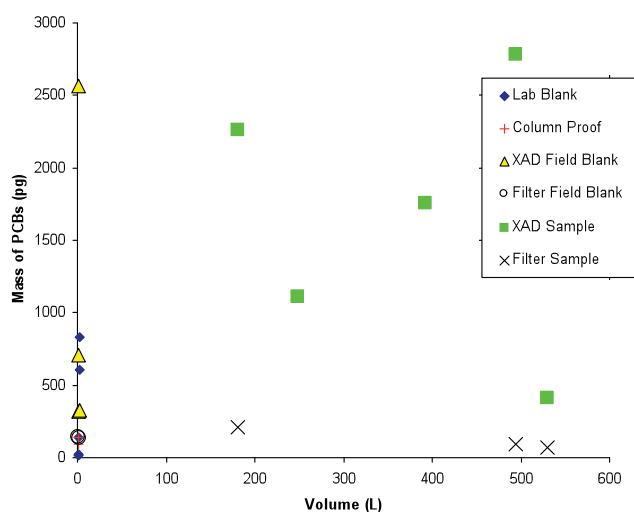


Figure 3. Relationship between sample volume (L) and absolute mass of PCBs (pg) in the samples and blanks. Blanks are shown using nominal volumes of 1-2 L.

using 1-2 L nominal volume for the various blanks. The amounts in the filters are similar to the amounts in the filter field blanks. Among the XAD samples, three contain greater amounts than the majority of the blanks, although not XAD Field blank 4, and XAD Sample 5 has amounts comparable to those in several of the lab and field blanks (Figure 3). Notably, there is no obvious increase in the mass of PCBs in the samples with increasing seawater volume. The sample containing the greatest mass of PCBs (sample 4, 494 L, 2781 pg) was collected back-to-back with XAD Field blank 4.

We interpret these results as indicating that blank levels represent a significant source of contamination for at least some of the samples, interfering with accurate quantification of PCBs. To attempt to correct for the blank levels, we calculated 'blank-corrected' congener concentrations by subtracting the mass of compound in the field blank from the mass of compound in the sample, then dividing this value by the sample volume to convert to concentration. For samples and field blanks collected back-to-back, we used the associated blank and sample values; for the other samples, we used the average field blank value. For the XAD samples, the average was calculated excluding Field blank 4.

Blank-corrected concentrations of 30 prominent congeners are shown in Figure 4 (AMAP list, Muir and Morita, 2003). Di-, tri- and tetrachlorobiphenyls

are the most prominent congeners in the XAD samples, with concentrations of 0.1 pg/L to as much as 1.0 pg/L, compared to concentrations mostly less than 0.01 pg/L for the more highly-chlorinated congeners. In the filter samples, low- and moderately-chlorinated congeners are equally prominent, with concentrations generally about 0.01-0.04 pg/L. Blank-corrected total PCB concentrations (sum of detected congeners) vary from about 0.04 to 0.39 pg/L. Blank-corrected total PCB concentrations in the XAD samples vary from 0.20 to 3.64 pg/L among XAD samples 1, 2, 4 and 5 and reach 10.0 pg/L in sample 3. The relatively elevated concentrations in sample 3 (the lowest-volume sample) may indicate that the blank-correction did not fully address the effect of blank contamination for this sample, although a concentration of 10.0 pg/L is consistent with previous seawater PCB results obtained using similar techniques (in situ pump collection, analysis at AXYS) in the Canadian High Arctic ( $7.0 \pm 5.0$  pg/L) and Resolute Bay ( $10.3 \pm 4.6$  pg/L) (Hargrave et al., 2000; Hargrave et al., 1992). The lower concentrations in the other samples are comparable to the total PCB concentrations (0.54-1.96 pg/L) reported for a study in the north Barents Sea and near the North Pole, in which strict ultra-clean techniques were employed (Gustafsson et al., 2005).

## Conclusions

New data collected in the CFL study area during May-June 2008 suggest that pumping of large volumes of seawater through rigorously cleaned columns handled with clean techniques can produce trustworthy data for PCB concentrations in the Canadian Arctic Ocean. In 2008, high-volume sampling was hindered by unusual ice conditions, which limited the sampling to only five samples (together with four blanks) and slightly lower volumes (181-530 L) than the targeted 500-1000 L. Nevertheless, the rigorous ultra-clean preparation, handling and sampling procedures that were employed to avoid contamination of the samples were generally effective, and low blank levels necessary for good quality-assurance were in most cases achieved. The source of excessive contamination in one field blank requires further investigation.



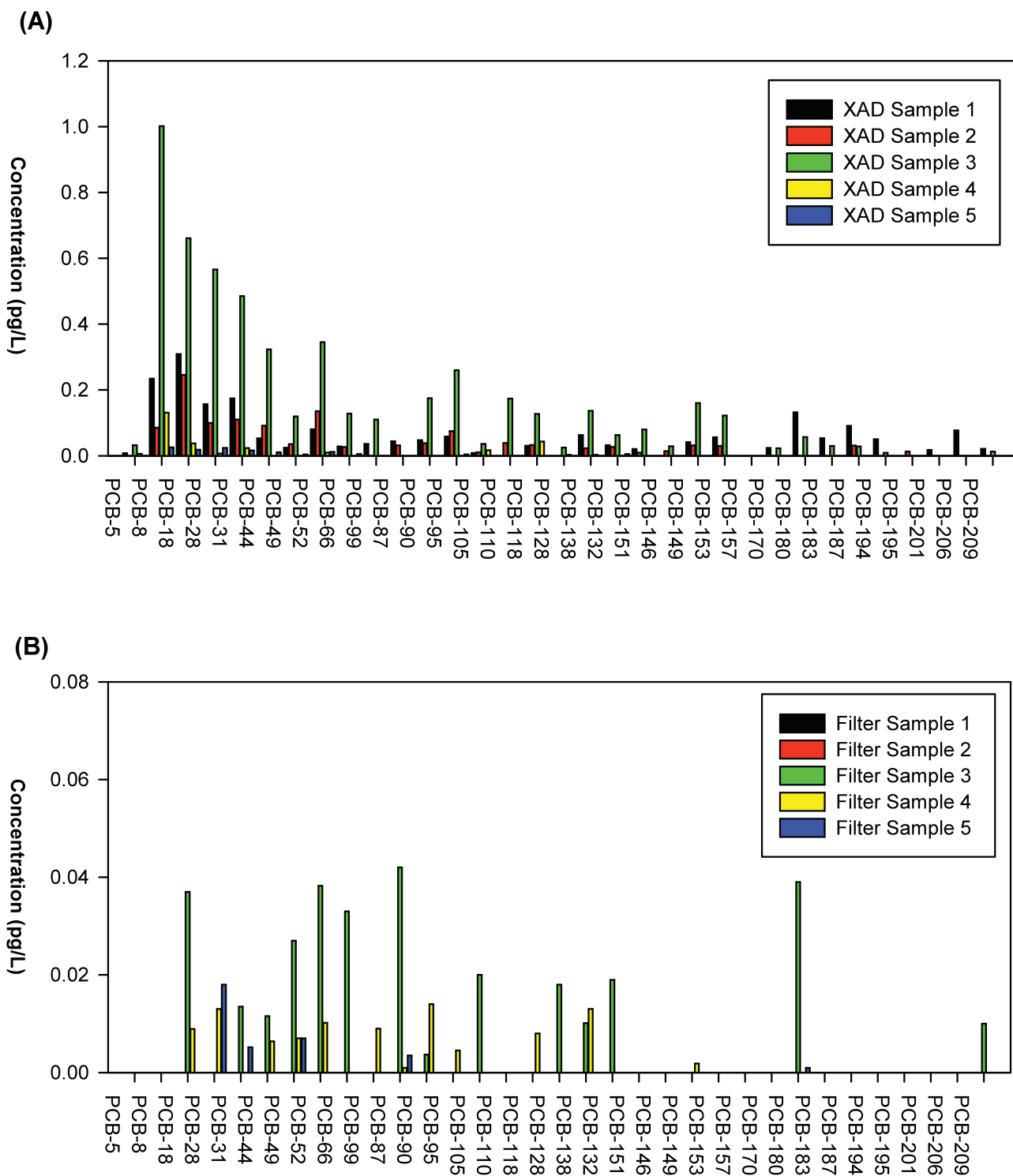


Figure 4. Blank-corrected concentrations of PCB congeners in the five XAD samples (A) and three analyzed filter samples (B).

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After correcting for the levels in the blanks, the concentrations of total PCBs in Canadian Arctic Ocean seawater (dissolved fraction) determined in this study are generally less than 4 pg/L. These values are much lower than concentrations (ca. 40-210 pg/L) reported previously (see, for example, data compiled in the Canadian Arctic Contaminants Assessment Report (CACAR, Muir and Strachan, 2003)) and lend support to proposals (Borga et al., 2005; Gustafsson et al., 2005; Muir and Strachan, 2003; Sobek and Gustafsson, 2004) that previous concentrations were affected by contamination. At 4 pg/L, the top 200 m of the Canada Basin ( $\sim 4 \times 10^6$  km<sup>2</sup>) would contain about 3000 kg of dissolved PCB. To overcome field and laboratory contamination, large volumes of seawater (target 1000 L) need to be collected in a very clean manner. Clearly it would also be helpful to find ways to reduce levels and variability in blanks associated with sample collection.

## Expected Project Completion Date

Additional sampling will be conducted as opportunities arise.

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# Environmental Trends Monitoring of flame retardants and pesticides in the Canadian High Arctic via Ice/Snow

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## Abstract

The purpose of this project was to collect snow and ice cores from the Devon Ice cap on Devon Island in the Canadian Arctic archipelago and measure contaminants such as new persistent organic pollutants and current use pesticides. Many pollutants emitted by industrial and agricultural activities in the south are efficiently removed from the atmosphere by snow. The remote Arctic ice caps on Devon and other Islands, where snow accumulation is preserved with limited summer melting, can therefore provide archives of this airborne pollution. Continuous records of pollution can be developed by taking cores and analyzing them for contaminants. In May 2008 a snow pit (7 m depth) was dug on the Devon ice cap (Devon Island). Snow/ice samples were collected at 25 cm intervals and later extracted under clean room conditions. A wide range of brominated flame retardants (BFRs) and current use pesticides were detected in the Devon ice cap samples. Among the BFRs detected, the predominant chemical

## Résumé

Le but de ce projet consistait à recueillir des carottes de glace et de neige de la calotte glaciaire de Devon sur l'île Devon dans l'archipel de l'Arctique canadien et de mesurer les contaminants tels que les polluants organiques persistants et les pesticides d'usage courant. La neige permet d'éliminer efficacement un grand nombre des polluants émis dans le cadre des activités industrielles et agricoles dans le sud de l'atmosphère. Les calottes glaciaires éloignées de l'île Devon ainsi que d'autres îles, où la neige est préservée par les fontes estivales limitées, peuvent par conséquent nous procurer des archives de la pollution atmosphérique. Il est possible d'établir des enregistrements continus de la pollution en recueillant des échantillons de carottes afin d'y analyser la présence de contaminants. En mai 2008, un puits a été creusé dans la neige (7 m de profondeur) dans la calotte glaciaire Devon (île Devon). Des échantillons de glace et de neige ont été recueillis à des intervalles de 25 cm et extraits par la suite en salle blanche. Un vaste éventail d'ignifugeants bromés (IB) et de

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was decabromodiphenyl ether, a widely used flame retardant. Other flame retardants detected included hexabromocyclododecane and bis(tribromophenoxy) ethane as well as pentabromoethyl benzene. A fifth BFR, decabromodiphenyl ethane, was undetectable.

## Key Messages

- Brominated flame retardants (BFRs) and current use pesticides (CUPs) were detectable in the Devon Island icecap;
- Concentrations of the BFRs varied seasonally and were weakly related to sulphate deposition which is highest in winter months
- The results demonstrate the value of glacial sampling for determining the deposition of some contaminants that are difficult to detect in fish or wildlife

pesticides d'usage courant ont été détectés dans les échantillons de la calotte glaciaire du Devon. Parmi les IB détectés, le produit chimique prédominant était le décabromodiphényléther, produit ignifuge utilisé à grande échelle. Les autres produits ignifuges détectés comprennent l'hexabromocyclododecane et le bis(tribromophénoxy)éthane ainsi que le pentabromoéthylbenzène. Un cinquième ignifugeant bromé, le décabromodiphényléthane, était indétectable.

## Messages clés

- Des ignifugeants bromés (IB) et des pesticides d'usage courant sont détectables dans la calotte glaciaire du Devon.
- Les concentrations d'ignifugeants bromés présentaient des variations saisonnières et une faible relation avec les dépôts de sulfate qui sont plus élevés pendant les mois d'hiver.
- Les résultats démontrent l'importance de recueillir des échantillons de glace afin de déterminer les dépôts de certains contaminants difficiles à déceler dans les poissons ou les animaux sauvages.

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## Objectives

1. Sample snow and ice from a snow pit on Devon Island and analyse for brominated flame retardants, other selected persistent organic chemicals and current use pesticides.
2. Examine the annual trends in deposition of these contaminants to the ice cap using back trajectory modelling.
3. Provide this information to the nearby communities (Grise Fiord) and to the Niquit Avatittinni Committee (Nunavut) on a timely basis.

## Introduction

Ice and snow cores from Canadian Arctic ice caps have been widely used to study current and historical inputs of both natural and anthropogenic (industrial or other) contaminants (Barrie et al. 1985; Gregor et al. 1995; Koerner et al. 1999;

Goto-Azuma and Koerner 2001; Zheng et al. 2003; Krachler et al. 2008).

Nitrate and sulfate aerosols, as well as microparticles carrying metals, are deposited in snow by dry fallout and/or precipitation scavenging (Barrie et al. 1985; Dietz et al. 1998; Kämäri et al. 1998). Summer melting can cause mobilization of water-soluble pollutants. However, usable deposition histories can still be reconstructed at well-studied sites (Koerner 1997; Grunet et al. 1998). Zheng *et al.* (2003) reported a 155 year high resolution reconstruction of lead deposition to the Devon Island icecap. The trend was comparable to that observed in central Greenland (Rosman et al. 1994) although with a later maximum. The results from the Devon icecap (75°N, 82°W) suggest that European sources are important at this site as they are in Greenland.

The situation with organic pollutants is more complex. Both gaseous and particle-bound organic

compounds are subject to efficient snow scavenging (Lei and Wania 2004). Organics with very low vapor pressures (VPs <0.001 Pa at 20°C) are primarily associated with particles at sub-zero temperatures typical of the atmosphere and snow surfaces in the high Arctic (Macdonald et al. 2000). Semi-volatiles (VPs 0.001-0.1 Pa at 20°C) such as PCBs and hexachlorocyclohexanes (HCH) have been shown to revolatilize as the snowpack changes in surface area and density and undergoes melting (Herbert et al. 2005; Burniston et al. 2007). Despite these limitations an increasing number of studies have documented temporal trends of persistent organochlorine (OC) pollutants (Gregor et al. 1995; Donald et al. 1999; Villa et al. 2003; Wang et al. 2008) and polycyclic aromatic hydrocarbons PAHs (Peters et al. 1995) and pesticides (Hermanson et al. 2005) in mid-latitude and Arctic glacial cores.

Decabromodiphenyl ether (BDE 209), the major “additive” brominated flame retardant (BFR) still in wide use, is found in the atmosphere almost exclusively on airborne particles (Su et al. 2007). Therefore, as noted above for metals such as lead, ice cores may also be good reservoirs for BDE209 and for other non-volatile high production volume organic contaminants present in the atmosphere of urban areas mainly on particles. Su et al. (2007) have shown that polybrominated diphenyl ethers (PBDEs) including BDE209 are detectable in air at Alert and are increasing in concentration. Wang et al. (2005) found BDE209 concentrations ranged from <0.5-41 pg/m<sup>3</sup> on particles in the Beaufort Sea. Breivik et al. (2006) examined latitudinal trends of BDE-209 in Canada using dated sediment cores and found that sedimentation fluxes declined exponentially with latitude. Apart from the sediment cores, which had limited temporal resolution due to low sedimentation rates in arctic lakes, there is little temporal trend information for BDE209 or information on its current rate of deposition. The major concern with BDE209 is that it will be debrominated both in the atmosphere, water, sediments and in biota, giving rise to lower brominated and more bioaccumulative and toxic BDEs

While PAHs, organochlorine pesticides and PCBs were determined in the Agassiz ice cap (Ellesmere Is) in the mid-1990s (Gregor et al. 1995; Peters et al. 1995; Franz et al. 1997) very little is known

about deposition of current-use pesticides (CUPs) in the Canadian Arctic. Hermanson et al. (2005) reported a series of CUPs including organophosphorus insecticides (chlorpyrifos, terbufos, diazinon, methyl parathion, and fenitrothion), triazine, dinitroaniline, and chloroacetamide herbicides (atrazine, metolachlor, pendamethalin) in the Austfonna glacier, the largest ice cap in Eurasia, on Svalbard.

## Activities in 2008/2009

### *Sample collection for organic contaminants:*

Samples were collected in May 2008 from the Devon Ice Cap, Nunavut (75° 20.4N, 82° 40.2 W) by our sampling team of Dan Walsh and Torsten Meyer. A 7 m deep snow pit was dug about 2 km upwind from the nearest temporary research site at the summit of the ice cap. Duplicate samples representing about 8L of snow were taken vertically at 25 cm intervals along the face of the pit using 4L polypropylene (PP) bottles. Separate large volume (32 L) samples were collected at 50 cm intervals in PP bottles. Snow density measurements (g/cm<sup>3</sup>) and Ion chemistry samples were collected at 10 cm intervals. All samples were held at <0°C during collection and then were shipped by air-freight to Burlington where they were transferred to commercial freezer space (-20oC) until analysis.

### *Analysis of PBDEs and CUPs:*

Individual 8L snow/ice samples were melted in a clean room at Canada Centre for Inland waters (HEPA and carbon filtered air) and melt water (3-5L) was extracted using XAD-2 resin columns. Larger samples (15-20 L) corresponding approximately to annual deposition layers were extracted separately using the same method. Extracts were screened for 27 individual PBDEs, as well as hexabromocyclododecane, bis(tribromophenoxy) ethane, decabromodiphenyl ethane and penta-bromoethyl benzene using GC-negative ion mass spectrometry (GC-NIMS).

CUPs were isolated and quantified as described by Hermanson et al. (2005). Extracts were screened for 45 CUPs (triazine/acetanilide herbicides, organophosphate (OP) insecticides and miscellaneous fungicides) by GC-low resolution MS (EI and NI modes).

### ***Dating and Flux calculations:***

Subsamples were analysed for sulfate ( $\text{SO}_4^{2-}$ ),  $\text{Cl}^-$ ,  $\text{Na}^+$  and  $\text{Ca}^{2+}$  (National Laboratory for Environmental Testing, inorganics lab). Year of deposition was estimated based on the water chemistry data and previous knowledge of snow deposition rates on the ice cap. Observed concentrations of all organics were corrected for density by multiplying concentrations measured in melt-water by the observed density of the snow. Fluxes were estimated as density-corrected concentration ( $\text{pg/kg}$ )  $\times$  the accumulation ( $\text{kg/m}^2$ ) for each sampling horizon.

## **Results**

### ***Brominated flame retardants:***

A wide range of BFRs were detected in the Devon ice cap samples. Concentrations of major compound groups in the upper 75 cm, representing deposition in 2007 and early 2008, are presented in Table 1. These concentrations are based on melt water and not adjusted for snow density. Highest concentrations by far were the PBDEs which were present at  $\text{ng/L}$  concentrations in all depths. Hexabromocyclododecane (determined as a single compound using GC-NIMS) and bis(tribromophenoxy) ethane were present at similar concentrations while pentabromoethyl benzene was present at sub pictogram/L concentrations. A fifth BFR, decabromodiphenyl ethane, was undetectable. Blank analyses, consisting of XAD resin and solvents ( $N=6$ ) had BFR levels  $<1\%$  of

measured values except for BDE 47 and 99 which ranged from about 30 to 50% of measured values. Method detection limits based on  $3\times\text{SD}$  of blank values were in the range of 0.5-80  $\text{pg/L}$  for BFRs except BDE209 which had a  $\text{MDL} = 530 \text{ pg/L}$ .

Decabromodiphenyl ether (BDE209) was the predominant PBDE followed by debromination products of deca (nona- and octabromoBDEs). BDE209 concentrations were significantly correlated with total nona- and octaBDEs ( $P<0.01$ ) but not with total di-hexaBDEs. BDE209 has been shown to be photolytically debrominated in a wide range of solid matrices (Soderstrom et al. 2004). Thus it is likely that the presence of nona- and octabromo-BDEs is due to debromination during atmospheric transport and during summer sunlight irradiation on the ice cap.

The deposition of BDE209 and other BFRs on the Devon Ice cap varied seasonally. Higher BDE209 coincided with higher sulfate and chloride concentrations in some horizons, however, concentrations were not significantly correlated. Further assessment of the historical variation of BFR deposition in the ice cap awaits dating of the snow pit.

### ***Current use pesticides:***

Only preliminary results for CUPs were available in time for this report. Results were similar to those reported previously (Muir and Zheng 2007). Seven CUPs were detected in almost all recent horizons – dacthal,  $\alpha$ - and  $\beta$ -endosulfan, endosulfan-sulfate, chlorothalonil, metolachlor, metribuzin, trifluralin,

**Table 1. Concentrations ( $\text{pg/L}$ ) of major groups of brominated flame retardants in snow samples from the 0-75 cm depth on the Devon Island ice cap**

Compound	Abbreviation	Mean	range
Pentabromoethyl benzene	PBEB	0.13	$<0.01$ -0.38
Heabromocyclododecane	HBCD	22	3.2-52
Bis(tribromophenoxy) ethane	BTBPE	23	12-43
decabromodiphenyl ethane	DBDPE	$<0.01$	$<0.01$
Polybrominated diphenyl ethers	Total PBDEs	20900	5040-33400
Nonabromo-diphenyl ethers	Total nonas	2060	860-3330
Octabromo-diphenyl ethers	Total octas	631	420-910
Heptabromo-diphenyl ethers	Total hepta	99	55-174
Di- to hexabromo-diphenyl ethers	Total di- to hexa-	266	79-540



phorate. Concentrations of  $\alpha$ -endosulfan and endosulfan sulfate were highest in recent horizons. Most of the CUPs detected are major use chemicals in Canada and the USA, as well as in Europe.

## Discussion and Conclusions

The analysis of snow/firn cores from the Devon Island ice cap has provided the first detailed information on the atmospheric deposition of BFRs in the High Arctic. Other measurements including arctic air (Su et al. 2007) and sediment cores (Breivik et al. 2006) have shown PBDEs are present in the Canadian Arctic but have not provided a high resolution picture of deposition. Combined with the data for CUPs in the ice cap, the results show that a wide range of semi-volatile and rather non-volatile particle borne organics are present in the Devon ice cap. They also illustrate that particle borne organics can be transported to the Arctic and result in similar or higher concentrations in arctic ice caps than many more volatile, environmentally mobile chemicals. Overall these results demonstrate the value of glacier samples and glacial depth profiles for providing information on current and past historical trends of organic chemicals input to the Arctic, which are difficult or impossible to obtain solely from analysis of annual collections of archived tissue samples.

## Project Completion Date

March 31, 2009

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# Improved Screening for New Arctic Contaminants

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## **Abstract**

There is a distinct possibility that some Arctic contaminants exist that have not been identified. They are neither detected, studied, monitored, nor evaluated, let alone regulated. This project uses computer models and a mechanistic understanding of how organic contaminants reach remote polar regions and how they bioaccumulate in the Arctic human food chain to identify currently unrecognized Arctic contaminants. This is achieved by predicting the properties of the multitude of organic substances in commerce and comparing them to the combination of properties that make a substance susceptible to becoming an Arctic contaminant. The influence of the choice of chemical property estimation method was found to be substantial and could lead to errors in screening. Some organic substances have been identified that may have the potential to reach the Arctic by long range oceanic transport. The outcome of the project are lists of substances that merit scientific and regulatory attention by virtue of their persistence, potential for bioaccumulation and potential for long range transport.

## **Résumé**

Il existe une forte possibilité que certains contaminants de l'Arctique n'aient pas encore été identifiés. Ils ne sont ni détectés, ni étudiés, ni contrôlés, ni évalués, ni bien sûr réglementés. Dans le but d'identifier les contaminants de l'Arctique encore non dépistés, le présent projet utilise des modèles informatiques et une compréhension mécanistique de la façon dont les contaminants organiques parviennent dans les régions polaires distantes et s'accumulent biologiquement dans la chaîne alimentaire humaine arctique. Il nous faut pour cela prédire les propriétés de la multitude de substances organiques que l'on trouve dans le commerce et les comparer à un ensemble de propriétés qui fait qu'une substance est susceptible de se transformer en un contaminant de l'Arctique. L'influence du choix de la méthode d'estimation des propriétés chimiques s'est avérée substantielle et pourrait entraîner des erreurs de tri. Il a été déterminé que certaines substances organiques ont le potentiel d'atteindre l'Arctique par le transport océanique à grande distance. Le résultat du projet est une série de listes de substances qui méritent une attention scientifique et réglementaire du fait de leur persistance, et de leurs potentiels d'accumulation biologique et de transport à grande distance.

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## Key Messages

- The choice of property estimation method was found to influence the outcome of Arctic contaminants screening of a significant number of organic chemicals, potentially leading to a large number of wrongly classified substances.
- This suggests that screening methods should preferably not rely on yes/no decisions with respect to partitioning property thresholds.
- A notable number of pesticide chemicals appear to possess partitioning properties that would allow them to undergo long range transport in the ocean, if they are persistent in water.

## Messages clés

- On a trouvé que le choix d'une méthode d'estimation des propriétés influençait les résultats du tri des contaminants de l'Arctique d'un grand nombre de produits chimiques organiques, ce qui pourrait mener à de nombreuses substances mal classées.
- Cela donne à penser que les méthodes de tri ne devraient pas, préférentiellement, relever de décisions par oui ou non pour ce qui est des seuils de propriété de séparation.
- De nombreux pesticides chimiques semblent posséder des propriétés de séparation leur permettant de voyager sur de grandes distances dans l'océan s'ils persistent dans l'eau.

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## Objectives

### *Long Term:*

Use computer models and a mechanistic understanding of how organic contaminants reach remote polar regions and how they bioaccumulate in the Arctic human food chain to identify currently unrecognized Arctic contaminants.

### *Short Term*

- Identify the chemical partitioning property estimation techniques best suited for use in the screening for Arctic contaminants
- Identify among the multitude of organic chemicals those substances that may undergo long range transport to the Arctic in the oceans.
- Improve the description of the atmospheric transport of particle-bound substances in global models used in Arctic contaminant screening.

## Introduction

Simulations with global scale fate and transport models such as Globo-POP make it possible to delineate quite precisely what chemical characteristics make an organic substance susceptible to accumulation in the Arctic (Wania, 2003, 2006). Similar model-based approaches aid in constraining the properties of substances bioaccumulating

in human food chains (Czub & McLachlan, 2004). Building upon the results of those models, Brown & Wania (2008) recently introduced a chemical screening methodology for persistent organic pollutants (POPs), which identifies chemicals that have the properties that allow for accumulation in the physical Arctic environment and in the Arctic human food chain. This method relied upon:

- a large data set of organic chemicals in commerce,
- partitioning and degradation properties of those organic chemicals estimated with the help of the quantitative structure-property relationships implemented in EPIWIN (US EPA, 2009),
- model calculations using hypothetical chemical properties that seek to constrain the property combinations that make a chemical susceptible to bioaccumulation and long range transport (Czub et al., 2008), and
- a parallel screening step that quantified structural resemblance with known POPs using a so-called POP score.

In addition to proving the feasibility and strength of this approach for identifying a list of candidates for Arctic contaminants, the study by Brown & Wania (2008) also identified numerous ways in which this approach could be improved and

expanded. This project thus sought to remedy several shortcomings that had been identified in the screening procedure for unidentified Arctic organic contaminants.

## Activities in 2008/2009

One of the criteria in the screening method by Brown and Wania (2008) is that the chemical needs to have partitioning properties that correspond to potentially elevated Arctic Contamination and Bioaccumulation Potential (AC-BAP). They had defined elevated AC-BAP as any value exceeding 10 % of the maximum AC-BAP value calculated for perfectly persistent organic chemicals. Czub et al. (2008) identified the maximum and the range of partitioning properties that leads to elevated AC-BAP by calculating the AC-BAP for hypothetical chemicals with a range of different combinations of  $K_{AW}$  and  $K_{OA}$ , i.e. the equilibrium partitioning coefficients between air and water

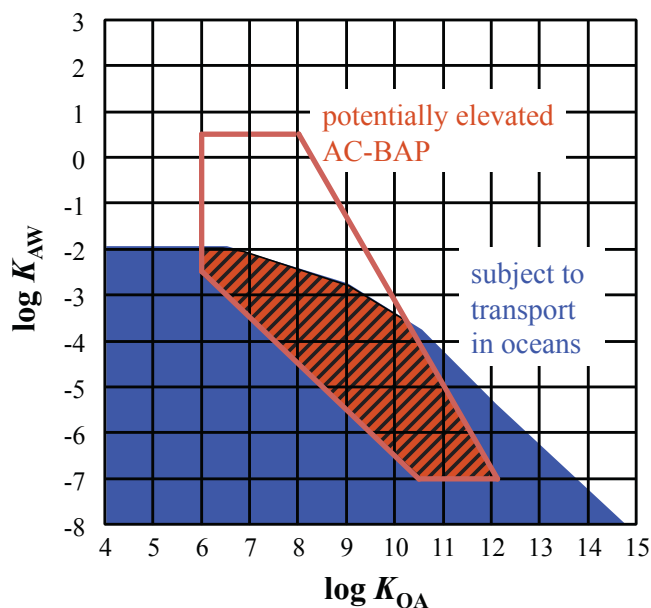


Figure 1. The chemical partitioning space defined by the equilibrium partitioning coefficients between air and water ( $\log K_{AW}$ ) and between octanol and air ( $\log K_{OA}$ ). The red outline designates chemical partitioning property combinations that may lead to a high potential for transport to the Arctic and accumulation in the Arctic marine food chain. The blue are designates chemical partitioning property combinations that allow for transport in the aqueous phase. The hatched area of overlap thus designates substance property combinations that may allow for transport to the arctic with ocean currents and subsequent bioaccumulation.

and between octanol and air.  $K_{AW}$  and  $K_{OA}$  define a coordinate system, also called the chemical partitioning space (Figure 1). The red outline in Figure 1 comprises the partitioning properties corresponding to elevated AC-BAP.

### *Testing the influence of the choice of partitioning property estimation method on the results of the screening for new Arctic contaminants.*

Brown and Wania (2008) used the atom/bond/fragment/group contribution methods implemented in the EPISuite (US EPA, 2009) to estimate the equilibrium partitioning coefficients  $K_{AW}$  and  $K_{OA}$ . Even though the confidence in the validity of the EPIWIN estimates of the screened substances is limited, they had to use them, because no other accessible method can estimate property data for more than 100,000 chemicals so easily. For a subset of approximately 700 organic chemicals, we now predicted  $K_{AW}$  and  $K_{OA}$  with two additional quantitative structure property relationships (QSPR). These were:

- SPARC<sup>1</sup>, a mechanistic perturbation models based on chemical structure (Hilal et al., 2007)
- Prediction of solute descriptors ("Abraham parameters") (ADME Boxes v. 4.1, Pharma Algorithms Inc.), which are then used in poly-parameter Linear Free Energy Relationship (ppLFER) (e.g. Niederer et al., 2006) to predict the partition coefficients

We then compared partitioning properties predicted from different QSPRs, and sought to identify chemicals and functional groups that lead to discrepancies in the partitioning property predictions of different QSPRs. We also evaluated to what extent discrepancies in the estimated partitioning properties affect the outcome of the screening procedure by Brown and Wania (2008), in other words, whether the same or different chemicals fall into the red outline of Figure 1, when different QSPR are being used.

Inherent in the use of a partitioning space defined by  $\log K_{AW}$  and  $\log K_{OA}$  is the assumption that the partitioning properties of octanol are a good surrogate for the partitioning properties of both

<sup>1</sup> SPARC online Calculator, version 4.2. Available online at <http://ibmlc2.chem.uga.edu/sparc/> 2009.



sedimentary and soil organic matter and for the organic matter in atmospheric particles. We explored whether this commonly made assumption has a significant impact on the results for the Arctic Contamination Potential (ACP) and whether these differences impact on the results of screening for elevated ACP. For this purpose, we used the Globo-POP model to calculate the ACP of perfectly persistent chemicals as a function of the three-dimensional partitioning space defined by  $K_{AW}$  (Henry's law constant),  $K_{PA}$  (equilibrium partitioning coefficient between atmospheric particles and gas phase), and  $K_{OC}$  (equilibrium partitioning coefficients between sedimentary and soil organic matter and water). In other words, by doing so, we no longer assumed that the organic matter in soil and sediments has the same partitioning characteristics as the organic matter in aerosol. We then screened a subset of substances, for which solute descriptors are available and which have partitioning properties within the applicability domain of Globo-POP, for elevated ACP by either using the originally developed two-dimensional chemical space map (based on  $K_{AW}$  and  $K_{OA}$ ) by Wania (2003) or the new three-dimensional map (based on  $K_{AW}$ ,  $K_{PA}$ , and  $K_{OC}$ ).

#### ***Screening for chemicals that may undergo long range transport in the oceans.***

One of the chemical properties used in the screening by Brown and Wania (2008) is the atmospheric OH radical oxidation half-life as predicted by EPI Suite (US EPA, 2009). The use of this screening criterion was based on the assumption that long-range transport to the Arctic occurs mainly via atmospheric transport. Even though the global scale model underlying the calculations by Czub et al. (2008) includes a description of oceanic transport, the screening procedure by Brown & Wania (2008) may thus have failed to flag chemicals undergoing long range transport in the oceans by eliminating chemicals that have short half-lives in the atmosphere. There may be substances that have short half lives in air, but are persistent in water. We thus conducted a further screening exercise which explores the possibility of long-range transport to the Arctic in the oceans.

Based on the sensitivity of model results on the input parameters describing oceanic mixing, Wania (2006) defined an area in the  $K_{AW}/K_{OA}$  chemical partitioning space which corresponds to chemicals

referred to as "swimmers". Such chemicals, located in the blue area of the chemical space in Figure 1, have a high potential for transport with water. The intersection of the blue area of the swimmers with the red outline of elevated AC-BAP thus designates substance property combinations that should favour transport of contaminants to the Arctic and the potential for bioaccumulation in an Arctic marine food chain (blue and red hatched area in Figure 1). We screened the large data set of more than 100,000 chemicals compiled in Brown and Wania (2008) to identify those that fall into that hatched area. Chemicals were also screened with the "POP score" as introduced by Brown and Wania (2008) to check for resemblance to known Arctic contaminants. Finally, the resulting list of chemicals was screened for presence on one or more high production volume (HPV) chemical lists indicating an increased potential for environmental emissions.

The partitioning properties as estimated by EPI Suite were used. As was done by Brown and Wania (2008), two data sets were used in chemical space screening, one based on Henry's Law Constants (HLC dataset) and one based on Vapor Pressures (VP dataset), and a chemical was considered to be in the hatched part of the partitioning space if either dataset locates the chemical within the defined areas. Ideally degradation in water should be used as a screening criterion, but the estimation of hydrolysis rate constants in EPI Suite has a very limited domain of applicability and so is not appropriate for screening the entire database of chemicals. The assumption is made here that if a chemical is labile in air (atmospheric OH radical oxidation half-life is less than 2 days) then long-range transport in surface waters will be favored over long-range transport in air. Also, the chemicals that are persistent in air and fall into the red outline would already have been identified in the original screening (Brown and Wania, 2008).

## **Results and Discussion**

#### ***Testing the influence of the choice of partitioning property estimation method on the results of the screening for new Arctic contaminants.***

Partition properties predicted for the same chemical by the four QSPR can vary by as much as 10 orders of magnitude, whereby discrepancies



between the predictions for  $K_{AW}$  tend to be larger than for  $K_{OW}$ . EPI Suite predicted values are closer to the measured ones. This is, however, of limited significance, because the measured data set is strongly biased in favour of EPI Suite, because the database of measured values came from the EPISuite and presumably had been used in the development of the QSPRs implemented in EPISuite.

To test the influence of the choice of partitioning property estimation method on the results of screening, we screened the subset of approximately 700 chemicals for those that have partitioning property combinations corresponding to elevated Arctic Contamination and Bioaccumulation Potential (AC-BAP) using different sets of partitioning properties. The total number of screened chemicals is not always the same, ranging from 624 for SPARC to 713 for EPISuite, because different QSPRs have different applicability domains.

The absolute number of chemicals that falls inside the red outline of Figure 1, and is thus flagged as a chemical with a potentially elevated AC-BAP, is quite similar for different QSPRs, ranging only

from 168 for Absolv to 175 for EPISuite (Figure 2). However, this does not account for the fact, that in each case a slightly different set of chemicals falls inside the red outline. Between 75 and 88 chemicals change in terms of their classification as having potentially elevated AC-BAP depending on the chosen QSPR, i.e. screening results differ for over 10% of the studied chemicals (Figure 3). Clearly, use of different QSPRs will give rise to a significant number of false positive or false negative screening results.

We found that the screening for chemicals with elevated ACP was only marginally affected by the application of octanol as a surrogate for partitioning to organic matter in soils, sediments and aerosol. Chemicals were classified similarly when using approaches based on two- and three-dimensional chemical partitioning maps.

#### ***Screening for chemicals that may undergo long range transport in the oceans.***

Only five chemicals met all of the previously stated screening criteria (within the hatched area of Fig. 1; non persistent in air; high POP score; on HPV list). These are two current use pesticides

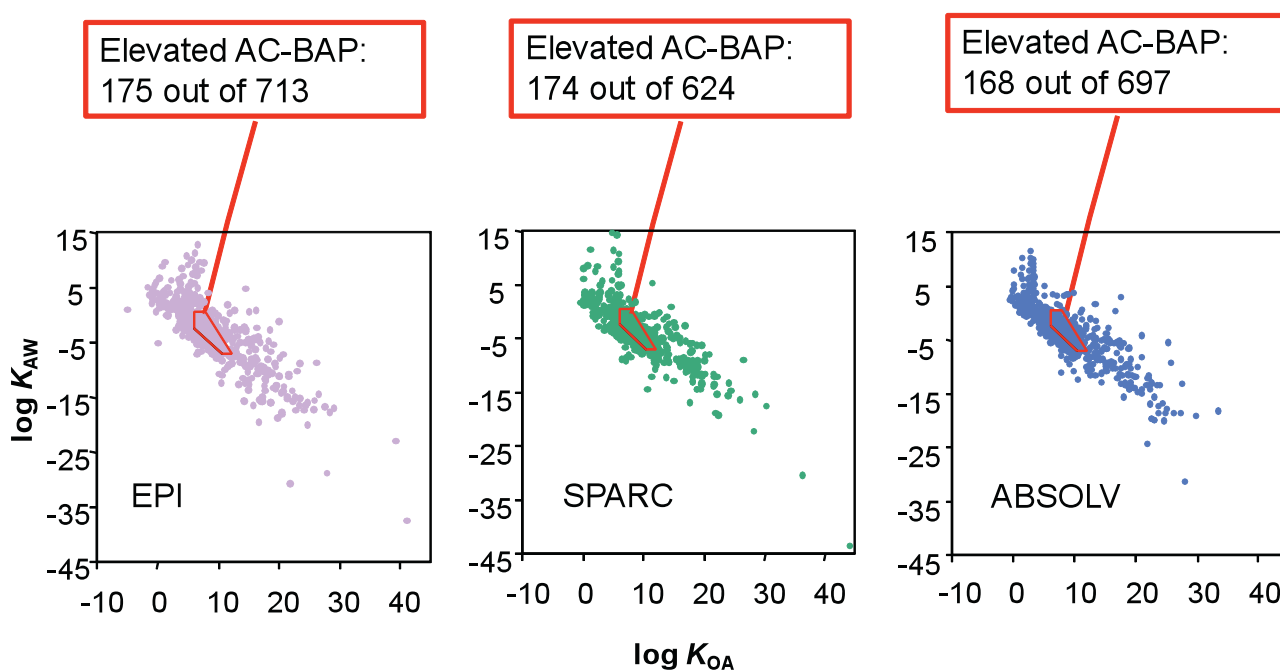


Figure 2. Location of the screened organic chemicals in the partitioning space, if  $K_{AW}$  and  $K_{OA}$  are being estimated with the EPISuite (left panel), SPARC (central panel) or ABSOLV (right panel). The number of screened chemicals and the number of chemicals falling inside the red outline, indicating a potential for elevated AC-BAP, is indicated above the panels.

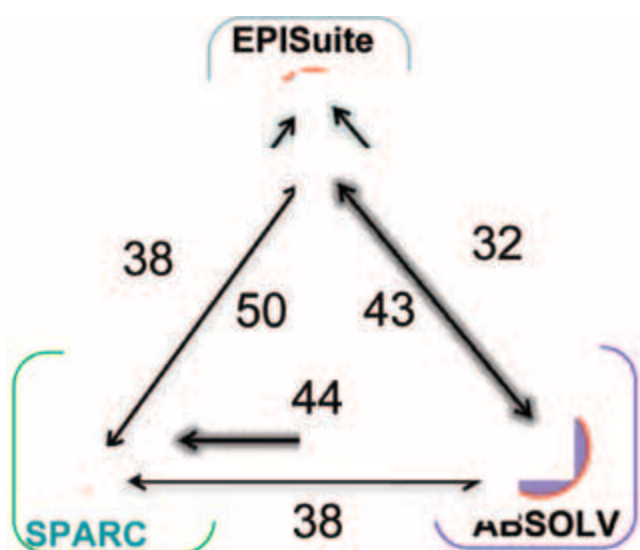


Figure 3. Number of screened chemicals that fall on different sides of the red outline in the  $K_{AW}/K_{OA}$  partitioning space depending on the property estimation method. For example, 44 of the substances that had been flagged as having high AC-BAP by ABSOLV, did not get flagged by SPARC.

endosulfan (CAS 11-52-97) and anilazine (CAS 10-10-53), two flame retardants chlorendic anhydride (CAS 11-52-75) and 1,3,5-tribromo-2-(2-propenyloxy)-benzene (CAS 327-88-95), and chlordene<sup>2</sup> (CAS 373-44-83), an intermediate in the manufacture of cyclopentadiene pesticides. The structure of these compounds is shown in Figure 4.

POP-like HPV chemicals which are persistent in air, but meet the other screening criteria for transport to the Arctic in ocean water include  $\gamma$ -hexachlorocyclohexane (lindane), hexachlorobenzene, pentachloronitrobenzene and a series of chlorinated cyclopentanes and cyclopentenones. These latter compounds had already been on the original list of potential Arctic contaminants by Brown and Wania (2008).

There are a total of 376 HPV chemicals which meet the screening criteria for transport to the Arctic in ocean water, but do not resemble existing

<sup>2</sup> 4,7-methano-1H-indene, 4,5,6,7,8-hexachloro-3a,4,7,7a-tetrahydro-

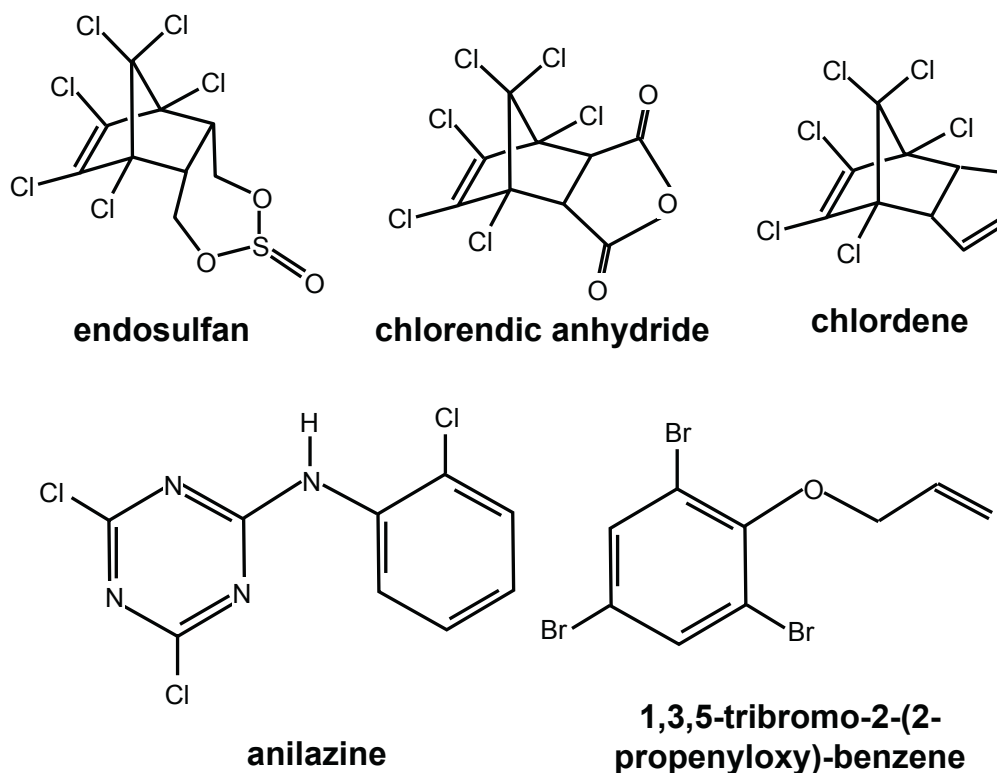


Figure 4. Structure of some high production of volume chemicals that have predicted properties that may make them potential Arctic contaminants, that can reach the Arctic by ocean transport.

POPs structurally (low POP score); 315 chemicals identified by the HLC dataset and 277 chemicals identified by the VP dataset. Many of these chemicals are either phenols or contain other phenoxy groups (21%) or are esters and organic acids (24%). Small aromatics like benzene and naphthalenes make up much of the remainder (30%). An interesting result is that 64 of the chemicals are pesticides; a fairly large portion of the identified chemicals (17%). Some pesticides included in the list are fenthion, parathion, diazinon, trifluralin, chlorpyrifos and alachlor. Out of the 64 pesticides 18 are also found on the US EPA Toxic Release Inventory (TRI).

## Conclusions

While useful in generating lists of substances that may require scientific and regulatory scrutiny, a screening that depends on the comparison of predicted substance properties against firm cut-off values inevitably suffers from errors that are associated with uncertain property predictions. In the future, screening efforts should be directed towards establishing chemical rankings based on a quantitative hazard evaluation. Such efforts should not ignore the uncertainties of predicted chemical properties but incorporate them in the procedure in an effort to identify and reduce the uncertainty that is most influential.

## Expected Project Completion Date

Work on additional improvements to the screening for new Arctic contaminants is currently ongoing. It is anticipated that the project will be fully completed by December 2009.

## Acknowledgments

Eldbjørg S. Heimstad of the Norwegian Institute of Air Research in Tromsø is acknowledged for calculating Abraham solute descriptor for a large number of organic substances.

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# Enhanced investigations of the factors affecting long-term contaminant trends in predatory fish in Great Slave Lake, the Northwest Territories

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## Abstract

We have been conducting enhanced studies of Great Slave Lake in order to better understand the factors affecting the different contaminant trends we are seeing in lake trout and burbot. We have examined the long-term weather record and note that there is a trend for air temperatures and precipitation to be increasing. Higher air temperatures may be enhancing productivity and growth rates for organisms living in the lake; higher precipitation may bring more nutrients and contaminants into the lake, including mercury. We also have examined the commercial fish record for the West Basin. This fishery focuses on whitefish and, while significant, has been operating below annual quotas. There is a trend of declining whitefish and lake trout catches in recent years (1989-2002) but no trend in mean whitefish age; lake trout age data have not been located and/or examined. While the data are incomplete, the commercial fishery does not appear to be changing dramatically with the concomitant potential to exert strong influences on fish biology and, ultimately, contaminant body burdens for West Basin fish, including lake trout: the fishery does not operate

## Résumé

Nous avons effectué des études améliorées du Grand lac des Esclaves afin de mieux comprendre les facteurs qui influencent les tendances des divers contaminants que nous observons chez la truite grise et la lotte. Nous avons étudié les dossiers météorologiques à long terme et remarquons que la température de l'air et les précipitations montrent une tendance à la hausse. Des températures de l'air plus élevées peuvent améliorer les taux de productivité et de croissance des organismes qui vivent dans le lac; des précipitations plus abondantes peuvent augmenter l'apport des nutriments et des contaminants dans le lac, y compris du mercure. Nous avons aussi étudié les dossiers des poissons commerciaux du bassin ouest. Cette pêche se concentre sur le ménomini et même si elle est importante, elle se situe sous les quotas annuels. On a observé une tendance à la baisse des prises de ménomini et de truite grise depuis quelques années (1989-2002), mais aucune tendance en ce qui concerne l'âge moyen du ménomini; les données sur l'âge de la truite grise n'ont pas encore été recueillies ni étudiées. Même si les données sont incomplètes, la pêche commerciale ne semble pas changer

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in the East Arm. Burbot collected from Lutsel K'e have been analyzed for mercury and show the same trend of increasing concentration as is being observed in the West Basin. Sediment cores were successfully collected in March 2009 and then later dated; chronology was very good. Examination of these cores for contaminant and productivity trends will help elucidate the factors driving trends in mercury, HCH, DDT, PCBs, etc. in Great Slave Lake burbot and lake trout. These factors will differ with the compound considered although changes in atmospheric inputs, climate, and nutrient inputs are believed to be the major drivers.

## Key Messages

- There is evidence of a long-term trend of increasing air temperature and precipitation for Great Slave Lake based on meteorological data for Yellowknife. These trends may be enhancing lake productivity and contaminant delivery to the lake from the watershed.
- There is less of evidence of a strong long-term change in the commercial fishery for the West Basin although whitefish and lake trout catches have been declining in recent years (1989-2002), most likely because of reduced fishing effort. This has not had strong impact on whitefish age. Thus, changes in the commercial fishery are not likely to be having a strong impact on observed trends in lipids and contaminant levels in lake trout. The commercial fishery does not operate in the East Arm. Burbot are a minor constituent of the commercial fishery.
- Burbot analyzed for mercury from the Lutsel K'e area of the East Arm are showing the same trend of increasing mercury as is being observed in burbot from the West Basin.
- Examination of dated sediment cores collected from Great Slave Lake for contaminant and productivity trends will help elucidate the

substantiellement avec le potentiel concomitant d'exercer de fortes influences sur la biologie des poissons et, en fin de compte, sur les fardeaux corporels des contaminants des poissons du bassin ouest, y compris la truite grise : il n'y a pas de pêche dans le bras oriental. Les lottes recueillies à Lutsel K'e ont été analysées quant au mercure et affichent les mêmes tendances à l'augmentation des concentrations que dans le bassin ouest. Des carottes sédimentaires ont été recueillies en mars 2009 et ont été datées par la suite; la chronologie était très bonne. L'étude de ces carottes pour ce qui est des tendances des contaminants et de la productivité permettra d'élucider les facteurs qui influencent les tendances relatives au mercure, aux HCH, au DDT, aux BPC, etc. chez la truite grise et la lotte du Grand lac des Esclaves. Ces facteurs différeront du composé étudié même que l'on croit que les principaux éléments sont les changements au niveau des apports atmosphériques, du climat et de l'apport des nutriments.

## Messages clés

- Il y a preuve d'une tendance à long terme vers une augmentation de la température de l'air et des précipitations au Grand lac des Esclaves, à partir des données météorologiques de Yellowknife. Ces tendances peuvent améliorer la productivité du lac et l'apport des contaminants au lac à partir du bassin hydrographique.
- La preuve est moindre d'un changement important à long terme de la pêche commerciale dans le bassin ouest, même si les prises de ménomini et de truite grise baissent depuis quelques années (1989-2002), probablement à cause d'une activité de pêche moindre. Cela n'a pas eu d'impact important sur l'âge du ménomini. Ainsi, les changements de la pêche commerciale n'auront probablement pas de conséquences importantes sur les tendances observées dans les niveaux de lipides et de contaminants chez la truite grise. Il n'y a pas de pêche commerciale dans le bras oriental. La lotte est un élément peu important de la pêche commerciale.
- La lotte analysée quant au mercure dans la région de Lutsel K'e dans le bras oriental montre la même tendance à la hausse du mercure que chez la lotte du bassin ouest.

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drivers affecting the different contaminant (mercury, HCH, DDT, PCB) trends being observed in lake trout and burbot collected from different areas of the lake.

- L'étude des carottes sédimentaires datées prélevées dans le Grand lac des Esclaves pour ce qui est des tendances relatives aux contaminants et à la productivité permettra d'élucider les facteurs qui influencent les tendances des différents contaminants (mercure, HCH, DDT, BPC) que l'on observe chez la truite grise et la lotte de différentes parties du lac.

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## Objectives

### *Short-term*

1. Collect and date sediment cores from the three regions in Great Slave Lake where lake trout and burbot are being analyzed for contaminant trends
2. As part of the sediment core sampling, collect water quality and plankton data for comparisons with observations made during earlier sampling periods and assess for changes in the winter limnology of Great Slave Lake.
3. Resume burbot sampling at Lutsel K'e to strengthen the long-term record for temporal trends in contaminants in the Great Slave Lake ecosystem.
4. Begin to review and synthesize the historic record for Great Slave Lake to assess trends in climate, commercial fish catches, and nutrient inputs.
5. Analyze archived invertebrate and fish samples from 1993, 1994 and 1995 collections for mercury.
3. Determine organic contaminant and mercury concentrations in various components of the Great Slave Lake food web, focussing on the Lutsel K'e and Fort Resolution areas and assess whether concentrations and biomagnification rates have changed since the mid 1990s when similar work was conducted. This research will be done in 2010.
4. As part of the 2010 food web sampling, determine benthos and plankton abundances and composition in the West Basin and East Arm and compare to similar collections made by us in 1994 and 1995. This will contribute to our assessments of changes in standing stocks and productivity in Great Slave Lake.
5. As part of meeting objectives 1-5, develop and test a series of hypotheses of the factors affecting the individual contaminant trends being observed in lake trout and burbot in various regions of Great Slave Lake.

### *Long-term*

1. Using dated sediment cores, determine time trends in persistent organic contaminants, mercury, and other metals in sediments in three regions of Great Slave Lake (West Basin near Hay River and near the Slave River at Fort Resolution, East Arm at Lutsel K'e). As part of this objective, investigate the relationship between time trends in contaminant concentrations and fluxes with contaminant time trends in fish collected from the same areas of the lake.
2. Determine time trends in the productivity at these three locations through chemical and microscopic analyses of these cores. Changes in lake productivity will affect various aspects of contaminant uptake and bioaccumulation by fish.
6. Work with local and other agencies to develop a long-term monitoring program for Great Slave Lake which will complement NCP's contaminant trend monitoring program. Also work to develop a research arm to this program. Together these programs should allow for improved community training and participation.
7. Participate in and contribute information to AMAP expert work groups for trend monitoring for POPs and mercury.
8. Communicate results to the communities and the commercial fisheries in a timely manner.



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## Introduction

The Northern Contaminants Program (NCP) has many objectives including determining contaminant levels and trends in biota such as fish which are important in traditional diets. Trend assessment is crucial for evaluating the success of international controls in reducing contaminant inputs to the north. However, a number of factors may enhance or mask contaminant trends. For example, while there is a continuing trend of reduced contaminant use and release in many parts of the developed world, large-scale developing economies such as those in India and China may be increasing contaminant release for compounds associated with those economies, e.g., mercury with the increased growth in the number and size of coal-fired power plants. Furthermore, global warming is a now recognized issue of concern with a multitude of implications to the functioning of aquatic ecosystems including productivity and contaminant biomagnification (Rouse et al. 1997; ACIA 2005; Prowse et al. 2006; AMAP 2006; Outridge et al. 2007).

Unlike the earlier NCP contaminant studies which were conducted in the 1990s and were broad-based in approach, considering contaminants in fish, invertebrates, and sediments in a number of aquatic ecosystems, the current NCP program is more focused and designed to detect time trends in key indicator organisms such as lake trout and burbot. For fish, in addition to contaminants being measured, a number of influential variables are measured, i.e., fish length, weight, gender, age, lipid content,  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  (Bache et al., 1972; Rasmussen et al. 1990; Bentzen et al. 1996; Evans et al. 2005 a, b). Other components of the ecosystem are not being measured although changes in these components may affect the biology of the fish and hence their contaminant body burdens, e.g., changes in trophic status, algal assemblages, carbon sources, etc. (Berglund et al. 2001; Houde et al. 2008). Long-term fish monitoring studies as part of the NCP trends program are beginning to detect trends in contaminant levels. For example, for lake trout from Great Slave Lake, mercury levels appear to be increasing and HCH and DDT levels decreasing while PCB trends are less evident. However, lipid levels are showing a decline over the same period and there is some evidence that lake trout are feeding more on littoral carbon than

in the mid 1990s (Evans and Muir 2007, 2008, 2009). A somewhat similar situation is occurring in Lake Laberge with lipid levels declining and growth rates increasing in lake trout (Ryan et al. 2005; Ryan 2006; Stern et al. 2008). The factors affecting these trends in fish contaminant body burden and biology are poorly understood. In a thorough study of contaminant trend monitoring of fish in Lake Ontario, Bentzen et al. (1999) recommended that a more appropriate design for contaminant monitoring of key species is to supplement such programs with periodic (ca. every 5 years) intensive sampling of the entire food web.

This project addresses the issue of the factors affecting the individual contaminant trends in lake trout and burbot in Great Slave Lake through a multipronged approach. It builds on previous NCP and other supported studies conducted in Great Slave Lake. It follows a somewhat similar approach as recently conducted by Ryan (2005; Ryan et al. 2006) on Lake Laberge where the sediment record was examined to assess long-term trends in contaminant inputs and changes in productivity, and food webs to assess changes in contaminant biomagnification rates and food web structure. In contrast to Lake Laberge, there is an extensive data base for Great Slave Lake including commercial fish records, water quality monitoring at Fort Fitzgerald on the Slave River, and high quality invertebrate and plankton collections which will allow us to place current conditions into a broader historic perspective. Great Slave Lake has experienced relatively few perturbations to its fishery with the commercial fishery operating since 1945 with catches to this day below quotas; unlike the Great Lakes, Great Slave Lake has not been invaded by exotics nor stocked with non-indigenous species. On the other hand, Great Slave Lake does present more of a challenge than Lake Laberge as the lake is large and complex with different ecological zones based primarily on depth, Slave River inflow, and local geology (Rawson 1950, 1951, 1953, 1956; Fee et al. 1985).

## Activities in 2008/2009

Activities in 2008-2009 focussed in three general areas. The first area was to improve our data base for assessing contaminant trends in lake trout and burbot and to work towards modelling contaminant biomagnification in lake trout and burbot food

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webs in the various regions of Great Slave Lake where the fish are being monitored. Accordingly we:

1. Located archived invertebrate and fish samples from 1993, 1994 and 1995 collections and submitted them for mercury analyses (Evans 1994, 1995). This will extend our mercury trend data for lake trout and burbot to 1993. It also will allow us to investigate mercury biomagnification for a food web at Lutsel K'e in the East Arm and near Fort Resolution in the West Basin for the early 1990s which can be compared with similar food webs in 2010.
2. Burbot were collected from Lutsel K'e for mercury and organochlorine analyses. Burbot monitoring at Lutsel K'e ceased after 2004. There were a number of reasons to continue this monitoring including the desire to compare contaminant trends in two species of fish being collected in the same location. In the West Basin, burbot are being monitored near the Slave River and lake trout north of Hay River.
3. Northern pike were captured during the burbot collections at Fort Resolution. Twenty fish were retained and were submitted for mercury analyses. This will extend our northern pike record which, when combined with the commercial fish record, goes back into the mid 1970s.

Our second general area of activity was to begin to synthesize the existing knowledge of the Great Slave Lake ecosystem. We have examined the long-term climate record for Yellowknife and the commercial fish record. In addition, we are examining the long-term water quality record for the Slave River. Changes in climate, fishing pressures, and nutrient inputs all have the potential to affect contaminant pathways and trends in lake trout and burbot.

Our third general area of activity was sediment coring studies. In March 2009, we collected sediment cores from the Hay River (site 12), Fort Resolution (site 19) areas which we previously investigated in 1994 (Evans et al. 1996). In addition, we collected a sediment core near Lutsel K'e in the East Arm. These cores were submitted for dating analyses. In addition, we conducted limnological investigations at each site including

temperature, oxygen, pH and conductivity profiles; water for nutrient and chlorophyll analyses; and water for phytoplankton analyses. The findings from these studies can be compared with our earlier research (Evans 1996). Dr. Stephanie Guildford from the University of Minnesota contributed to this study, focussing on measuring algal pigment composition and photosynthetic capacity with depth; this work was part of a larger program we were conducting in lakes in the Inuvik area (Ogbebo 2009 a, b). We expect to work with Dr. Guildford again in 2010.

## Results

The long-term climate record at Yellowknife is showing a clear trend for increasing temperatures and precipitation over the May-September period (Fig 1); similar trends are evident on an annual basis. Warmer temperatures should enhance productivity and growth rates while higher precipitation rates should enhance the delivery of nutrients and other chemicals from the watershed into the lake.

We have started to examine the commercial fish record focusing on the combined catch for the West Basin and total catches for whitefish, the focus of the fishery, and lake trout for trends (Yaremchuk et al. 1989; Low and Read 1987, 1993; Low et al. 1989; Read and Taptuna 1995, 1997, 2001, 2003). In the early years of the fishery (Fig. 2), lake trout were caught in large numbers but since the 1970s, the fishery has been directed towards whitefish. The commercial quota has ranged from 2,175,000 kg over 1975-1976 to 1,545,455 kg over 1976-1978, and was 1,727,400 kg for the 1990-2002 season. Thus, the fishery has been operating below its quota. Whitefish catches have been declining since the late 1980s; mean age shows little change over 1983-2002 suggesting the decline in catch is not due to overharvesting but possibly reduced fishing effort. Tallman and Friesen (2007) reported that whitefish were increasing in mean age and size over 1972-1995 which they related to low exploitation pressures. Lake trout are showing a somewhat similar trend of reduced catches. Therefore, unlike as in Lake Laberge, changes in contaminant levels and lipids in lake trout in Great Slave Lake do not appear to be driven by a drastic relaxation in commercial

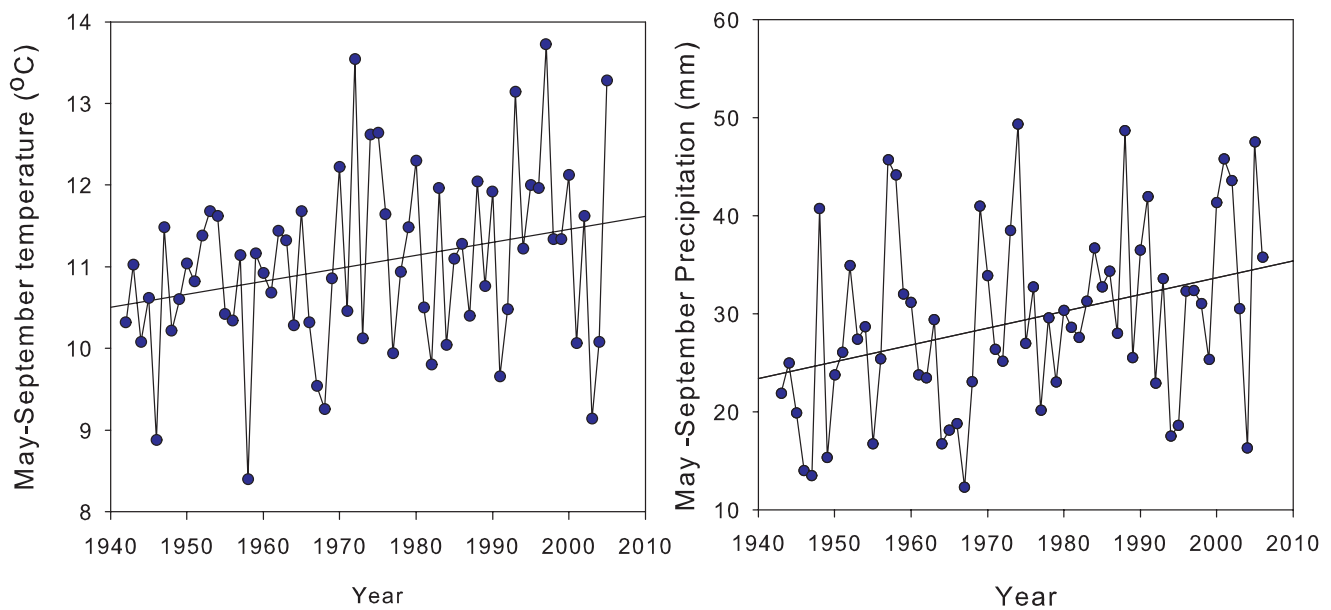


Figure 1. Temperature and precipitation trends for Yellowknife.

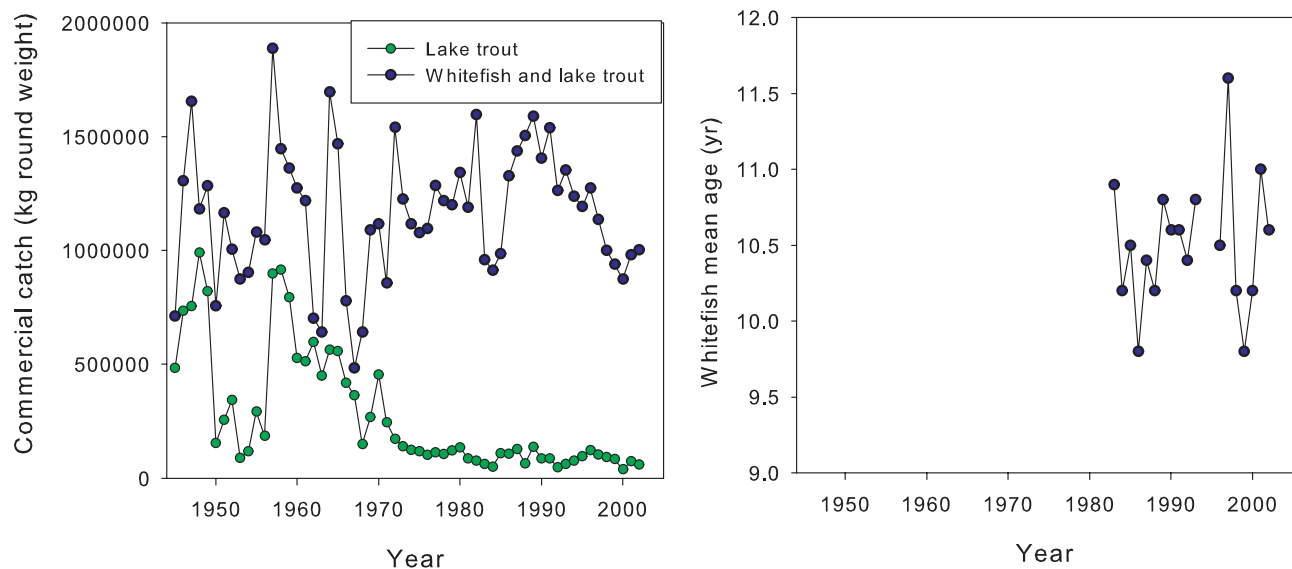


Figure 2. Trends in fish catch and mean age of lake whitefish for the commercial fishery on Great Slave Lake.

fishing pressures with a concomitant change in lake trout (and whitefish) mean age, size, and growth rates. Burbot are a minor component of the commercial fishery.

A recently released report by Glozier et al. (2009) states that total phosphorus and dissolved phosphorus concentrations increased in the Slave River at Fort Fitzgerald over 1989-2006 but not 1996-2006; this seems to support Evans et al.'s (1996)

reported increase in lake productivity in the West Basin based on organic carbon measurements of cores collected offshore of the Slave River and Hay River. It is not clear why a trend of increased phosphorus concentrations is not evident in the 1996-2006 record but may be related to sample size and/or Slave River flow rates which should be higher with greater precipitation; this could result in nutrient dilution. Examination of the more

recent sediment core record and flow rates should help clarify these trends.

Burbot from Lutsel K'e have been examined for mercury while organochlorine analyses are ongoing; burbot are showing a continuing trend of mercury increase (Fig. 3) although the causal factors have not been determined. Mercury also is increasing in burbot from Fort Resolution with the more recent values the highest on record. Some of the more recent mercury increases in Fort Resolution burbot may be related to an increase in the size of the burbot provided for analyses.

The March 2009 sediment coring on Great Slave Lake was successful although cold weather hampered some of our collections. A sediment core was collected in the extreme west end of the West Basin at site 12 (Evans et al. 1996) and represents the general area where lake trout are being sampled; the Slave River influence (with respect to suspended sediments) is weakened here and the site may be influenced by the Hay River. The second core was collected at site 19, northwest of Fort Resolution and in an area of strong Slave River influence, but in an area where datable cores could be obtained. The third core was collected in the East Arm, near Lutsel K'e, in an inlet where sedimentation rates were sufficiently high as to provide for good time resolution. All cores have

been dated and provide sediment records which are in general agreement with cores collected in March 1994 and 1995 (Fig. 4). The  $^{137}\text{Cs}$  maximum agrees well with  $^{210}\text{Pb}$ -assigned dates. The average sedimentation rates are  $348 \text{ g/m}^2/\text{yr}$  for site 12,  $532 \text{ g/m}^2/\text{yr}$  for site 19, and  $654 \text{ g/m}^2/\text{yr}$  for the East Arm site; adjustments have not been made for focussing. Furthermore, sedimentation rates have varied over time as has been observed in Lake Laberge (Ryan 2006) and a large multitude of lakes in northern Canada as recently reported in Muir et al. (2009). Sedimentation rates have increased most markedly with time at the East Arm site and least for site 12.

Limnological sampling was conducted as part of the March 2009 study with temperature, oxygen, pH and conductivity measured with depth. Water was collected from two depths and measured for chlorophyll, nitrite-nitrate, ammonia, dissolved phosphorus, dissolved organic carbon, total phosphorus, particulate organic carbon, and particulate nitrogen. Phytoplankton also was preserved for algal composition and abundance studies.

Specific conductivity illustrates several features of the physical limnology of Great Slave Lake (Fig. 5). Specific conductivity was lower at the two West Basin sites in 2006 than 1994. For site 12, a higher conductivity layer was observed in the upper 20 m

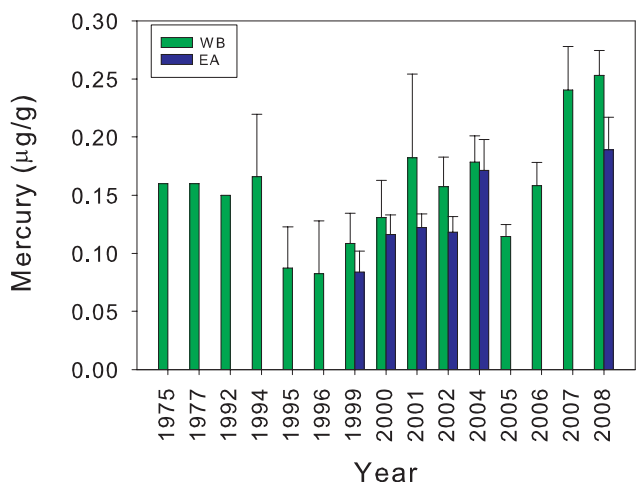


Figure 3. Time trends in mercury concentrations in burbot from the West Basin (green) and East Arm at Lutsel K'e (blue). Since 1993, West Basin fish have been caught from the Fort Resolution area while fish caught at earlier times were from commercial fishery.

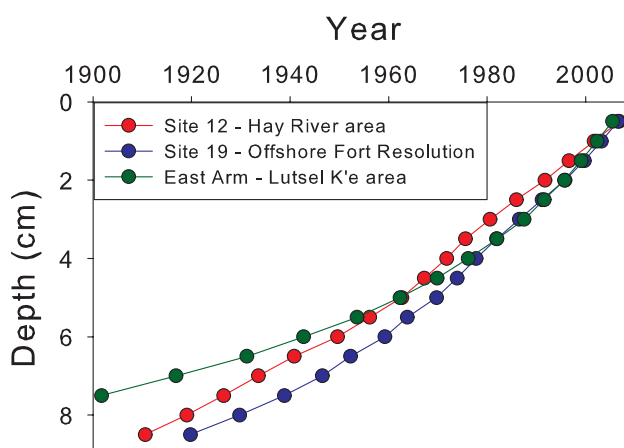


Figure 4. Assigned dates (constant rate of supply model) to sediment cores collected in Great Slave Lake, March 2009.

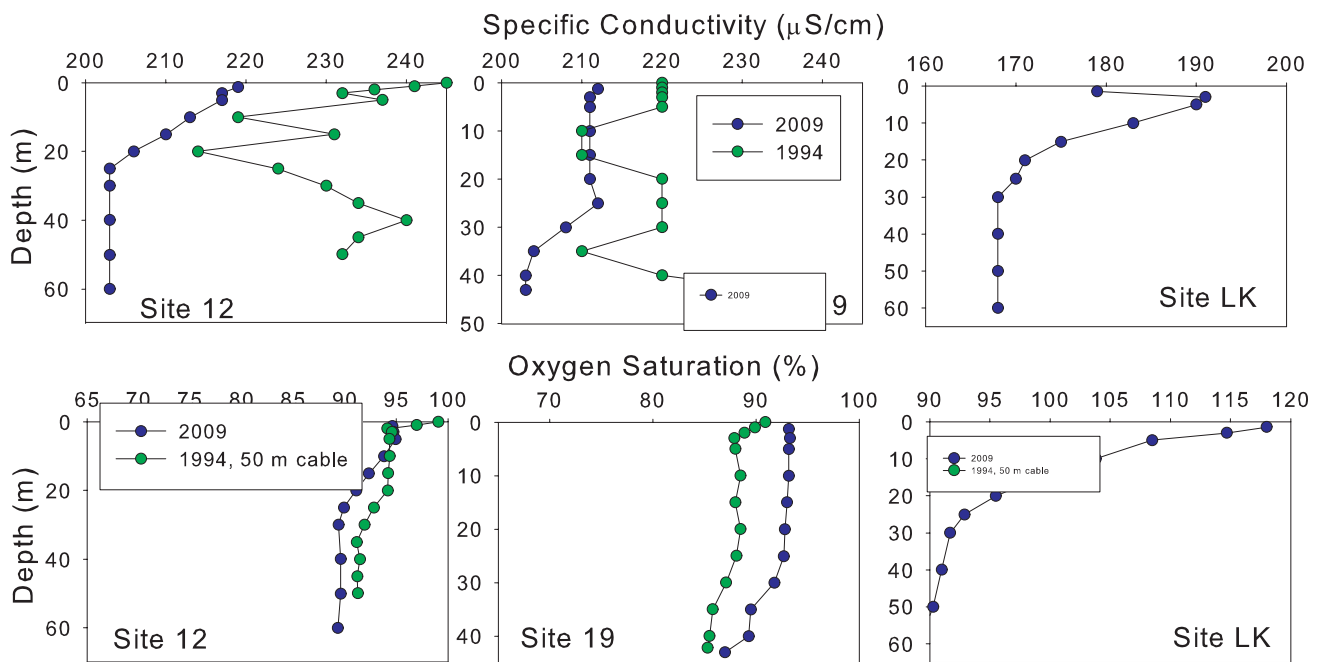


Figure 5. Specific conductivity and percent oxygen saturation as determined at sites 12 (offshore of Hay River) and 12 (offshore of the Slave River) in the West Basin and site LK near Lutsel K'e in the East Arm in 1996 (green dots) and 2009 (blue dots). In 1996, sampling was conducted only to 50 m because of instrument cable length.

of the water column in both years; this water may have been from the Hay River and/or Slave River. Interestingly, in both years, conductivity was lower in the upper 20 m of the water column at site 19 than site 12 although site 19 is closer to the Slave River; possibly at this time of the year, site 19 was being influenced by low-conductivity outflow from the East Arm. Conductivity was substantially lower at the Lutsel K'e site, reflecting the weaker influence of the Slave River in the East Arm and geological influences. Again, conductivity was higher in the upper 30 m than deeper waters with some indication of lower conductivity inflow just under the ice.

As is typical with winter months, oxygen concentrations and percent saturation were high. Percent saturation was less at site 12 in 2006 than 1996 while the converse was observed at site 19 (Fig. 5). Oxygen saturation was surprisingly high at the Lutsel K'e site. This was unexpected and may be an instrument error. On the other hand, there appeared to be algal bloom occurring under the ice based on other field measurements (fluorometric).

## Conclusions

Our study to date has shown that Great Slave Lake is undergoing various long-term changes with mean air temperature and precipitation increasing and an apparent trend of increasing phosphorus inputs to the lake via the Slave River; the sediment core record up to 1994 also suggested that lake productivity was increasing. The commercial fishery is showing evidence of a decline in whitefish and lake trout catches although the fishery continues to operate below the assigned quota. There is no shift in the mean age of lake whitefish over 1993-2002 suggesting, on a first look, that changes in commercial fishery catches are unlikely to be strongly influencing whitefish and, by inference, lake trout growth rates, lipid concentrations, and ultimately, contaminant levels. The greatest changes occurring in the lake ecosystem, i.e., those related to changes in climate and nutrient inputs, will be investigated through the detailed examination of the dated sediment cores collected in 2009. The focus of these analyses will be on assessing contaminant trends, changes in lake productivity, and the contribution of the littoral to offshore communities.



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# Spatial patterns of contaminants in arctic seabirds

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## **Abstract**

As part of an International Polar Year study on dietary change in marine birds, adult thick-billed murrelets and northern fulmars were collected from a number of locations throughout the Canadian Arctic during 2007 and 2008. We took advantage of this opportunity to carry out a spatial analysis of contaminants for two species of seabirds. Preliminary results suggest that thick-billed murrelets breeding at the higher latitude Arctic colonies have higher mercury concentrations than those breeding at low Arctic colonies in northern Hudson Bay and Hudson Strait. There were also significant differences found for mercury in northern fulmars between the two colonies sampled in the high Arctic and off eastern Baffin Island.

## **Key Message**

- Preliminary results from the spatial analysis of mercury in livers of adult thick-billed murrelets from five colonies across the eastern Canadian Arctic suggest a latitudinal difference with birds breeding at the higher latitude Arctic colonies having higher mercury concentrations than those breeding at low Arctic colonies in northern Hudson Bay and Hudson Strait.

## **Résumé**

Lors d'une étude réalisée dans le cadre de l'Année polaire internationale sur le changement de régime alimentaire des oiseaux marins, des guillemots de Brünnich adultes et des fulmars boréaux ont été recueillis en plusieurs endroits de l'Arctique canadien en 2007 et en 2008. Nous avons profité de cette occasion pour effectuer une analyse spatiale des contaminants de deux espèces d'oiseaux de mer. Les résultats préliminaires donnent à penser que les guillemots de Brünnich qui se reproduisent dans les colonies arctiques à plus hautes latitudes ont des concentrations de mercure plus élevées que ceux qui se reproduisent dans les colonies arctiques à plus basses latitudes du Nord de la baie d'Hudson et du détroit d'Hudson. D'importantes différences ont aussi été constatées dans les concentrations de mercure chez les fulmars boréaux des deux colonies étudiées dans l'extrême arctique et au large de l'Est de l'île de Baffin.

## **Message clé**

- Les résultats préliminaires de l'analyse spatiale du mercure dans le foie des guillemots de Brünnich adultes effectuée dans cinq colonies de l'Est de l'Arctique canadien donnent à penser qu'il existe une différence latitudinale, les oiseaux se reproduisant dans les colonies à plus hautes latitudes ayant des concentrations de mercure

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plus élevées que ceux qui se reproduisent dans les colonies de l'Arctique à plus basses latitudes du Nord de la baie d'Hudson et du détroit d'Hudson.

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## Objectives

Adult birds were collected as part of a dietary study funded under the International Polar Year. We proposed to analyze livers from those birds to: (i) update earlier spatial information on contaminants in thick-billed murres, (ii) provide spatial information for northern fulmars, and (iii) generate spatial information for newer contaminants such as brominated flame retardants and perfluorinated compounds in seabirds.

## Introduction

One of the objectives of the core monitoring component of the NCP Blueprint for Environmental Trends Related to Human Health and International Action is to generate information on spatial trends of contaminants and to broaden the range of species for which we have information. The last spatial survey of contaminants carried out for arctic seabirds was in 1993 (Braune et al. 2002) and included only the legacy persistent organic pollutants (POPs) and total mercury (Hg). The results of that survey showed that concentrations of total Hg and many of the legacy POPs were significantly different in birds from Prince Leopold Island compared with other Canadian arctic colonies. As part of an International Polar Year (IPY) project to assess changes that have occurred in arctic marine food webs, adult birds were collected from a number of locations throughout the Canadian Arctic during 2007 and 2008 to compare dietary data from a previous survey of marine bird diets throughout Nunavut, made during the 1970s and 1980s, with current dietary information. Those birds are also being analyzed for stable isotope ratios of carbon and nitrogen in order to generate further information related to diet. The study focussed on colonies of thick-billed murres but also included some work on northern fulmars. Using the samples collected for the IPY study, we proposed to carry out a spatial survey of contaminants in 2008, including newer contaminants such as BFRs and PFCs,

in seabirds. These collections provide a unique opportunity to: (i) update earlier spatial information on contaminants in thick-billed murres, (ii) provide spatial information for northern fulmars which were not included in the 1993 survey, and (iii) generate spatial information for newer contaminants such as BFRs and PFCs in seabirds.

## Activities in 2008/2009

### *Sample collection/analysis:*

As part of an IPY study on dietary change in marine birds being carried out by Dr. Tony Gaston of Environment Canada, adult thick-billed murres and northern fulmars were collected from a number of locations throughout the Canadian Arctic during 2007 and 2008 (Figure 1). The birds were dissected with the help of students from Arctic College (Iqaluit). The birds had their gastro-intestinal tracts removed for diet analysis, and muscle samples have been sent for stable isotope (N, C) analysis. Livers from 10 adult murres (5 males, 5 females) from each of the following four locations are being analyzed for the legacy POPs, BFRs, PFCs, and total Hg: Prince Leopold Island, Digges Island, Akpatok Island, the Minarets, plus 5 murres (females) from Coats Island. Additionally, 10 adult fulmars (5 males, 5 females) are being analyzed from Prince Leopold Island, and from the Minarets/Cape Searle (~30 km from the Minarets).

### *Analytical methods:*

Analyses of the legacy POPs, PBDEs, HBCD, PFCs, and total mercury (Hg) are carried out at the National Wildlife Research Centre (NWRC) laboratories at Carleton University in Ottawa, Ontario. The legacy POPs are analyzed by gas chromatography using a mass selective detector (GC/MSD) according to NWRC Method No. MET-CHEM-OC-06B. Analyses of the 14 standard PBDE congeners and total- $\alpha$ -HBCD are carried out using GC-low resolution MS also according to NWRC Method No. MET-CHEM-OC-06B. PFCs are analyzed using HPLC/MS/MS



Figure 1. IPY sampling locations for adult seabirds in 2007 and 2008.

in negative electrospray mode (ESI<sup>-</sup>) according to NWRC Method No. MET-WTD-PFC-01. PFCs analyzed include 10 PFCAs (including PFOA), 4 PFSAAs (including PFOS), 3 FTUCAs, PFOSA and 3 FtOHs. Total Hg is analyzed using an Advanced Mercury Analyzer (AMA-254) equipped with an ASS-254 autosampler for solid samples according to NWRC Method No. MET-CHEM-AA-03G. The method employs direct combustion of the sample in an oxygen-rich atmosphere. Quality assurance/quality control (QA/QC) for all analyses is monitored by Laboratory Services at NWRC, Ottawa, Ontario, which is an accredited laboratory through the CAEL-SCC and has participated in the NCP's QA/QC Program. All samples will be archived in the NWRC Specimen Bank.

## Results

Only results for the total Hg analyses have been reported to date. There were no significant differences ( $p > 0.05$ ) in Hg concentrations found

between male and female thick-billed murres from each of the four colonies from which both sexes were sampled. Likewise, there were no significant differences found between male and female northern fulmars from each of the two colonies sampled. Preliminary results from the spatial analysis of Hg in livers of adult thick-billed murres from the five colonies showed that both males and females from Prince Leopold Island and the Minarets had significantly higher Hg concentrations (ANOVA - males:  $n=20$ ,  $F_{(3,16)}=47.988$ ,  $p<0.00001$ ; females:  $n=25$ ,  $F_{(4,20)}=13.787$ ,  $p=0.000015$ ) than birds from the three Hudson Strait/Hudson Bay colonies (Coats Island, Digges Island, Akpatok Island) (Figure 2). Although Hg in livers of male and female northern fulmars were not significantly different between the Minarets and Prince Leopold Island when sexes were analyzed separately ( $t$ -tests – males:  $n=10$ ,  $t_{(8)}=1.99$ ,  $p=0.082$ ; females:  $n=10$ ,  $t_{(8)}=2.25$ ,  $p=0.055$ ), fulmars from Prince Leopold Island did have

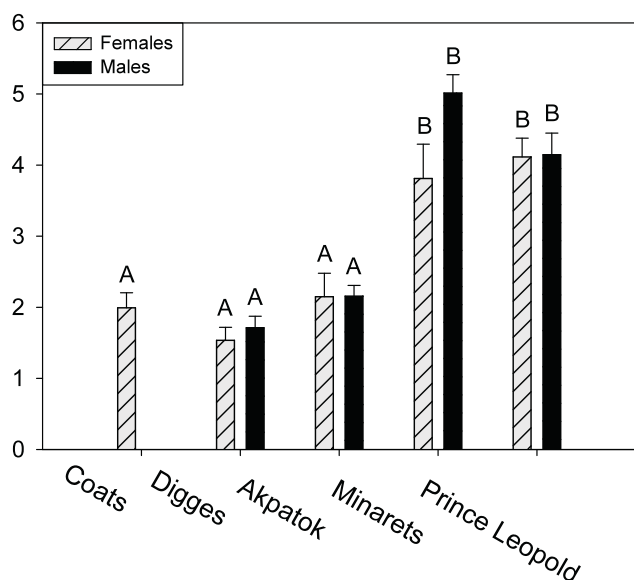


Figure 2. Mean hepatic concentrations ( $\pm$  standard error) of total Hg ( $\mu\text{g g}^{-1}$  dry wt) in thick-billed murres collected from five colonies across the eastern Canadian Arctic, 2007-08. Different letters indicate statistically different Hg concentrations among colonies and sexes.

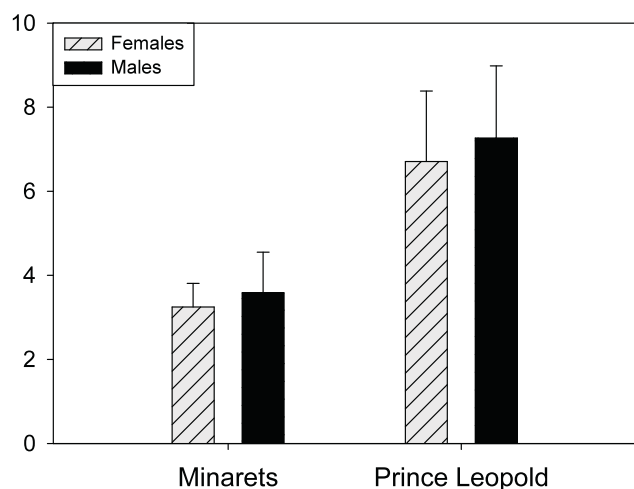


Figure 3. Mean hepatic concentrations ( $\pm$  standard error) of total Hg ( $\mu\text{g g}^{-1}$  dry wt) in northern fulmars collected from two colonies in the eastern Canadian Arctic, 2007-08.

significantly higher Hg concentrations ( $n=20$ ,  $t_{(18)}=3.12$ ,  $p=0.0059$ ) than the birds from the Minarets when the sexes were analyzed together (Figure 3).

## Discussion and Conclusions

The last spatial survey of contaminants in Canadian arctic seabirds was carried out in 1993 and included only the legacy POPs and total Hg (see Braune et al. 2002). That survey included eggs of thick-billed murres from four arctic locations (Prince Leopold Island & Coburg Island in the high Arctic, Digges Island & Coats Island in northern Hudson Bay). The results of that survey showed that concentrations of total Hg and all of the major groups of the legacy POPs except  $\Sigma\text{HCH}$  were significantly different in the murres from Prince Leopold Island compared with other arctic colonies. There were no significant differences found for the major groups of legacy POPs nor for total Hg in eggs from the two low arctic colonies at Coats Island and Digges Island. It is interesting to note that, although significant differences were found between the two colonies at Prince Leopold Island and Coats Island for total Hg and all of the major legacy POPs groups except for HCHs in 1993, by 1998, significant differences between those two colonies remained only for  $\Sigma\text{PCB}$ ,  $\Sigma\text{DDT}$ , dieldrin and total Hg (Braune et al. 2002). Northern fulmars were not included in the 1993 survey.

Mercury concentrations in marine biota from the Canadian Arctic are substantially higher than elsewhere in the circumpolar Arctic, and increases in Hg concentrations have been documented over the past few decades for a number of other Canadian Arctic species (Braune et al. 2005). Preliminary results from the spatial analysis of Hg in livers of adult thick-billed murres from five colonies located throughout the eastern Canadian Arctic in 2007-08 suggest a latitudinal difference with birds breeding at the higher latitude colonies (Prince Leopold Island, the Minarets) having higher Hg concentrations than those breeding at low Arctic colonies (Coats Island, Digges Island, Akpatok Island). Similar latitudinal differences have been observed by Dietz et al. (1996) for Greenland species and Braune et al. (2002) in the Canadian Arctic. As was found in the 1993 survey, murres from Prince Leopold Island still have higher Hg levels than those from the low Arctic colonies



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as do the murrelets from the Minarets off eastern Baffin Island. Stable isotope data will, hopefully, shed some light on the role of diet in determining the inter-colony differences in Hg concentrations. The knowledge that Hg levels in seabirds are higher at the Prince Leopold Island and Minarets colonies, coupled with the fact that concentrations at Prince Leopold Island appear to be steadily increasing (see Braune 2009, this report), underscore the need to continue monitoring of these colonies and to evaluate the Hg levels found in the context of marine ecosystem health.

## Expected Project Completion Date

December 31, 2009

## Acknowledgements

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# Effect of climate change on diet and contaminant exposure in seabirds breeding in northern Hudson Bay

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## Abstract

Thick-billed murres in northern Hudson Bay have been monitored for contaminants since the early 1990s. Dietary data for these birds from the 1980s and 1990s suggest that there has been a change in diet over that time period. As part of an International Polar Year (IPY) study, fish samples were collected in 2007 and 2008 from adult murres bringing food to their chicks on Coats Island in northern Hudson Bay. It was proposed that these samples be analyzed for total mercury, legacy POPs as well as brominated flame retardants (BFRs) and perfluorinated compounds (PFCs) to determine if exposure of thick-billed murres to contaminants through their diet on the breeding grounds has changed with a change in diet.

## Key Messages

- The shift in the diet from arctic cod and benthic fish species to capelin and sandlance is reflected in the decreased  $\delta^{15}\text{N}$  values found in murre eggs at the Coats Island thick-billed murre colony during 1998-2007.

## Résumé

On surveille les contaminants dans les guillemots de Brünnich du Nord de la baie d'Hudson depuis le début des années 1990. Les données alimentaires sur ces oiseaux recueillies dans les années 1980 et 1990 donnent à penser qu'il y a eu un changement de régime au cours de cette période. Lors d'une étude réalisée dans le cadre de l'Année polaire internationale (API), des échantillons de poisson ont été prélevés en 2007 et 2008 chez les guillemots adultes rapportant de la nourriture à leurs petits, sur l'île Coats, au nord de la baie d'Hudson. On a proposé d'analyser le mercure total, les POP résiduels, les ignifugeants bromés et les composés de perfluorocarbène que contenaient ces échantillons, afin de déterminer si l'exposition des guillemots de Brünnich aux contaminants du fait de leur régime alimentaire sur les sites de reproduction avait changé en raison d'un changement de régime alimentaire.

## Messages clés

- Le changement de régime alimentaire, de la morue polaire et des espèces de poissons benthiques au capelan et au lançon, se reflète dans la baisse des valeurs de  $\delta^{15}\text{N}$  trouvées dans les œufs de la colonie de guillemots de Brünnich de l'île Coats entre 1998 et 2007.

- The arctic cod fed to thick-billed murre chicks occupy a higher trophic position and have higher concentrations of total Hg in them than either the capelin or sandlance.
- The change in diet may be influencing the contaminant trends reported for thick-billed murres at the Coats Island colony.
- La morue polaire qui était donnée aux petits guillemots de Brünnich se trouve en haut de la pyramide alimentaire et contient des concentrations de Hg total plus élevées que le capelan ou le lançon.
- Le changement de régime alimentaire peut influencer les tendances en matière de contaminants observées chez les guillemots de Brünnich de la colonie de l'île Coats.

## Objectives

It is proposed that fish samples collected from Coats Island in 2007 and 2008 be analyzed for total Hg, organochlorines (OCs), PCBs, BFRs and PFCs to determine if exposure of thick-billed murres to contaminants through their diet on the breeding grounds has changed with a change in diet.

## Introduction

Thick-billed murres (*Uria lomvia*) breeding on Prince Leopold Island on Lancaster Sound in the high Arctic have been monitored for contaminants since 1975 (Braune 2007) and those breeding on Coats Island in northern Hudson Bay have been monitored since 1993 (Braune et al. 2002). Over the period 1975 to 2007,  $\delta^{15}\text{N}$  values, as an index of trophic position, have not shown a consistent directional change in eggs of thick-billed murres from Prince Leopold Island, whereas  $\delta^{15}\text{N}$  values in murre eggs from Coats Island from 1998-2007 are consistently lower than 1993 values (Figure 1). At Prince Leopold Island in the high Arctic, there has been no trend reported in summer ice cover (Davidson et al. 2008) whereas warming ocean conditions and longer ice-free periods have been documented for Hudson Bay (Gaston et al. 2009, Davidson et al. 2008). Dietary studies show that there has also been a shift in the diet of thick-billed murres in northern Hudson Bay from arctic cod and benthic fish species to capelin and sandlance over the period 1980-2002 (Gaston et al. 2003). As part of an International Polar Year (IPY) study on dietary change in marine birds, fish samples were collected in 2007 and 2008 from adult birds bringing food to their chicks on Coats Island. We proposed to analyze those samples for total Hg, legacy POPs (e.g. OCs, PCBs) as well as brominated

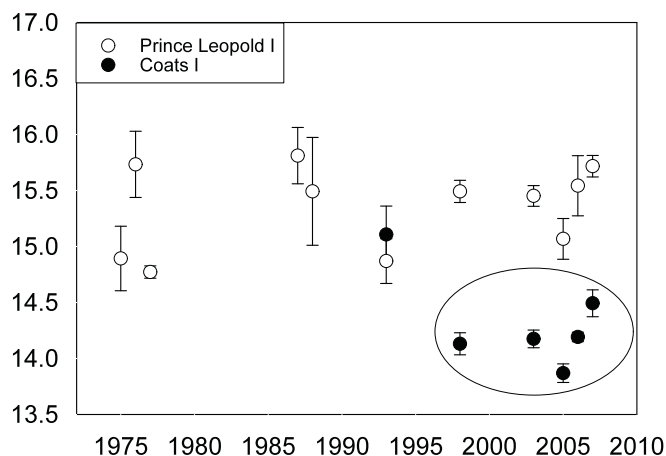


Figure 1. Change in  $\delta^{15}\text{N}$  (as an index of trophic position) in eggs of thick-billed murres from Prince Leopold Island in Lancaster Sound (1975-2007) and Coats Island in northern Hudson Bay (1993-2007).

flame retardants (BFRs) and perfluorinated compounds (PFCs) to determine if the different fish species have different contaminant profiles which would affect the exposure of the birds to contaminants through their diet on the breeding grounds.

## Activities in 2008/2009

### Sample collection/analysis:

Whole fish brought back by adult thick-billed murres to their chicks on Coats Island were sampled in 2007 and 2008. Previous samples were also available from 2004. The samples from 2004 and 2007 were analyzed for stable isotopes of C and N at the University of Winnipeg (Dan Bailey's lab) during the course of other studies. Fish samples available from 2004 and 2007 included arctic shanny (*Stichaeus punctatus*), daubed shanny

(*Leptoclinus maculatus*), banded gunnel (*Pholis fasciata*), fourline snakeblenny (*Eumesogrammus praecisus*), Atlantic poacher (*Leptagonus decagonus*), scuplin sp. (e.g. *Myoxocephalus scorpioides*, *Triglops pingelii*, *T. murrayi*), eelpout (*Gymnelus viridis*), capelin (*Mallotus villosus*), sandlance (*Ammodytes hexaptera*) and arctic cod (*Boreogadus saida*). Only sagittal sections remained from those fish which had already been analyzed for stable isotopes. Whole capelin and amphipods were available for contaminant analysis from 2008. Sagittal sections were pooled, as necessary, to make up the required mass for contaminant analysis. Whole fish were analyzed individually. All samples (individuals or pools) were analyzed for total Hg. However, due to limited sample mass, not all samples were analyzed for OCs, PCBs, BFRs and PFCs.

#### Analytical methods:

Analyses of the legacy POPs (OCs, PCBs), BFRs (PBDEs, HBCD), PFCs and total Hg are being carried out at the National Wildlife Research Centre (NWRC) laboratories at Carleton University in Ottawa, Ontario. The legacy POPs are analyzed by gas chromatography using a mass selective detector (GC/MSD) according to NWRC Method No. MET-CHEM-OC-06B. Analyses of the 14 standard PBDE congeners and total- $\alpha$ -HBCD are carried out using GC-low resolution MS also according to NWRC Method No. MET-CHEM-OC-06B. PFCs are analyzed using HPLC/MS/MS in negative electrospray mode (ESI) according to NWRC Method No. MET-WTD-PFC-01. Total Hg is analyzed using an Advanced Mercury Analyzer (AMA-254) equipped with an ASS-254 autosampler for solid samples according to NWRC Method No. MET-CHEM-AA-03G. The method employs direct combustion of the sample in an oxygen-rich atmosphere. Quality assurance/quality control (QA/QC) is monitored by NWRC Laboratory Services which is an accredited laboratory through the CAEAL and has participated in the NCP's QA/QC Program. All remaining samples will be archived in the Environment Canada Specimen Bank housed at NWRC in Ottawa.

## Results

To date, we have received only the Hg results. However, since we have not yet received the stable isotope data for the 2008 samples, the following

analyses are based only on the 2004 and 2007 data. The arctic cod fed to thick-billed murre chicks occupy a higher trophic position and have higher concentrations of total Hg in them than either the capelin or sandlance (Figure 2). In fact, Hg levels in the cod were about 74% higher than in the capelin.

## Discussion

The trophic positions of thick-billed murres, as indicated by  $\delta^{15}\text{N}$  values in eggs, were similar at Prince Leopold Island and Coats Island in 1993 (Figure 1). Arctic cod is the main prey of thick-billed murres at Prince Leopold Island in the high Arctic (Gaston and Bradstreet 1993, Davidson et al. 2008) and, until the mid-1990s, was the most common prey item found in the diet of nestling murres throughout the Canadian Arctic (Gaston and Jones 1998). However, during the period 1980-2002, Gaston et al. (2003) documented a shift in the diet of thick-billed murres in northern Hudson Bay from arctic cod and benthic fish species (e.g. sculpins, shannies, blennies, gunnels, eelpouts, Atlantic poachers) to capelin and sandlance, a trend which has continued into more recent years (Gaston et al., unpublished data). Given that arctic cod and a number of the benthic fish species occupy a higher trophic position than either capelin or sandlance (Figure 2), this dietary shift could explain the decrease we observed in

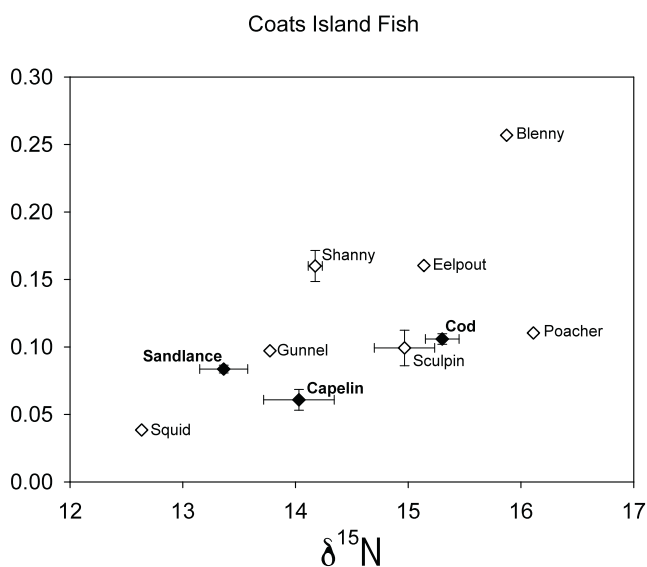


Figure 2. Trophic position ( $\delta^{15}\text{N}$ ) vs total Hg concentration ( $\mu\text{g g}^{-1}$  dry wt) in some prey items of thick-billed murres at Coats Island.

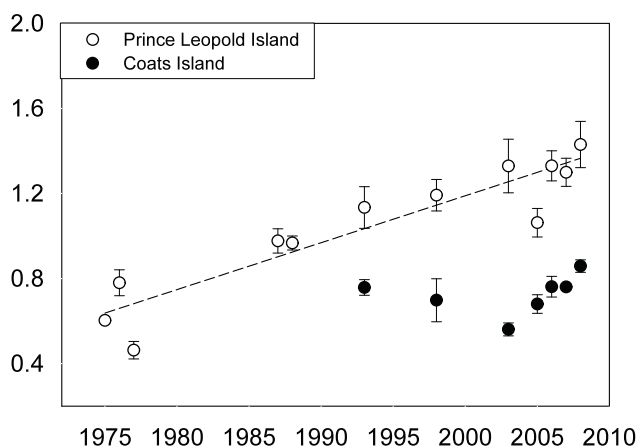


Figure 3. Concentrations ( $\mu\text{g g}^{-1}$  dry wt) of total Hg in eggs of thick-billed murres from Prince Leopold Island (1975-2008) vs Coats Island (1993-2008).

trophic position as reflected by  $\delta^{15}\text{N}$  values in the murre eggs at the Coats Island colony during 1998-2007 (Figure 1). Total Hg levels in thick-billed murre eggs have been increasing at Prince Leopold Island whereas there is no clear Hg trend at the Coats Island murre colony (Figure 3). We hypothesize that the lack of a trend in Hg concentrations over time at the Coats Island colony may not reflect environmental Hg trends, rather it may be due to a shift in prey species taken resulting in a lower exposure to Hg via the diet over time. However, a change in dietary composition could also lead to a change in the nutritional/energetic value of the diet which would, in turn, require a change in the mass consumption of certain prey species in order to maintain a minimum necessary energetic intake (see Harris et al. 2008, Hebert et al. 2008, Österblom et al. 2008). Using dietary composition data in conjunction with data on energetic content of the prey species (see Elliott and Gaston, 2008), the next step will be to calculate expected changes in contaminant exposure using existing bioenergetics information for these birds.

## Expected Project Completion Date

March 31, 2010

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# Interspecies sensitivity of arctic marine birds to methylmercury exposure: a pilot study

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## **Abstract**

Total mercury (Hg) has been increasing in marine birds and mammals in some regions of the Canadian Arctic and in West Greenland over the past several decades. There is also evidence to suggest that current Hg exposures may pose a health risk to some people and animals in the Arctic. The most bioavailable and toxic form of Hg is methylmercury (MeHg) and nearly 100% of the Hg transferred by breeding female birds to their eggs is MeHg. Given that reproduction is one of the most sensitive endpoints of MeHg toxicity, we propose to use protocols developed by the United States Geological Survey (USGS) to bring eggs of arctic seabirds into the laboratory where they will be dosed with graded concentrations of methylmercury chloride to determine the relative sensitivity of the developing embryo to MeHg. However, before proceeding with this type of research, we conducted a pilot study to determine the feasibility of transporting eggs from an arctic seabird colony to the National Wildlife Research Centre in Ottawa and successfully incubating the eggs artificially. We also wanted to determine if neurochemical changes, which have been shown to occur in association with increasing Hg

## **Résumé**

Le mercure (Hg) total augmente depuis quelques dizaines d'années chez les oiseaux et mammifères marins de certaines régions de l'Arctique canadien et de l'Ouest du Groenland. Des observations donnent aussi à penser que les expositions actuelles au mercure peuvent présenter un risque pour la santé de certains habitants et animaux de l'Arctique. La forme la plus biodisponible et toxique du Hg est le méthylmercure (MeHg) et près de 100 % du Hg que les oiseaux femelles qui se reproduisent transmettent à leurs œufs est le MeHg. Étant donné que la reproduction est l'une des extrémités les plus sensibles de la toxicité du MeHg, nous proposons d'utiliser des protocoles établis par le United States Geological Survey (USGS) pour rapporter des œufs d'oiseaux marins arctiques au laboratoire, où on leur administrera des concentrations progressives de chlorure de méthylmercure pour déterminer la sensibilité relative de l'embryon en cours de développement au MeHg. Toutefois, avant d'effectuer ce type de recherche, nous avons réalisé une étude pilote afin de déterminer s'il était possible de transporter des œufs depuis une colonie d'oiseaux marins arctiques au Centre national de la recherche faunique d'Ottawa et de les incubier avec succès dans



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concentrations in brains of adult birds, could be detected and measured in the brain tissue of developing avian embryos.

## Key Messages

- Seabird eggs collected from Canadian Arctic colonies can be successfully transported to a southern location and artificially incubated.
- Preliminary results show that neurotransmitter receptor densities in brains of developing avian embryos can be detected with no tissue-specific issues.

des conditions artificielles. Nous voulions aussi déterminer s'il était possible de détecter et de mesurer, dans les tissus cérébraux des embryons d'oiseaux en cours de développement, les changements neurochimiques qui surviennent avec l'augmentation des concentrations de Hg dans le cerveau des oiseaux adultes.

## Messages clés

- Il est possible de transporter plus au sud et d'incuber dans des conditions artificielles des œufs d'oiseaux marins recueillis dans les colonies de l'Arctique canadien.
- Les résultats préliminaires indiquent que les densités des récepteurs des neurotransmetteurs dans le cerveau des embryons d'oiseaux en cours de développement peuvent être détectées et ne présentent pas de problème propre aux tissus.

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## Objectives

### Long-term

- To determine the relative sensitivities of methylmercury (MeHg) exposure for arctic marine bird species based on the most sensitive life stage, embryonic development.
- Based on the MeHg sensitivity and existing Hg temporal trend data (i.e. rates of Hg increase/change in eggs), predict when environmental Hg exposure levels may reach critical thresholds for reproductive impairment in the species tested.
- To determine the effects of MeHg exposure on the neuro-signaling pathways of the developing avian brain using changes in neurochemical parameters (e.g. neuroreceptor concentrations) as a biomarker of Hg exposure; and relate neurochemical changes to decreased hatchability following MeHg exposure *in ovo*.

### 2008-09

- To ascertain the feasibility of transporting eggs from an arctic seabird colony to the National Wildlife Research Centre in Ottawa and successfully incubating the eggs artificially to the pipping stage (starring of eggshell indicating beginning of hatch).

- To determine if receptors for the neurotransmitters acetylcholine (muscarinic [mACh]) and glutamate (N-methyl-D-aspartic acid [NMDA]) could be detected and confidently measured in developing avian brain tissue.

## Introduction

Elemental mercury (Hg) is highly volatile, and gaseous Hg<sup>0</sup> partitions readily into the atmosphere where it can undergo long-range atmospheric transport. Due to a variety of factors (see Macdonald et al. 2005), polar regions are global sinks for Hg. Mercury biomagnifies up the food chain (Atwell et al. 1998; Campbell et al. 2005) making those species feeding at high trophic positions more vulnerable to Hg exposure via their diet. Canadian arctic marine bird species which feed at higher trophic levels include the glaucous gull (*Larus hyperboreus*), ivory gull (*Pagophila eburnea*), northern fulmar (*Fulmarus glacialis*) and thick-billed murre (*Uria lomvia*) (Campbell et al. 2005).

Although arctic biota have been exposed to a range of organic and inorganic contaminants, Muir et al. (1999) concluded that, up until 1997, there had been relatively little study of the biological effects of these chemicals. Unlike the situation in the

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Baltic Sea or the Great Lakes, there is still currently little evidence to support or refute arguments that chemical contaminants are present in sufficient quantities to affect reproduction or survival in Canadian Arctic marine biota (Fisk et al. 2005). Total Hg has been increasing in marine birds and mammals in some regions of the Canadian Arctic and in West Greenland over the past several decades (Braune et al. 2005a,b). In particular, retrospective analyses of Hg in archived seabird eggs from the Canadian Arctic have shown steady increases in several species of marine birds breeding in the Arctic (Braune 2007). As well, the ivory gull, a High Arctic marine bird which has experienced a dramatic population decline in the Canadian Arctic in recent decades, has been found to contain among the highest concentrations of Hg measured in arctic seabird eggs (Braune et al. 2006). There is also evidence to suggest that current Hg exposures may pose a health risk to some people and animals in the Arctic (Fisk et al. 2005).

The most bioavailable and toxic form of Hg is methylmercury (MeHg) and nearly 100% of the Hg transferred by breeding female birds to their eggs is MeHg (Wiener et al. 2003). Mercury is an extremely potent embryo toxicant, and dietary Hg is rapidly transferred to avian eggs on a dose-dependent basis, making reproduction one of the most sensitive endpoints of Hg toxicity (Wolfe et al. 1998). Where no embryotoxic data exist for wild species, embryotoxic thresholds of Hg based on captive breeding studies using species such as mallards (*Anas platyrhynchos*) and ring-necked pheasants (*Phasianus colchicus*) are often used generically for a wide range of avian species (Heinz et al. 2009). These extrapolations from laboratory results for mallards and pheasants to wild species have been used even though there is no reason to believe that the embryos of all birds are equally sensitive to the harmful effects of MeHg (Heinz et al. 2009). Given the great expense and logistical difficulties inherent in conducting controlled laboratory breeding studies with wild birds, Heinz et al. (2006) developed a protocol by which the eggs of wild birds could be brought into the laboratory and injected with graded concentrations of methylmercury chloride to determine threshold levels of harmful effects. Although they have now generated MeHg dose-response relationships for

26 different avian species (Heinz et al. 2009), no work has been done on alcids (e.g. murre) or procellariids (e.g. fulmars).

A number of studies have shown that neurochemical parameters (such as neuroreceptor concentrations) are significantly affected by low-level dietary exposure to MeHg in adult birds and mammals; thus there is a potential to use specific neurochemical changes as biomarkers of Hg exposure and effects in wildlife (Basu et al., 2007; Basu et al., 2006; Scheuhammer et al., 2008). However, the effects of *in ovo* exposure to MeHg on developing avian neurochemical pathways are not currently known. Because low doses of MeHg are generally more toxic in developing embryos than in adults, it is of considerable interest to determine the effects of MeHg exposure on the neuro-signaling pathways of the developing brain, and to relate these changes to more commonly used endpoints of toxicity, such as decreased hatchability.

## Activities in 2008/2009

The project leader and two colleagues met with Gary Heinz of the USGS and his colleagues at the Patuxent Wildlife Research Center in Maryland to learn the methodological protocols for MeHg dosing of eggs and artificial incubation. Several new contact incubators were purchased for the pilot study using non-NCP funds.

The pilot study, which was to determine the feasibility of transporting eggs from an arctic seabird colony to the National Wildlife Research Centre (NWRC) in Ottawa and successfully incubating the eggs artificially to the pipping stage (starring of eggshell indicating beginning of hatch), was moderately successful. A total of 15 fresh northern fulmar (*Fulmarus glacialis*) eggs were collected from Prince Leopold Island (74°02'N, 90°05'W) in early June 2008 for the study. In the wild, northern fulmar eggs typically have an incubation period of 47-49 days (Hatch and Nettleship 1998). The eggs were stored in the Polar Continental Shelf Project (PCSP) warehouse at Resolute Bay at an ambient temperature of ~13°C until they could be flown to Ottawa four days later. Storage of unincubated eggs for several days at cool temperatures prior to placement in an incubator is an accepted practice in the poultry industry (Fasenko 2007). Upon arrival at NWRC in Ottawa, one cracked egg was removed and the remaining

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14 eggs were placed in a Brinsea Z6 contact incubator after being weighed and candled to determine stage of development. Eggs were weighed and candled once a week during artificial incubation.

Although a few recent studies have reported neurochemical changes in association with increasing Hg concentrations in brains of adult birds (e.g. Scheuhammer et al. 2008), there are no reports of neurotransmitter receptor densities in brains of developing avian embryos. The ability to measure neurotransmitter receptors in the embryonic brain is a prerequisite for our proposed research for this particular endpoint. Thus, preliminary studies were conducted to determine if receptors for the neurotransmitters acetylcholine (muscarinic [mACh]) and glutamate (N-methyl-D-aspartic acid [NMDA]) could be detected and confidently measured in developing avian brain tissue.

## Results

Two of the 14 northern fulmars eggs transported from Prince Leopold Island back to Ottawa showed signs of early development upon arrival at NWRC. However, the other 12 eggs showed no signs of development suggesting that they had not been subject to any incubation at the colony. About 27 days into incubation, 10 of the 14 eggs were removed from the incubator because they were showing no signs of development. The remaining four eggs continued to develop until they were terminated at pip (starring of eggshell indicating beginning of hatch) or within days of pipping. Embryos were decapitated and brain samples for Hg and neuroreceptor density analyses were taken from the four embryos.

Analyses of the four fulmar embryo brains sampled for neurotransmitter receptors are currently underway. However, in another study conducted at NWRC using herring gull (*Larus argentatus*) eggs from the Great Lakes, brain tissue was dissected from pipping herring gull chicks following artificial incubation of eggs collected from the wild, and both NMDA and mACh receptors were detected with no tissue-specific issues. Preliminary results show NMDA receptor levels that are comparable to those found in adults, whereas mACh receptor levels appear to be on average higher than in adults.

## Discussion and Conclusions

Although the success rate for the pilot study using northern fulmar eggs was only 29%, it seems that the hatching success (# eggs hatched / # eggs laid) for northern fulmars at the colony on Prince Leopold Island is also naturally relatively low, averaging only 57% (Mallory et al. 2009). Therefore, given the relatively small sample size for the pilot study, in conjunction with a naturally low hatching success rate in the wild, we do not believe that the limited success of the pilot study was necessarily due to issues related to transport and incubation conditions. The fact that the 10 eggs, which were removed about half way through incubation, did not show any signs of development whereas the other four eggs developed, suggests that it was not the artificial incubation conditions which compromised development. Further, at the same time as the fulmar eggs were being collected for the pilot study, four glaucous gull eggs were also collected from a small colony north of Resolute Bay, held in storage at the PCSP warehouse, and transported to NWRC under the same conditions as the fulmar eggs. Those eggs were also artificially incubated, and three of the four eggs (75%) developed before being terminated within days of pipping. Therefore, we believe that seabird eggs can be successfully collected from Canadian arctic colonies, transported to NWRC, and artificially incubated to pipping.

The ability to measure neurotransmitter receptors in the embryonic brain is a prerequisite for our proposed research for this particular endpoint. Since there were no previous reports of neurotransmitter receptor densities in brains of developing avian embryos, preliminary studies were conducted to determine if receptors for the neurotransmitters acetylcholine (muscarinic [mACh]) and glutamate (N-methyl-D-aspartic acid [NMDA]) could be detected and confidently measured in developing avian brain tissue. These two neurotransmitter receptors play important roles in the regulation and control of reproductive hormones and, therefore, their perturbation may signify early impacts to reproductive potential. Preliminary results using herring gull eggs from the Great Lakes show that both NMDA and mACh receptors could be detected with no tissue-specific issues.

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## Expected Project Completion Date

March 31, 2012

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# PCB Effects in Black Guillemots in Saglek, Labrador

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## **Abstract**

Thirty-five seabird chicks (black guillemots) were collected in 2007 from different parts of Saglek Bay, Labrador. PCB levels in the livers of the guillemots have declined significantly since 1999, as a result of a PCB clean-up at Saglek. We assessed several measures of health and development in the guillemot chicks, to determine the current impacts of PCBs and the effectiveness of the PCB remediation. The lower PCB levels in 2007 were no longer associated with changes in sex hormones, liver EROD activity and vitamin A levels. However, elevated PCB levels were still associated with changes in thyroid hormone levels in plasma (T4) and brain neurotransmitter enzyme activity (ChE and MAO). The results of this study will enable us to derive toxicity thresholds for PCBs in an Arctic seabird, based on PCB levels in sediment, forage fish, or seabird eggs or tissues. These thresholds will be useful to assess the risks associated

## **Résumé**

Trente-cinq oisillons marins (guillemots à miroir) ont été recueillis en 2007 dans différentes parties de la baie Saglek, au Labrador. Les niveaux de BPC présents dans le foie des guillemots ont considérablement diminué depuis 1999 grâce aux efforts de nettoyage des BPC dans la région de Saglek. Nous avons évalué plusieurs mesures de la santé et du développement des jeunes guillemots afin de déterminer les impacts actuels des BPC et l'efficacité des mesures correctives pour réduire les BPC. Les niveaux inférieurs de BPC en 2007 ne sont plus associés à des changements des hormones sexuelles, de l'activité de l'EROD du foie et des niveaux de vitamine A. Cependant, des niveaux élevés de BPC sont toujours associés à des changements des niveaux des hormones thyroïdiennes dans le plasma (T4) et de l'activité des enzymes neurotransmetteurs du cerveau (ChE et MAO). Les résultats de cette étude nous permettront d'établir les seuils de toxicité des



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with marine PCB levels measured elsewhere in the North. The results will also be useful in assessing the success of the PCB remediation efforts at Saglek. This project has been carried out as a partnership with Environment Canada, the Environmental Sciences Group, the Nunatsiavut Government and Parks Canada. Findings will be communicated back to the local community in Nain in collaboration with the Nunatsiavut Government.

## Key Messages

- PCB concentrations in the livers of black guillemot chicks have dropped significantly from 1999 to 2007.
- PCB levels continue to be highest in guillemots nesting closest to Saglek beach.
- Many adverse effects associated with elevated PCB levels in the seabird chicks in 1999 have disappeared in 2007.
- However, elevated PCB levels in seabird chicks closest to the Saglek beach are still associated with lower than normal thyroid hormone levels (thyroxine) in the blood and elevated levels of two neurotransmitter enzymes in the brain (cholinesterase and monoamine oxidase).
- Using the black guillemots as an indicator of marine ecosystem health, our results indicate major improvements since 1999 but not yet a full return to normal conditions.

BPC pour les oiseaux marins arctiques fondés sur les niveaux de BPC contenus dans les sédiments, les poissons à fourrage, les œufs ou les tissus des oiseaux marins. Ces seuils seront utiles pour évaluer les risques liés aux niveaux de BPC dans la mer mesurés dans d'autres régions du Nord. Ces résultats seront également utiles pour évaluer le succès des mesures correctives contre les BPC mises en œuvre dans la région de Saglek. Ce projet a été effectué dans le cadre d'un partenariat avec Environnement Canada, le Groupe des sciences de l'environnement, le gouvernement du Nunatsiavut et Parcs Canada. Les conclusions de l'étude seront présentées à la collectivité locale à Nain, en collaboration avec le gouvernement du Nunatsiavut.

## Messages clés

- Les concentrations de BPC contenues dans le foie des jeunes guillemots à miroir ont considérablement diminué entre 1999 et 2007.
- Les niveaux de BPC demeurent les plus élevés chez les guillemots dont les nids sont établis à proximité de la plage de Saglek.
- De nombreux effets négatifs associés à des niveaux élevés de BPC constatés chez les oisillons en 1999 avaient disparu en 2007.
- Cependant, les niveaux élevés de BPC chez les oisillons à proximité de la plage de Saglek sont toujours associés à des niveaux d'hormones thyroïdiennes inférieurs à la normale (thyroxine) dans le sang et à des niveaux élevés de deux enzymes neurotransmetteurs dans le cerveau (cholinestérase et monoamine oxydase).
- Si nous utilisons les guillemots à miroir comme indicateur de la santé de l'écosystème marin, nos résultats font état d'améliorations considérables depuis 1999 mais non d'un retour complet à la normale.

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## Objectives

1. To assess the effectiveness of PCB remediation efforts at Saglek in reducing PCB levels in the marine foodweb and adverse effects in seabird chicks.
2. To establish PCB-exposure thresholds for sub-lethal adverse effects in an Arctic seabird

(black guillemot), which is used as a bio-indicator species for monitoring both:

- i) the long-term trends of contaminants in the Arctic, and
- ii) the ecological impacts of local sources of marine pollution.

## Introduction

The black guillemot is an Arctic seabird that feeds almost exclusively on benthic fish and invertebrates in inshore marine ecosystems. It has proven to be an excellent indicator of PCB biomagnification from contaminated marine sediments. It has also proven to be relatively sensitive to adverse impacts from PCB exposure. Our studies in 1999 at Saglek, Labrador found adverse impacts on the immune and endocrine systems, and on organ development in guillemot chicks in nests within 6 km of a point source of marine PCB pollution (ESG 2002; Kuzyk et al. 2003, 2005; Burgess (*unpublished data*). We returned in 2007 to repeat the guillemot study and assess any ecological improvements following the clean-up of PCB-contaminated soil at Saglek. Black guillemots are used in northern Canada and Europe to monitor contaminant trends in marine ecosystems (Fisk et al. 2003, 2005). The results of this project will provide toxicity thresholds for interpreting the toxicological significance of PCB levels observed in guillemot tissues.

## Activities in 2008/2009

Tissue samples were collected from 35 black guillemot chicks in 2007. PCB congener concentrations were measured in the guillemot livers in 2007. In 2008-09, tissue samples were sent for toxicological analysis of the following biomarkers:

- i) free and total T3 and thyroxine T4 (thyroid hormone) levels in plasma
- ii) estradiol and testosterone (sex hormones) levels in plasma
- iii) vitamin A (retinol & retinyl palmitate) levels in liver
- iv) EROD enzyme activity in liver
- v) neurotransmitter enzyme activity (AChE & MOA) in brain

The first four analyses were conducted at Environment Canada's National Wildlife Research Centre in Ottawa. The fifth set of analyses were carried out by Nil Basu at the University of Michigan.

## Results

### Liver PCB Levels:

In 2007, PCB analyses of 91 congeners were conducted at Axys Analytical on 12 livers from black

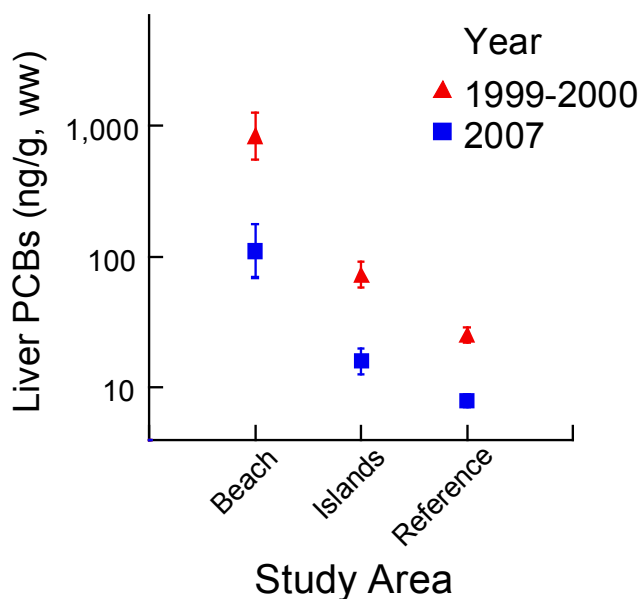


Figure 1. Geometric mean concentrations ( $\pm$  SE) of  $\Sigma$ PCB congeners ( $\text{ng g}^{-1}$ , wet wt.) in livers of juvenile black guillemots in three areas of Saglek Bay, Labrador in 1999-2000 and 2007.  $N = 31$  (1999-2000) and 35 (2007).

guillemot chicks from the highly-contaminated Beach group, 10 from the moderately-contaminated Islands group, and 13 from the least-contaminated Reference group.  $\Sigma$ PCB concentrations in the 35 livers ranged from 5  $\text{ng/g}$  (wet wt) in the Reference group to 1,143  $\text{ng/g}$  in the Beach group. Guillemot nestling livers collected from the Beach group had a geometric mean  $\Sigma$ PCB concentration 6.8 times greater than guillemot nestlings collected from the Islands group and 12.5 times greater than guillemot nestlings collected from the Reference group (Table 1); both differences were significant ( $p < 0.001$ ). The mean  $\Sigma$ PCB concentrations in the Islands and Reference group were not statistically different.

Comparisons of  $\Sigma$ PCB concentrations in guillemot livers between 1999-2000 and 2007 revealed significant declines over time (Figure 1).

### Thyroid hormones:

Total thyroxine (T4) hormone concentrations in the plasma of male guillemot chicks were negatively associated with liver  $\Sigma$ PCB levels ( $R^2 = 0.57$ ,  $p = 0.005$ ), but not in female chicks (Figure 2). The same pattern was observed in free T4 levels in plasma of male chicks ( $R^2 = 0.33$ ,  $p = 0.05$ ). No differences were found in total or free triiodothyronine (T3) levels in plasma.

**Table 1. Mean concentrations of  $\Sigma$ PCB congeners in liver and biomarkers of endocrine, liver and brain function in juvenile black guillemots from Saglek Bay, Labrador, 2007. See Liver PCB Results for explanation of Beach, Islands and Reference groups.**

Biomarker	Sex	N	Beach	Islands	Reference	Mean <sup>1</sup>
$\Sigma$ PCB Congeners in Liver (ng g <sup>-1</sup> , wet wt)	both	35	116 <i>A2</i>	17 <i>B</i>	9 <i>B</i>	G
Plasma Total T4 (ng ml <sup>-1</sup> )	male	12	9.2	12.8	17.8	A
	female	17	17.5	18.7	17.7	A
Plasma Free T4 (ng ml <sup>-1</sup> )	male	12	17.7	18.9	21.8	A
	female	17	26.8	27.7	23.5	A
Plasma Total T3 (ng ml <sup>-1</sup> )	both	29	2.6	2.6	2.6	A
Plasma Free T3 (ng ml <sup>-1</sup> )	both	29	3.6	4.2	3.5	A
Plasma Testosterone (ng ml <sup>-1</sup> )	both	29	0.122	0.098	0.073	A
Plasma Estradiol (ng ml <sup>-1</sup> )	both	29	nd <sup>3</sup>	nd	nd	–
Liver Retinol (ug g <sup>-1</sup> )	both	35	27	18	28	G
Liver Retinyl Palmitate (ug g <sup>-1</sup> )	both	35	354	222	357	G
Liver EROD Activity (pmol min <sup>-1</sup> mg protein <sup>-1</sup> )	both	32	10	12	9 <sup>4</sup>	A
Brain ChE Activity (RFU min <sup>-1</sup> ng protein <sup>-1</sup> )	both	35	2.8 <i>A</i>	2.9 <i>A</i>	1.3 <i>B</i>	G
Brain MAO Activity (RFU min <sup>-1</sup> ug protein <sup>-1</sup> )	both	35	0.88 <i>A</i>	0.93 <i>A</i>	0.69 <i>B</i>	A

<sup>1</sup> A = arithmetic mean, G = geometric mean

<sup>2</sup> different letters indicate significant differences among means ( $p < 0.05$ )

<sup>3</sup> nd = not detected, limit of detection for estradiol = 0.01 ng ml<sup>-1</sup>

<sup>4</sup> two outliers omitted

EROD = ethoxyresorufin *O*-deethylase; ChE = cholinesterase ; MAO = monoamine oxidase

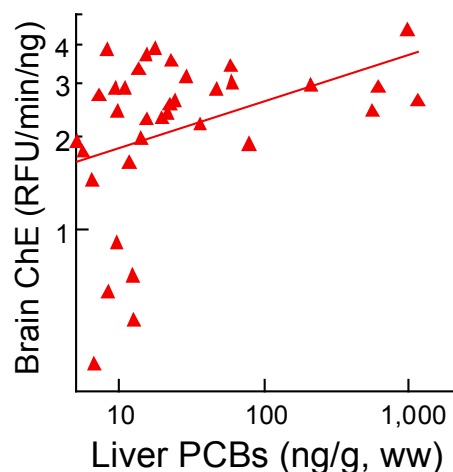


Figure 2. Total plasma thyroxine (T4) concentrations (ng ml<sup>-1</sup>) and liver  $\Sigma$ PCB congener concentrations (ng g<sup>-1</sup>, wet wt.) for 35 juvenile black guillemots in 2007.

#### **Sex hormones:**

While plasma testosterone levels increased with PCB exposure (Table 1), the trend was not significant ( $p > 0.05$ ). All estradiol concentrations in plasma were below the limit of detection of 0.01 ng ml<sup>-1</sup> in 2007.

#### **Vitamin A in Liver:**

There were no associations found between liver  $\Sigma$ PCB levels and liver retinol or retinyl palmitate concentrations in male, female or all chicks combined ( $p > 0.05$ ). There were no differences found among PCB exposure groups (Table 1).

#### **Ethoxyresorufin *O*-deethylase (EROD)**

##### **Activity in Liver:**

There was no association found between liver  $\Sigma$ PCB levels and EROD activity in liver ( $p > 0.05$ ). There were no differences found among PCB exposure groups (Table 1).

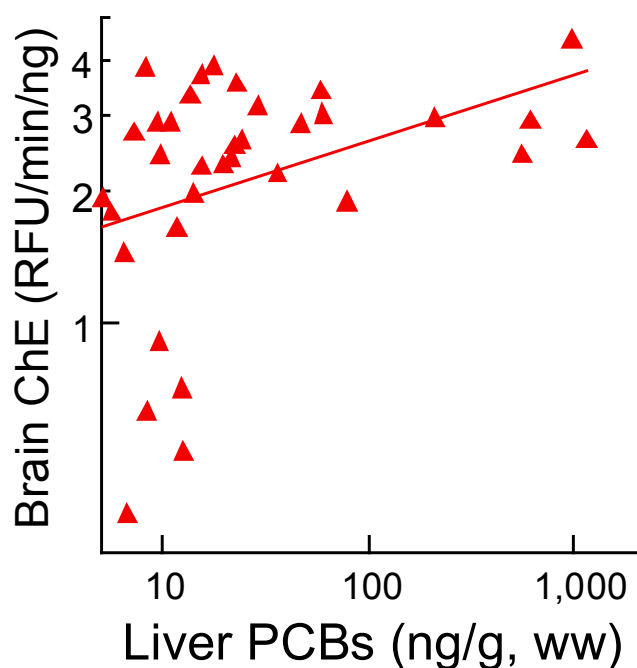


Figure 3. Brain cholinesterase (ChE) activity ( $\text{RFU min}^{-1} \text{ ng protein}^{-1}$ ) and liver  $\Sigma\text{PCB}$  congener concentrations ( $\text{ng g}^{-1}$ , wet wt.) for 35 juvenile black guillemots in 2007.

#### *Neurotransmitters in Brain:*

Both cholinesterase and monoamine oxidase enzyme activities were significantly elevated in the brains of Beach and Islands guillemots compared to the Reference birds ( $p < 0.05$ , Table 1, Figure 3). These endpoints were not assessed in the 1999 guillemot study.

## Discussion and Conclusions

Significant declines in PCB concentrations in the guillemot livers between 1999-2000 and 2007 indicate that the remediation of PCB-contaminated soil at Saglek has been effective in reducing PCB levels in the marine ecosystem. Future PCB levels in marine biota are expected to continue to decline.

Changes in the following endpoints were associated with elevated PCB exposure in black guillemot chicks in 1999 but no longer in 2007:

- plasma testosterone and estradiol concentrations increased,
- liver EROD activity increased, and

- retinol and retinyl palmitate concentrations decreased.

PCB concentrations in the guillemots in 2007 appear to have declined below the levels associated with these adverse effects.

However, three toxicological endpoints are associated with elevated PCB concentrations in the 2007 guillemot chicks near Saglek beach:

- plasma total and free thyroxine concentrations declined with elevated PCB levels, and
- brain cholinesterase and monoamine oxidase enzyme activities increased with elevated PCB levels in the birds.

Several other endpoints are currently being assessed and may provide further insights on the impacts of current PCB levels at Saglek on marine ecosystem health.

Using the black guillemots as an indicator of marine ecosystem health, our results indicate major improvements since 1999 but not yet a full return to normal conditions.

## Expected Project Completion Date

Our partners are working to complete the analysis of several more toxicological endpoints. Prof. Andrew Iwaniuk is currently assessing the impacts of PCB exposure on the brain anatomy of the black guillemot chicks. Dr. Laura Rogers is currently assessing the impacts of PCB exposure on the histopathology of the gonads and several other endocrine organs. The results of these analyses are expected by March 2010.

## Acknowledgements

We would like to thank the Director-General Environment of the Department of National Defence, ArcticNet and the Northern Contaminants Program for partial funding of this project. This work is a collaboration between Environment Canada, the Environmental Sciences Group, the Nunatsiavut Government and Parks Canada. We thank Marion Winters, Aimee McIntosh, Tim Pottle, Danielle Baikie, and Craig Burden for their assistance with the field work.

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# Influence of climate warming on scavenging of POPs by suspended particulate organic matter in lake waters, as recorded in sediments of Amituk Lake, Cornwallis Island

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## Abstract

Recent Arctic climate warming is known to have significantly increased the primary productivity of northern lakes, resulting in higher waterborne concentrations of algal-derived organic matter (OM), and higher fluxes of algal-derived carbon into sediments. This one-year project investigated whether these accelerating processes could have influenced the accumulation rates of legacy POPs in Amituk Lake sediments on Cornwallis Island, Nunavut. Results indicated that the sedimentary POPs profiles integrated two superimposed and sometimes opposing historical processes: atmospheric POPs deposition reflecting distant global emissions, and increased POPs scavenging by algal OM during periods of higher productivity. The relative balance between these processes varied over time and between compounds. The strongest evidence for a scavenging effect occurred with some of the more highly-chlorinated POPs (HCHs, chlordanes and dieldrin), during periods of rapid productivity increases and decreasing global usage. For other compounds (DDT, PCBs), there was

## Résumé

On sait que les récents changements climatiques ont considérablement augmenté la productivité primaire des lacs septentrionaux, entraînant une augmentation des concentrations hydriques de matières organiques provenant des algues ainsi que des flux de carbone provenant des algues dans les sédiments. Le but de ce projet d'une durée d'un an consistait à étudier si l'accélération de ces processus avait pu influencer les taux d'accumulation des anciens polluants organiques persistants (POP) dans les sédiments du lac Amituk de l'île Cornwallis, dans le Nunavut. Les résultats indiquent que les profils des POP sédimentaires intègrent deux processus historiques superposés et parfois opposés : les dépôts de POP atmosphériques témoignant d'émissions globales distantes et une augmentation du piégeage des POP par des matières organiques provenant des algues pendant les périodes de productivité accrue. L'équilibre relatif entre ces processus variait avec le temps et en fonction des composés. Les preuves les plus solides de l'effet de piégeage ont été constatées avec certains des POP les plus chlorés (HCH,



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minimal apparent relationship with algal productivity, and their sedimentary profiles were most closely related to production and use trends. This project underscores the need to consider climate-driven limnological changes in lakes when attempting to reconstruct atmospheric deposition rates of POPs and other contaminants.

## Key Messages

- The reconstructed atmospheric deposition rates of some, but not all, persistent organic pollutants (POPs) recorded in Amituk Lake sediments were magnified or diminished by coincidental variations in the amount of algal organic matter in lakewaters, resulting in sediment POPs profiles giving an inaccurate record of global production histories.
- These findings underscore the need to consider climate-driven limnological changes in lakes when attempting to reconstruct atmospheric deposition rates of POPs and other contaminants from sediments, e.g. as a means to judge the effectiveness of international regulations.

chlordanes et dieldrine) pendant les périodes d'augmentation rapide de la productivité et de diminution de l'usage global. Les autres composés (DDT, BPC) présentaient une relation apparente minimale avec la productivité des algues et leurs profils sédimentaires étaient plus étroitement liés aux tendances de production et d'usage. Ce projet souligne la nécessité de tenir compte des changements limnologiques attribuables au climat que subissent les lacs dans les efforts de reconstituer les taux de dépôts atmosphériques des POP et des autres contaminants.

## Messages clés

- Les reconstitutions des taux de dépôts atmosphériques de certains polluants organiques persistants (POP) (mais pas de tous) ont été amplifiées ou réduites par des variations concomitantes des quantités de matières organiques provenant des algues dans les eaux des lacs. Par conséquent, les profils de POP sédimentaires présentaient une image erronée de l'historique de la production globale.
- Ces conclusions soulignent la nécessité de tenir compte des changements limnologiques attribuables au climat dans les efforts de reconstitution des taux de dépôts atmosphériques des POP et des autres contaminants dans les sédiments comme moyen d'évaluer l'efficacité de la réglementation internationale.

## Objectives

1. Carry out analyses of organochlorine, polychlorinated biphenyl, and polybrominated compounds in sediment samples spanning the 20<sup>th</sup> Century, in a core collected from Amituk Lake in 2003.
2. Calculate flux rates over the last century for these compounds, using <sup>210</sup>Pb and <sup>137</sup>Cs dates and sedimentation rates determined for each core slice.
3. Test for associations between the fluxes of POPs and specific algal-derived forms of organic carbon, and determine whether the concentration and flux profiles more closely resemble global production patterns, within-lake algal productivity, or a combination of the two processes.

## Introduction

Persistent organic pollutants (POPs), including organochlorine pesticides like DDT and toxaphene, and industrial products such as PCBs, are frequently found in Arctic biota and in the abiotic environment (Braune et al. 2005; AMAP 2004). Lake sediments are a sink for these compounds, and sediment cores have been used to determine modern and historical fluxes of POPs into lakes all over the world, including in the Arctic (Muir et al. 1995; Tomy et al. 1999; Rawn et al. 2001; Stern et al. 2005).

One essential, but often unrecognized, assumption in using sediments for reconstructing atmospheric fluxes of contaminants is that the processes involved in environmental pathways and fate have

remained unchanged over time, so that the only source of variation in the sedimentary fluxes of contaminants is their atmospheric fluxes (Macdonald et al. 2005). However, this assumption is challenged by the fact that Arctic lakes have undergone unprecedented limnological change over the past century, including large increases in primary productivity, and alterations in dominant phytoplankton composition and benthic communities (Smol et al. 2005).

Primary productivity increases generally increase the concentration of biogenic particles in the water column, including phytoplankton, algae and their organic exudates and detritus. POPs are known to preferentially adsorb onto organic matter particles and colloids because of their hydrophobic behaviour, with the amount of adsorption possibly influenced by the degree of compound chlorination (Totten et al. 2001; Moon et al. 2004). Stern et al. (2005) reported that the post-1950 fluxes of some POPs (especially toxaphene and PCBs) in Lake DV-09 sediments on Devon Island were strongly correlated to changes in diatom numbers, which was taken as possible evidence of a climate-driven effect on the sedimentary accumulation of these compounds. Since then, we have carried out organic carbon speciation studies on Amituk Lake and Lake DV-09 sediments (see Fig. 1) using the well-established organic geochemical methodology called “RockEval” (data reported in Outridge et al. 2007). RockEval carbon speciation and quantification can more accurately reveal past algal productivity and biomass patterns than diatom numbers alone. Here, we examine possible relationships between POPs accumulation rates and algal productivity as reflected by sedimentary organic carbon patterns in Amituk Lake sediments.

## Activities in 2008/2009

The upper 14 slices of Amituk cores 1, 2 and 3 collected in 2003 corresponded to the last century (i.e. 1900 to 2003). We used RockEval organic carbon data from core 1, and analysed sediments from core 2 for POPs; there was insufficient material remaining from core 1 for POPs determinations. Both cores were  $^{210}\text{Pb}$  dated and showed similar sedimentation patterns and chronologies (Fig. 1). Core 2 dates were used for convenience in plotting both sets of parameters. The POPs analytical work was carried out using standard

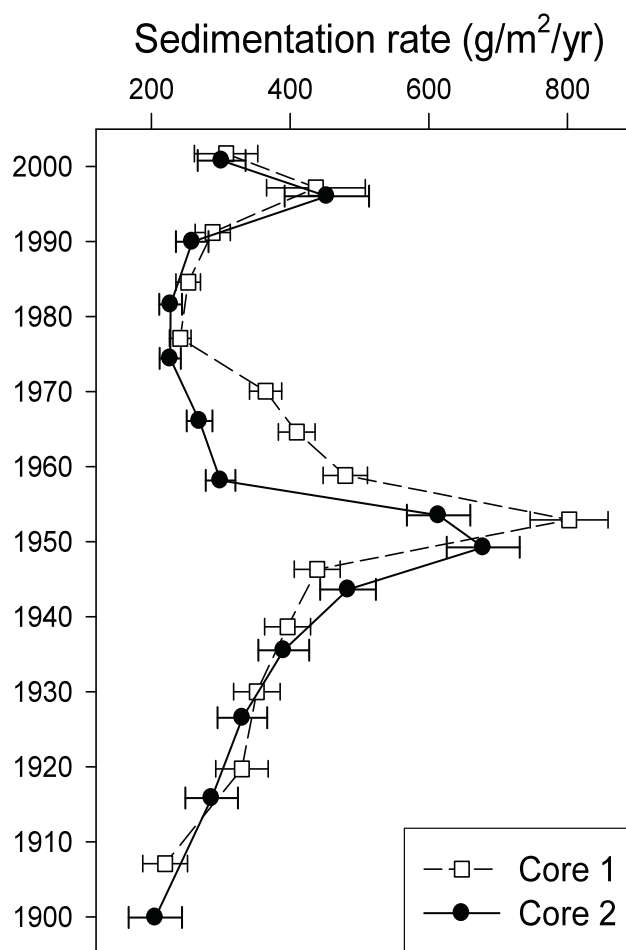


Figure 1. Sedimentation rates (error bars show  $\pm 2$  S.D. estimates) for cores 1 and 2 in Amituk Lake. (Core 1 was used for RockEval organic carbon analyses, and core 2 for POPs analyses)

techniques at a recognized specialist laboratory at DFO Winnipeg (Stern) which regularly participates in international and NCP interlaboratory QA/QC studies. Freeze-dried sediments were combined with anhydrous sodium sulfate and extracted in an accelerated solvent extractor (ASE 200, Dionex Canada, Oakville, ON). PCB 30 and octachloronaphthalene recovery standards were added before extraction. Sulfur was removed by treatment of the extracts with activated copper powder. Extracts were reduced in volume and fractionated on 1.2% deactivated Florisil. Fractions were then analyzed by high-resolution gas chromatography with electron capture detection (HRGC-ECD), using a 60 m DB-5 capillary column (0.25 mm ID, 0.25  $\mu\text{m}$  film thickness, J&W Scientific). Compounds are quantified by

comparing chemical peak areas (identified by retention time) to those of commercially available standards of known concentration. Sediment from a slice lower in the core (estimated as ~16<sup>th</sup> Century) was processed to serve as a procedural blank, which was used to correct the 20<sup>th</sup> Century data when necessary. Statistical associations were tested between the fluxes of POPs and the “S2” form of organic carbon (derived in this lake almost entirely from kerogen-like substances from algal cell walls, Outridge et al. 2007) using linear regression analysis (SigmaStat 2.0). Data were not transformed because they satisfied the necessary assumptions of data normality and constancy of variances.

## Results

Average northern hemisphere temperature trends during the 20<sup>th</sup> Century (Fig. 2a) were closely tied to temporal variations in algal-derived S2 carbon concentrations in Amituk Lake sediment (Fig. 2b and subsequently). Amituk S2 fluxes were almost identical to those in Lake DV-09 in terms of the

timing of increases and fluctuations and in their common 750% increase above their S2 fluxes since 1854 (Outridge et al. 2007). The S2 carbon history in Amituk gave a better agreement with air temperature history than did diatom numbers, which were absent from the sediment profile prior to 1950, probably because of diatom silica dissolution.

The clearest evidence for whether or not Amituk sediment POPs concentrations and fluxes were influenced by algal productivity or global production histories (or both) may be seen in those compounds with documented production or emission data against which to compare historical patterns of algal carbon sedimentation (Fig. 2).  $\Sigma$ HCH concentrations increased starting around 1950 and peaked in the early 1970s, a pattern that agreed well with known US and Chinese production data (Fig. 2b). However, after 1980 both  $\Sigma$ HCH and S2 carbon concentrations increased at a time when HCH production is believed to have declined significantly. Reduced dilution of POPs and S2 concentrations by lower sedimentation rate during this period cannot explain these findings

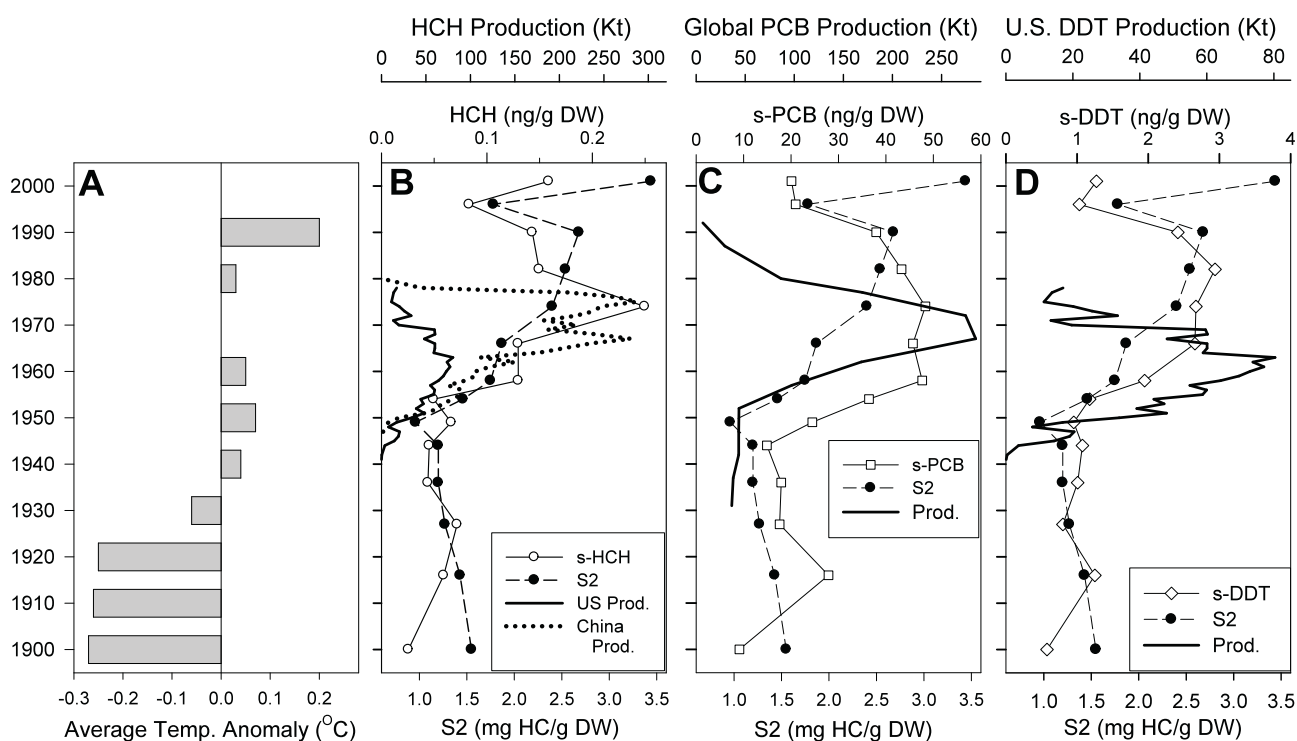


Figure 2. 20<sup>th</sup> Century decadal temperature deviations (A), and Amituk Lake sedimentary concentrations of algal-derived S2 carbon and POPs for which global production data are available (B.  $\Sigma$ HCHs; C.  $\Sigma$ PCBs; and D. DDT) (Northern hemisphere temperature data are deviations from the 1961-90 climate average, IPCC (1990), S2 carbon data from Outridge et al. (2007), POPs data from this study)

**Table 1. Results of correlation tests between fluxes of algal-derived S2 carbon and POP compounds into Amituk Lake sediments since 1900.**

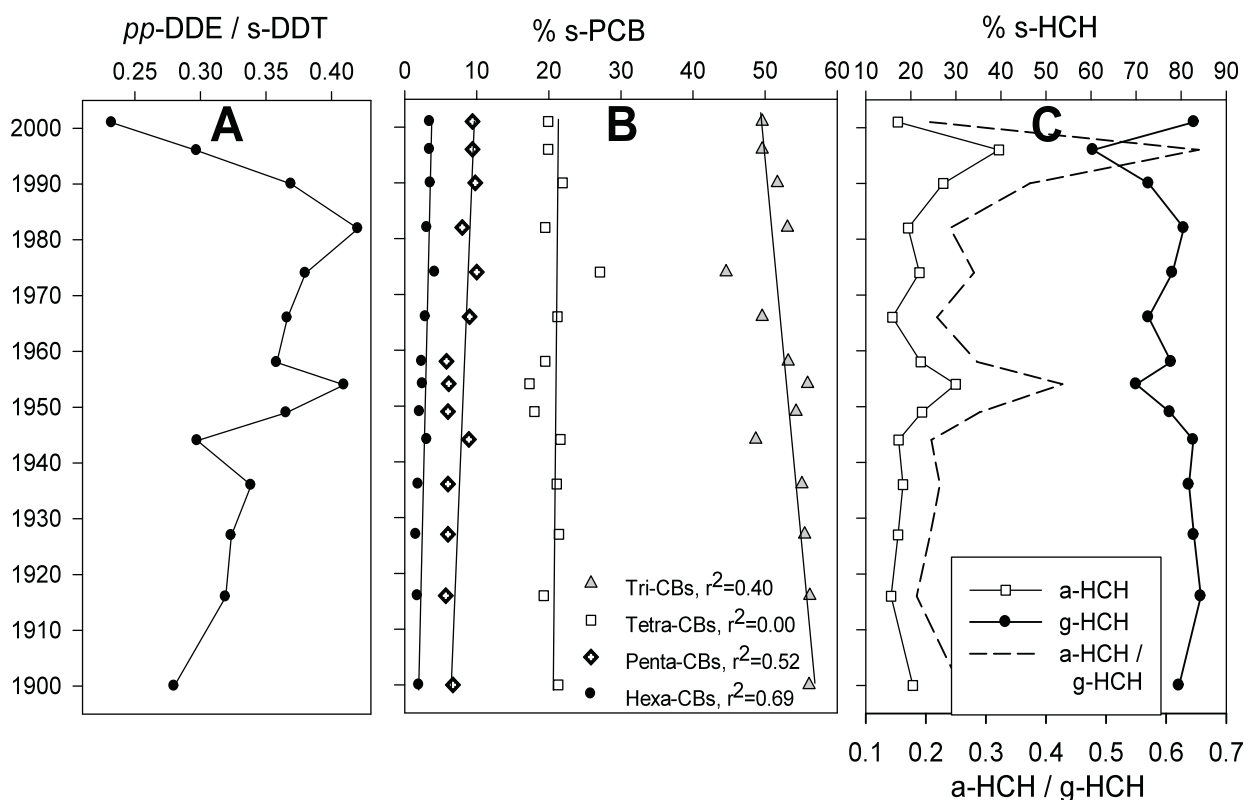
POPs Compound	S2 Correlation (r <sup>2</sup> value)	Significance level
ΣChlordane	0.46	**
ΣDDT	0.22	NS
Dieldrin	0.46	**
ΣHCH	0.55	**
ΣPCA	0.39	*
ΣPCB	0.14	NS

(NS – not significant ( $P > 0.05$ ), \* -  $P < 0.05$ , \*\* -  $P < 0.01$ , total df = 13)

because sedimentation was not reduced at this time (Fig. 1). Overall, the fluxes of S2 and ΣHCH in Amituk Lake were significantly correlated during the 20<sup>th</sup> Century ( $r^2 = 0.55$ ,  $P < 0.01$ ; Table 1).

ΣPCB concentrations attained constant levels of 43–48 ng/g DW over 25 years between 1958 and 1982, spanning the period of major global PCB production (Fig. 2c). After 1980, sediment [ΣPCB] declined by about half, at a markedly slower rate than global production. PCB and S2 fluxes were not correlated (Table 1). ΣDDT concentrations in Amituk Lake sediments increased shortly after U.S. production increased in the early 1940s (Fig. 2d), but remained relatively high (declining no more than 50% from its maximum) after 1953 despite a large decline in production. S2 flux was not related to DDT flux (Table 1). DDT remains in use in some countries, and this could account for the continuing high flux.

The input of “new” DDT into the Arctic was also indicated by the decline since ~1980 in pp-DDE as a proportion of ΣDDT (Fig. 3a). The PCB homologue profiles were dominated as expected by tri-CBs and tetra-CBs (Fig. 3b), but the relative abundance of homologues changed over time. Tri-CBs declined significantly in relative terms whereas penta- and hexa-CBs increased. ΣHCH in



**Figure 3. Changing proportions of (A) DDE, a DDT breakdown product, (B) PCBs and (C) HCH homologues in Amituk Lake sediments.**

Amituk sediments was dominated by  $\gamma$ -HCH, with the proportion of  $\alpha$ -HCH spiking in the mid-1950s and especially in the 1990s (Fig. 3c).

While the histories of production and emissions of chlordane, PCAs (pentachloroanisoles) and dieldrin are generally known, detailed information is not available, and so these compounds are plotted separately. Maximum [s-PCA] occurred in the mid-1970s in Amituk sediments and have remained relatively high since then (Fig. 4a); s-PCA and S2 fluxes were significantly correlated (Table 1).  $\Sigma$ Chlordane concentrations were highest in the 1970s but declined by only about 50% in the following decades up to 2001 (the upper slice in our core, Fig. 4b). Chlordane fluxes generally followed S2 carbon fluctuations during the 20<sup>th</sup> Century (correlation  $r^2=0.46$ ,  $P<0.01$ ; Table 1), and s-CHL fluxes remained only 10-20% below the 1970s maximum up to 2001. Dieldrin, also a widely-used insecticide during the 1960s to 1980s displayed considerable variability in Amituk sediments (Fig. 4c). Maximum dieldrin fluxes occurred in

1959, 1982, and in 2001, and were correlated with S2 carbon flux ( $r^2=0.46$ ,  $P<0.01$ , Table 1).

## Discussion and Conclusions

The POPs patterns in Amituk Lake sediments indicated two simultaneous processes were at work to varying degrees: atmospheric deposition of POPs related to their known production histories, and changing rates of POPs sedimentation (scavenging) due to temporally varying concentrations of algal organic matter in the water column. The relative importance of each of these processes varied over time, as the global production and emissions of the compounds changed and as lake productivity fluctuated with climate trends; it also differed between POPs compounds.

There were a number of similarities between the findings of Stern et al. (2005) concerning POPs and diatom abundance in Lake DV-09 on Devon Island, and those for Amituk reported here. The advantage of the present study is that the S2 carbon data gives a more precise and reliable measure of

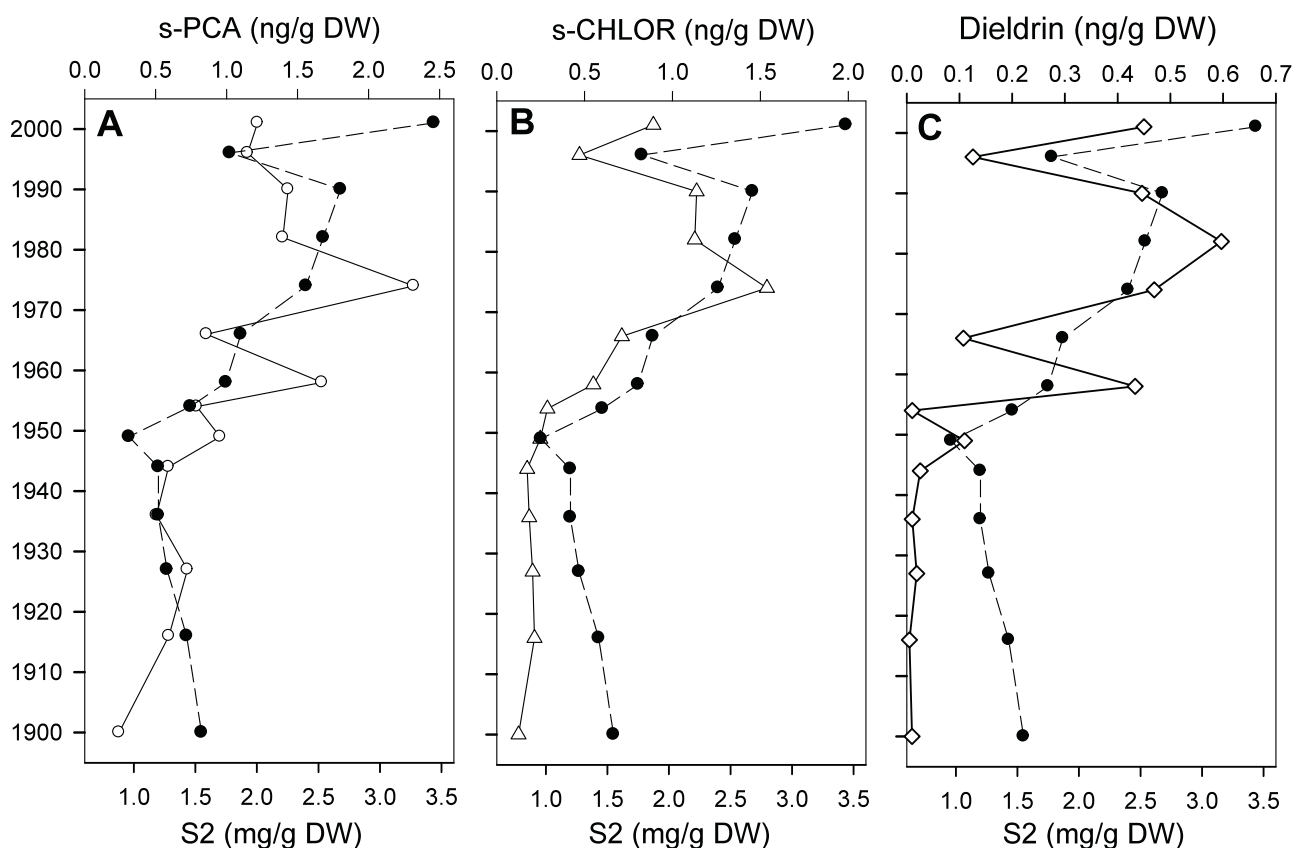


Figure 4. Concentrations of  $\Sigma$ Chlordane,  $\Sigma$ PCA and Dieldrin, as well as S2 carbon, in Amituk Lake sediments.

changing algal productivity and biomass through time than diatom enumeration. The substances in the present study which showed the most suggestive evidence of an algal scavenging effect included the highly chlorinated compounds  $\Sigma$ HCH,  $\Sigma$ Chlordane, and dieldrin. Stern et al. (2005) reported strong associations with changing diatom abundance for these compounds as well as PCBs and other chlorinated POPs in Lake DV-09. The key period from the perspective of separating a possible algal scavenging effect from a deposition effect is after 1980, when the production and use of many of those POPs were curtailed or eliminated and algal productivity increased significantly.

The dissociation of the sedimentary trends of these POPs from their global trends of declining production and use during the 1980s to 2001 could be evidence of an influential process at work that is operating independently from atmospheric deposition. HCH concentrations and fluxes remained high two decades after its global production ceased, and even increased in the most recent sediment layer in our cores. For chlordane and dieldrin, in the absence of a precise production or emission history with the same high degree of resolution as the Amituk algal carbon record, our interpretation is more cautious. The use of chlordane, an agricultural insecticide, was banned in Canada, the U.S.A. and Europe in the 1980s but continued to be used elsewhere to some degree until 1997 (PANNA 1997). The lack of a significant decline (<20%) in  $\Sigma$ Chlordane flux in Amituk after the 1980s is surprising given an atmospheric deposition interpretation, but is explicable in the light of the relatively high and increasing rates of algal productivity during those decades. Dieldrin's use was severely curtailed in the 1980s and eliminated globally in the 1990s, but Amituk dieldrin concentrations and fluxes remained at near-maximal levels up to 2001. These POPs-S2 associations over recent decades may be evidence of a recently-increased rate of algal scavenging acting to maintain sediment fluxes of POPs at levels above their atmospheric deposition trends.

The sediment records of DDT, PCBs and of HCH during the 1950s to 1970s, appeared to mimic their respective global productions without perceivable influence from within-lake processes. The HCH data is especially valuable because the close agreement in timing between the ramping up and

decline of US and subsequently Chinese HCH production, and the sediment record during those decades, confirms the accuracy of the core's  $^{210}\text{Pb}$  dating. It is notable that algal productivity was relatively constant during this period, which may explain why HCH and PCB production changes were so precisely mimicked in the sediments.

Based on the S2 carbon data, the within-lake process of organic matter particle scavenging of POPs is suggested to be an important factor in producing the measured sedimentary profiles of certain POPs in Amituk Lake. Other environmental processes described by Macdonald et al. (2005) could also be simultaneously influencing POPs accumulation rates in Arctic lake sediments, including climatic alterations of air mass transport to and precipitation scavenging in the Arctic, compound soil-to-air volatility in southern source regions and condensation from Arctic air, and the rate and seasonal timing of POPs flushing from lake catchments in springtime, among others. The findings of this study emphasize the need to consider possible natural system changes as mitigating or enhancing factors that may confound the reconstruction of atmospheric POPs deposition rates using northern lake sediments. The implications for science and policy could be important. For example, if increased algal scavenging is maintaining current sediment POPs accumulation rates at abnormally high levels relative to atmospheric deposition, the erroneous conclusion may be drawn that international POPs emissions controls were less effective than was the case. Further exploration of this topic is warranted.

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# ***Education and Communications***





# Northern Contaminants Program Synopsis Report

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## ◆ **Project team:**

Yukon First Nations

Yukon Contaminants Committee

## **Abstract**

The Council of Yukon First Nations (CYFN), in partnership with the Yukon Contaminants Committee, has been involved with different projects, including air measurements at Alert and Little Fox Lake; and the health risk assessments for mercury levels in fish in the Yukon.

The key Yukon message remains that the benefits of eating traditional foods far outweighs the risks of trace amounts of long-range contaminants that may be found. This message comes from a CYFN-McGill Dietary Study that was completed in 1998, which analyzed the risks and benefits of eating traditional foods. While the results of this study showed that the benefits of consuming traditional foods far outweigh the risks, there continued to be concerns expressed. In order to better understand what people were hearing and retaining through our efforts to communicate, we organized an in-depth survey of Yukon communities. We want to know if the messages “Our Traditional Foods Are Safe to Eat” were getting out, and that the training and workshops that we were conducted over the years had been beneficial. We had over 300 respondents. The main message was that community people wanted their own workshops, which we have tried to incorporate over the past few years.

The information coming from the NCP is sent to the First Nations and to the general assemblies. If a concern arises the Yukon Contaminants Committee

## **Résumé**

Le Conseil des Premières nations du Yukon (CPNY), en collaboration avec le Comité des contaminants du Yukon, a participé à différents projets, y compris des mesures de l’atmosphère à Alert et à Little Fox Lake et une évaluation des risques pour la santé posés par les niveaux de mercure dans les poissons du Yukon.

Le principal message pour le Yukon reste que les avantages de consommer des aliments traditionnels surpassent largement les risques présentés par les quantités négligeables de contaminants qu’ils pourraient contenir. Ce message émane d’une étude sur l’alimentation effectuée par le CPNY et McGill terminée en 1998, qui analysait les risques et les avantages de consommer des aliments traditionnels. Les résultats de cette étude démontrent que les avantages de consommer des aliments traditionnels l’emportent largement sur les risques, cependant certaines préoccupations subsistaient. Afin de mieux comprendre ce que la population avait entendu et retenu de nos efforts de communication, nous avons entrepris un sondage approfondi auprès des collectivités du Yukon. Nous souhaitons savoir si le message selon lequel « nos aliments traditionnels sont sans danger pour la consommation » était clairement communiqué et si les formations et les ateliers que nous avons organisés au cours des années avaient porté fruit. Nous avons obtenu plus de 300 réponses. Le principal message qui est ressorti de ce sondage

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Chair is informed. We continue to communicate the benefits of traditional food in the Yukon and to continue to support NCP research that show that our foods are safe to eat.

## Key Message

- Our Traditional Foods are Safe to Eat

This key message may change depending on what is going on with Yukon fish.

était que les habitants des collectivités souhaitent bénéficier de leurs propres ateliers, demande à laquelle nous nous sommes efforcés de répondre au cours de ces dernières années.

L'information provenant du PLCN est transmise aux Premières nations et aux assemblées générales. Le président du Comité des contaminants du Yukon est informé en cas de problème. Nous continuons à souligner les avantages des aliments traditionnels au Yukon ainsi qu'à soutenir les recherches du PLCN afin de démontrer que nos aliments sont sans danger pour la consommation.

## Message clé

- Nos aliments traditionnels sont sans danger pour la consommation.

Ce message clé peut changer en fonction de la situation des poissons dans le Yukon.

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## Objectives

- To enhance the confidence of Yukon First Nations in making informed decisions about traditional food consumption and other health-related factors.

## Introduction

Over the years there have been substantial amounts of contaminants information disseminated to Yukon and First Nation communities through workshops, conferences and training. In the Yukon, long-range contaminant sources are low and traditional foods such as caribou and moose are safe to eat. Our communication efforts have included workshops, radio shows, posters, presentations, poster contests, curriculum development, front-line training courses, a Yukon wide survey and most recently the concentration has been to participate in research with a focus on mercury.

Mercury emissions from Asia have increased between 1990 and 2000 while other regions such as Europe and North America have decreased in that time period. Modeling has shown that air masses enriched with mercury pollution from the Pacific Rim enter the western Canadian Arctic. There has not been any atmospheric mercury measurements collected in

the western Canadian Arctic in the past to ascertain the impact of these transport events. To address this gap in knowledge there has been funding from the NCP allocated for the Intercontinental Atmospheric Transport of Anthropogenic Pollutants to the Arctic (INCATPA), to take measurements of mercury in the air at Little Fox Lake. This is an International Polar Year (IPY) project that began in the summer of 2006. This year we are fortunate to have a summer intern through this project who has developed posters and presentations and is doing community outreach activities in various communities.

Complementing this research is a project to complete regionally relevant health risk assessments (HRAs) for mercury levels in fish in the Yukon lakes. In the Yukon, the Chief Medical Officer (CMO) is responsible for providing health advice to Yukon. In the case of health advice or specific health advisories pertaining to environmental contaminants such as mercury, the CMO undertakes this responsibility in consultation with the Yukon Contaminants Committee (YCC) and Health Canada (as required). The CMO is an active member of the YCC, and is therefore a team member for this project. Mercury data was compiled, analyzed and presented to the CMO in conjunction with northern health experts and

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YCC representatives. The research, led by Dr. Laurie Chan, indicated that the levels of mercury in most fish were below Health Canada Guidelines and do not pose a risk at average or even at well above average consumption levels.

## **Activities in 2008/2009**

- Supervised an INCAPTA intern, Tonya Makletzoff
- CYFN General Assembly, July 2009
- Community General Assemblies in Yukon and Northern BC
- Distributed fact sheets, synopsis reports. AMAP reports, CACAR I and II
- Participate in meetings of preliminary research results

## **Results**

In 2008-2009 there was an effort to present NCP information to regional and community general assemblies, including the Council of Yukon First Nations. Booths were set up in various community general assemblies, including Burwash Landing, Haines Junction, Watson Lake and Dawson City. With results just coming in from work over the past year there will be an effort to bring them to communities in the form of posters, presentations and general assemblies. This will have to be done in coordination with the communities, CYFN, researchers and YCC.

However, larger sizes of lake trout and burbot, at least in certain locations, do contain enough mercury to warrant limitations on intake by women of childbearing age and children under 12. A specific guideline to this effect is being developed by Yukon Government with guidance from the CMO and the YCC.

## **Discussion and Conclusion**

Both the on-going monitoring program and the new research projects are both exciting and informative. While our main focus has been to emphasize the important nutritional value of our traditional foods it's also important that we continue to monitor and provide new information to communities on research results, both preliminary and conclusive. With this in mind the next phase of the project should include posters, presentations and handouts for communities.

## **Expected Project Completion Date**

On-going

## **Acknowledgements**

Yukon First Nations communities, Tonya Makletzoff, Laurie Chan, Jody Butler Walker, and Pat Roach



# Continuing to meet the information needs of Nunatsiavummiut

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## **Abstract**

In 2008-09 the ongoing activities of the Northern Contaminants Researcher (NCR) continued to communicate and educate the Nunatsiavummiut population on contaminants, wild foods and health issues so that they may make informed decisions in their daily lives. With two committees within NG, the Interim Research Committee and an NCP funded Nunatsiavut Health and Environment Review Committee (NHERC), the NCR is providing advice and assistance to the development of a research protocol for Nunatsiavut as well as ensuring that contaminant issues/information are now provided to each of the communities and gathered in a regional hub, therefore strengthening the already existing relationship between the NCR and the region.

One of the main areas of concern to Nunatsiavummiut is the contamination at an old military site which is approximately 1km from the community of Hopedale. This site also resulted in contamination round some homes in a subdivision within the community. While this is obviously not a long-range contaminant issue, Nunatsiavummiut need to understand both local and long-range

## **Résumé**

En 2008-2009, le responsable de la recherche sur les contaminants dans le Nord (RRCN) a poursuivi ses activités afin de diffuser aux Nunatsiavummiut de l'information sur les contaminants, la faune et la flore sauvages et les questions de santé et de les éduquer de sorte qu'ils puissent prendre des décisions éclairées dans leur vie de tous les jours. En sa qualité de membre de deux comités au sein du gouvernement du Nunatsiavut, soit le comité intérimaire de la recherche (*Interim Research Committee*) et le comité d'examen de la santé et de l'environnement du Nunatsiavut (*Nunatsiavut Health and Environment Review Committee* ou NHERC) qui est financé par le Programme de lutte contre les contaminants dans le Nord (PLCN), le RRCN offre de l'aide et des conseils en vue de l'élaboration d'un protocole de recherche pour le Nunatsiavut et veille à ce que les questions ou l'information relatives aux contaminants soient dorénavant communiquées à chacune des collectivités et regroupées au sein d'un centre de coordination régionale, renforçant ainsi les liens déjà solidement établis entre le RRCN et la région.

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issues in order to put the contaminants issue in context. Nunatsiavut Government coordinated this project by providing resources and logistical support, assisting in some field work sampling and ensured community concerns were addressed. A Human Health Risk Assessment (HHRA) conducted last year shows that there is significant risk posed to people visiting the site. This project is now underway to a clean up plan and the NCR is working side by side with the Government of Newfoundland and Labrador and the community of Hopedale to ensure that community members are informed and included in all stages of the work in Hopedale.

L'un des principales préoccupations des Nunatsiavummiut est la contamination d'un ancien site militaire situé à un kilomètre environ de la collectivité de Hopedale. Ce site est également à l'origine de la contamination observée aux alentours de quelques demeures dans l'un des secteurs de la collectivité. Même s'il ne s'agit manifestement pas d'un problème de contaminant à grande distance, les Nunatsiavummiut doivent saisir à la fois les enjeux locaux et à long terme pour pouvoir situer le problème des contaminants dans son contexte. Outre qu'il a coordonné ce projet en fournissant des ressources et un soutien logistique et en collaborant à certains travaux d'échantillonnage sur le terrain, le gouvernement du Nunatsiavut a veillé à ce que les préoccupations de la population soient abordées. Selon les résultats de l'évaluation du risque à la santé humaine (ERSH) menée l'an dernier, les personnes qui se rendent sur les lieux courent un danger non négligeable. La phase de nettoyage du projet est maintenant en cours et le RRCN collabore étroitement avec le gouvernement de Terre-Neuve-et-Labrador et la collectivité de Hopedale pour garantir que les membres de la collectivité sont informés et consultés à toutes les étapes des travaux effectués à Hopedale.

## Key Messages

- The NCR strives to, in a culturally relevant manner, continue its communication efforts on contaminants, research and the environment, conduct research and promote mutually beneficial relationships between the communities of Nunatsiavut and outside scientists.
- Nunatsiavummiut have a number of concerns about their health and the environment around their communities. The creation of the NHERC has helped these communities come together with common issues, gain understanding of common issues in other regions and understand what NCP's mandate is.
- NHERC has become a portal for communication and exchange of information on contaminants issues/research in the region.
- A much-needed research facility has been approved for funding for the Nunatsiavut region.

## Messages clés

- Tout en tenant compte des différences culturelles, le RRCN poursuit ses efforts en matière de communication de renseignements sur les contaminants, la recherche et l'environnement, mène des recherches et facilite les relations entre les collectivités du Nunatsiavut et les scientifiques d'ailleurs.
- Les Nunatsiavummiut ont un certain nombre de préoccupations au sujet de leur santé et de l'environnement autour de leurs collectivités. La création du NHERC a permis à ces collectivités de faire front commun en ce qui a trait aux enjeux collectifs, d'analyser les problèmes également observés dans d'autres régions et de saisir la nature du mandat du RRCN.
- Le NHERC est devenu un portail de communication et d'échange de renseignements sur les problèmes ou la recherche ayant trait aux contaminants dans la région.
- Le financement d'installations dont la région du Nunatsiavut a absolument besoin a été approuvé.

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## Objectives

The objective of the Northern Contaminants Program (NCP) is to reduce or, wherever possible, eliminate contaminants in traditionally harvested foods while providing information that assists informed decision making by individuals and communities related to their food use.

This year the Nunatsiavut Government NCP Researcher:

- Assisted residents of Nain, Hopedale, Postville, Makkovik, Rigolet and Happy Valley-Goose Bay by providing information about risks, the means to reduce risks, and information on the benefits of traditionally harvested foods to support residents in making informed decisions;
- Assisted communities of the Nunatsiavut and the Lake Melville area in becoming involved with contaminant issues and activities that affect the people and the region through the creation of the NHERC
- Sits on the Nunatsiavut Government's Interim Research Advisory Committee
- Assisted in securing funds for a much needed research centre which will be constructed in Nain in 2009/2010. This research centre will be equipped with the facilities that are much needed in the region. This includes a wet and dry lab, accommodations, library, freezers etc.
- As part of her ongoing work on the issue of local contaminant sources at abandoned military sites, the NCR was invited to a conference on abandoned Military Dewline sites and traveled to Timmons, ON in the fall of 2008 to give a presentation on the Hopedale experience and learn about similar experiences by Ontario First Nations, who are dealing with similar issues and shared their information about the contaminated sites in Ontario. The expectation of this type of networking and sharing of information about these sites will support the need to clean up these sites.

## Introduction

Nunatsiavummiut are sustained by the animals, birds, fish and plants of the region. Research has

shown that contaminants are present in these food sources due to the environment being contaminated by varying sources of pollution (local and distant) which are then transported to the north via atmospheric and oceanic mechanisms. The levels of contaminants in these foods which sustain Nunatsiavummiut and potential effects they may have on residents of the coastal communities are of concern to the Inuit of Nunatsiavut. The Nunatsiavut Government Research Office helps to ensure that Nunatsiavummiut are heard and their concerns with regard to both national and local contaminants issues are assessed and communicated. The current issues for Nunatsiavut Inuit are similar to those of other Inuit regions, but there are also local concerns that need to be communicated and the minds of Nunatsiavummiut to be settled with regard to some perceptions regarding local contaminants.

Hopedale, Labrador, a community of approximately 625 residents, has a former US Air Force early-warning radar station near the community, has been voicing concern of possible contamination for a number of years. In the last couple of years, the NG collaborated with the Environmental Sciences Group (ESG) in an effort to assess the situation in Hopedale. This project is now being lead by the Government of Newfoundland and Labrador. The NCR is working closely with the Provincial Government and the community to begin working on a clean up plan.

In order for Nunatsiavut Inuit to be informed about wise food choices the NG Research Office must coordinate efforts between Inuit Tapiriit Kanatami, NCP, university and government researchers and our communities. Following the objectives of the Northern Contaminants Program, the NG's Researcher acts as a key resource person and provides information to the population in a culturally relevant and plain language format on contaminant-related issues. The NHERC has proven to be a very helpful tool in ensuring that each Inuit community is represented and has a voice, not only when dealing with NCP related research, but also through our National Inuit Organization; ITK. ITK is also a member of this committee.

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## Activities in 2008/2009

### *Communication*

#### **Avativut Newsletter**

This newsletter is a publication that the NG Research Office has developed and used to communicate to the Labrador Inuit population about such things as the benefits and risks of wild foods, the most recent knowledge on contaminants, health and environmental issues and to update people on current research activities in the region. The existence of a regional person and their involvement in this publication has enhanced the NG's ability to communicate such information in a culturally-relevant and timely manner. Without this person, this newsletter would not be published. The main focus of the Avativut newsletter is on the benefits of consuming wild foods so as to reinforce the understanding among the general population that they are still the most nutritious foods for Nunatsiavummiut to eat. Additional updates of new and ongoing research activities and relevant facts about health and environment issues in the region are also provided in this newsletter which is printed in English and Inuktitut (Labrador dialect). A section of the Avativut Newsletter is also set aside for environmental news from each of the Nunatsiavut communities, giving each community a chance to share their concerns and accomplishments with the coast regarding environment and health issues.

#### **Participation In Research Projects**

Although many of the issues that the NG is currently dealing with related to the issue of contaminants are associated with local source problems (e.g. Hopedale radar base) in the eyes of community members there remains the need to address these concerns. The public does not make the separation between local and long range sources and therefore communication activities in the region this year have had to deal with the issue of contaminants in a coordinated and combined manner. The NHERC has made some progress in explaining the difference through community representatives. Regional review of NCP proposals through NHERC has also raised awareness of projects in communities in Nunatsiavut, thus creating interest in having similar research done in other communities.

#### **Nunatsiavut Nuluak: Researchers and Inuit working toward understanding influence of climate change and human activities within three fiord ecosystems in northern Labrador**

Nunatsiavut Nuluak project, funded by ArcticNet and co-led by the Nunatsiavut Government and the Environmental Sciences Group of the Royal Military College of Canada, was initiated in the spring of 2006 to address Inuit concerns of environmental change and human activities which may be contributing to these changes in northern Labrador. The objectives of the program are to create a baseline inventory and comparative assessment of three fiord based ecosystems (Saglek, Anaktalak and Nachvak Fiords) and document and incorporate Inuit knowledge and concerns in all facets of the program. In the summer of 2008, the Nunatsiavut Government and ArcticNet Nunatsiavut Nuluak Office, with support from the Nasivvik Centre and International Polar Year, were able to give 5 students a chance to work side by side with Inuit from Nunatsiavut and Nunavik as well as researchers from several universities and government agencies in the Torngat National Park. Anita Fells, Sheena Merkuratsuk, Elias Obed, Dorothy Angnatok and Minnie Okkuatsiak spent five weeks rotating in different areas of research this summer. These areas included sampling of small organisms in water to vegetation, research on glaciers, collection of samples and more.

#### **Temporal Trends and spatial variations in persistent organic pollutants and metals in sea-run char from the Canadian Arctic**

Sea-run Arctic char are eaten by many Arctic communities and are valued as a good food source in addition to other food items such as seals and appropriate store bought food. From time to time, the government measures the levels of various chemicals in the food that people eat to make sure that the food is safe to eat. The government also does this to see if chemical levels are changing. Some of these programs measuring chemicals in fish are linked with scientific studies which are investigating different aspects of the environment. For these reasons, we conducted a study of the level of chemical levels in sea-run char harvested by Nain. This study is part of the Northern Contaminants Program which is measuring chemicals in



many kinds of animals including seals, beluga, and freshwater fish. This study is being conducted by Environment Canada.

Dr Evans visited Nain this year along with Jonathan Keating to demonstrate how they dissect the charr once they have arrived in Saskatoon.

The NCR ensured that Dr. Evans' visit would be of benefit to both the community and Dr. Evans by utilizing different avenues of communication that are known to be successful in the region. This included a radio interview with the OKalaKatiget Society, which is aired regionally, a community open house and a public demonstration at the local fish plant of how fish are dissected and what Dr. Evans' looks for during her research. The NCR was present at the open house, participated in the public demonstration on fish dissection, helping to communicate on techniques used to locate the gonad (ear bone) etc. of char.

### **Nasivvik Centre for Inuit Health and Changing Environments**

The Nasivvik Centre is a multidisciplinary research and training centre funded by the Canadian Institutes of Health Research-Institute of Aboriginal Peoples' Health. The Centre is focused on building capacity in Inuit health

research through trainee support and strategic funding initiatives in key environmental health areas of importance to Inuit communities.

The Nasivvik Centre also provides part of the funding for the Inuit Research Advisor (IRA) in Nunatsiavut, John Lampe. In cooperation with the NCR and the Nunatsiavut Government, the IRA position has led to better overall research coordination among projects funded under the various research programs going on in the North and an enhancement of training opportunities associated with research. The NCR is a member of the Board of Directors for the Nasivvik and takes part in meetings and conference calls to deal with any issues regarding the centre including reviewing funding proposals.

### **Nunatsiavut Health and Environment Review Committee**

The Nunatsiavut Contaminants Committee (Now the NHERC) began in late 2007. This committee consists of the NCR who is the Chair of the committee, the Nunatsiavut Inuit Research Advisor, Director, Nunatsiavut Government Environment Division, NG Dept. of Health, ITK, NCP and a representative from each of the 5 communities along the coast and Upper Lake



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Melville (ULM). The current co-chair is from Northwest River (ULM), Ed Tuttau. During its first year we have experienced some difficulty maintaining membership however have reached a point where the Committee is well-established. The NCR is the Chair of the committee and is responsible for securing funding for the committee and coordinating the committee activities, preparing for face to face meeting and teleconferences which include logistical planning, travel arrangements, orientation of new members, preparing minutes, disseminating proposals for review, collecting information, preparing documents and insuring regional concerns are forwarded to the NCP. Terms of Reference for the NHERC have been developed.

### ***Ongoing Daily Communications and Research Coordination***

In addition to these specific activities, a number of ongoing communication responsibilities are fulfilled by the NG Research Office staff. Daily activities of the Research Office include responding to community concerns, providing information to the Nunatsiavut Government, communities, and individuals on issues relating to contaminants, the environment and health, and acting as a liaison for the various people proposing to, and currently conducting research in the region. Additionally, the research staff acts as a liaison for interactions between the regional organizations and ITK and the NCP. This involves regular interaction with various individuals and ongoing communications efforts.

## **Discussion and Conclusions**

The NCR continues to be an essential part of the work of the Nunatsiavut Government. This is especially true for the residents of Hopedale, and will continue to be so for other Nunatsiavut communities who have concerns about contaminants. The NG Research Office continues to educate and empower the people of Labrador to better understand and deal with contaminants in their environment and traditional foods, and be aware of research and general environmental issues.

The Nunatsiavut Government Contaminants Researcher continues to:

- Support the activities undertaken by NG and NCP in providing information on research about contaminants, their effects on wildlife and

humans through consumption of wild foods which are based on the varied language and geographic needs of individuals and communities of Labrador;

- Enhance decision making abilities of Labrador Inuit through the delivery of information on risks and benefits of contaminants and wild foods relevant to the region in an accurate, timely and accessible manner;
- Develop regionally relevant resource materials in consultation with the coastal communities and appropriate agencies (i.e. Department of Health and Social Development, DIAND-NCP, ITK etc.). These materials include educational materials such as a quarterly newsletter; all publications are produced in both Inuktitut and English;
- Use the research results from studies conducted in the region to aid in effective delivery of information;
- Be a member of the NG Interim Research Advisory Committee, responsible for interacting with and assisting outside researchers with community consultations. This assistance also includes negotiating research agreements between researchers and community organizations and reporting project results to communities in a timely and responsible manner. The Research Office determines, in consultation with community representatives who are responsible for communication on contaminant, health and environment issues, which medium(s) best suit the information needs of the community, etc. This person will continue to assist in the development of a Protocol and Guidelines for Research conducted in Nunatsiavut.
- Continue to act as Chair of the NHERC, ensuring that communities are involved with in and informed about community concerns in the region, coordinates all logistical and administrative aspects of the committee
- Take part in research projects and communication of research results when appropriate.

## **Date of Completion**

This is an ongoing project in Nunatsiavut.



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## Acknowledgements

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# Developing Communication and Research Monitoring Infrastructure for the Inuvialuit Settlement Region

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## **Abstract**

All three of the projects proposed by the Inuvialuit IRA were completed, including two editions of the Health and Environment Newsletter, a community tour to all six Inuvialuit communities and the revamping of the health and environment page on the IRC website. Other deliverables and objectives that were also achieved include the building of regional partnerships within and outside of the ISR, the continued standardization of the IRA position with regional and national organizations (review), and obtaining detailed lists of research from licensing agencies and funding bodies. The IRA also maintained presence in existing committees and joined new ones that helped to ensure the IRA was in the forefront of research. The IRA also took part in other projects of importance to Inuvialuit including the Inuit Health Survey and Health Foods North. All of these activities have led to the new ways of effectively improve the IRA position and make it more successful for Inuit. In the coming year, the work and headway that was made will continue to be implemented. Some of the proposed projects and continuation of the work to date include, expanding the Health and

## **Résumé**

Les trois projets proposés par le conseiller en recherche inuvialuit (CRI) ont été menés à terme, y compris la diffusion de deux numéros du bulletin traitant de la santé et de l'environnement (*Health and Environment Newsletter*), une tournée des six collectivités inuvialuites et le remaniement de la page ayant trait à la santé et à l'environnement du site Web du Conseil régional inuvialuit. Au nombre des autres travaux et objectifs réalisés, signalons également l'établissement de partenariats régionaux au sein de la région inuvialuit désignée (RID) et à l'extérieur de celle-ci, la normalisation accrue des rapports du CRI avec les organismes régionaux et nationaux (examen) et l'obtention de listes détaillées des activités de recherche provenant d'organismes responsables de la délivrance de permis ou d'organismes de financement. De surcroît, le CRI a siégé au sein des comités existants et s'est joint à de nouveaux comités afin de se tenir au courant des derniers développements dans le domaine de la recherche. Il a également pris part à d'autres projets d'importance pour les Inuvialuit, entre autres l'étude sur la santé des Inuits (*Inuit Health Survey*) et celle portant sur les aliments de santé dans le Nord

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Environment newsletters from two to four editions in the year, establishing a regional contaminants and research working group, and holding another round of community visits. Community tours will be done based on the suggestions made by the communities and lessons learned from the IRA. With recommendations made at the IRA review, standardization will be implemented at all levels and organizations involved with the IRA position including, funding bodies, host organizations and support organizations. The IRA will also continue to be part of projects, working groups and committees that are of interest to Inuvialuit including, the Nasivvik Board of Directors, NWT RCC, and the Inuit Tipingat Committee. Overall, this year had been another year of adjustments and we are continuing to see the IRA position develop. With new IRAs in two of the four regions this year alone, there is a definite need to make the position more present in the scientific community and at home in Inuit regions before it can be truly utilized by all.

(*Health Foods North*). Toutes ces activités se sont traduites par une plus grande efficacité du CRI, qui sert d'autant mieux les intérêts des Inuits. L'an prochain, les travaux et progrès réalisés se poursuivront. Voici quelques-uns des projets proposés et travaux effectués jusqu'à présent : diffusion d'un plus grand nombre de bulletins sur la santé et l'environnement, passant de deux à quatre numéros par année, formation du groupe de travail régional sur les contaminants et la recherche et organisation d'une autre tournée des collectivités. Ces tournées seront organisées en fonction des suggestions formulées par les collectivités et des leçons apprises par le CRI. Suivant les recommandations faites dans le cadre de l'examen effectué par le CRI, la normalisation s'étendra à tous les niveaux et à tous les organismes travaillant avec le CRI, y compris les organismes de financement, d'accueil ou de soutien. Le CRI continuera à faire partie de projets, groupes de travail et comités intéressant les Inuvialuits, entre autres, le conseil d'administration du Centre Nasivvik, le Comité régional des contaminants des Territoires du Nord-Ouest et le comité Inuit Tipingat. Dans l'ensemble, l'année a été consacrée à des rajustements et le poste de CRI continue à évoluer. Des nouveaux conseillers en recherche inuite ayant été nommés durant l'année dans deux des quatre régions, il ne fait pas de doute que, pour pouvoir être utiles à tous les intervenants, les conseillers en recherche inuite doivent désormais être plus présents au sein de la collectivité des scientifiques et dans les régions inuites.

## Key Messages

- More youth and elder involvement is needed, which can only be made possible through dedication and actual intention to work with Inuit.
- We need a way to ensure that researchers budget money in their project for training and capacity building outreach in the ISR.
- ARI needs to include in their application/forms specific questions that are of significance to IRC and Inuvialuit. A recommendation that will be made is for ARI to make researchers identify if they are willing to add credits and acknowledgements in research (co-authorship, references, etc) for Inuit people.

## Messages clés

- Il faut mobiliser un plus grand nombre de jeunes et de personnes âgées, d'où la nécessité de faire preuve de bonne volonté et de démontrer l'intention claire de travailler avec les Inuits.
- Nous devons nous assurer que les chercheurs préparent les budgets de projet en prévoyant de financer la formation de même que le volet du renforcement de la capacité au sein de la RID.
- L'Institut de recherche Aurora doit inclure dans les demandes ou formulaires des questions qui ont de l'importance pour les Conseil régional inuvialuit et les Inuvialuits. On lui recommandera de demander aux chercheurs d'indiquer s'ils seraient disposés à inclure des mentions de source ou à indiquer l'origine de l'information

- We need to better streamline the licensing process, so that community organizations and IRC know how it works. (Pamphlet that was made by ARI was good, but they are meant for researchers, and there needs to be one made from a community perspective, so that everyone understands their place in research reviews.

utilisée aux fins de la recherche (coauteurs, références, etc.) pour ce qui est des Inuits.

- Nous devons simplifier encore plus le processus de délivrance de permis, afin que les organismes communautaires et le Conseil régional inuvialuit sachent de quelle manière il fonctionne. L'Institut de recherche Aurora a préparé un bon dépliant qui s'adresse toutefois aux chercheurs; il devrait y en avoir un destiné aux collectivités afin que tous les intervenants sachent quel rôle ils jouent dans l'examen de la recherche.

## Objectives from 2008-2009 year

- Continue with communication initiatives (website, meetings, etc)
- Continue to make regional newsletters relevant to communities
- Community visits must be done close together (keep the message going)

## Introduction

Since the IRA position was introduced into Inuit regions in 2005, the purpose of the position has been to improve the way that research is done in the North. A lot of work has been accomplished to better make the IRA position work for communities, host organizations and funding bodies. It started off with monthly teleconferences that encouraged more partners to get involved in our work, and ever since IRA's have been active in many aspects of this change and have gained many partners such as ICC-Canada, ITK and NAHO. For almost four years, IRA's have been introduced slowly into the scientific community and attended major events such as the ArcticNet Annual Scientific Meetings, and the NCP Results Workshops where we have influenced scientists (licensing and consultation), and paved the way for new protocols to be implemented in the North (Tri-Council Policy statement). At these events IRA's have been developing regional posters to explain to researchers and the scientific community how work should be conducted in Inuit regions. Some of these explanations have included information about licensing research, seeking community involvement and keeping with

regional guidelines or expectations for research (How to harmoniously blend TK and scientific knowledge). In addition, IRA's have developed tools to help them in their job, such as research monitoring forms, pamphlets, newsletters and information packages. They have also taken part in training in ethics, and scientific process for example. IRA's have also contributed their thoughts and concerns about research to other publications such as the ArcticNet newsletter, and by reporting back to our funders (ArcticNet, Nasivvik and Northern Contaminants Program). Another significant aspect of the IRA position is the increasing the presence of Inuit not only at big meetings but at smaller regional and national committees where decisions are made (NWT Regional Contaminants Committee). Here, IRA's are able to more actively associate with researchers, new partners and other regional organizations that otherwise would not be as accessible to them. All this progress has come in spurts of activity that has been possible because of the past and present partners that we have worked with. There have been several different IRA's and IRA coordinators as well as different supporting bodies throughout the years. Each and everyone contributed their time and resources to make to position functional and recognized today.

## Activities in 2008/2009

### May 2008-April 2009

The Inuvialuit HERC/IRA successfully completed all three of the projects that were proposed for the year. The *IRC Health and Environment Newsletters* (2 editions) were completed and distributed in

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September 2008 and April 2009. The scope of the newsletter was intended for Inuvialuit beneficiaries currently residing in the Inuvialuit Settlement Region, regional organizations and our national partners. The summer newsletter had the greatest distribution and was introduced at the NCP Results workshop in September and brought into many other meetings and events in and outside of the ISR from October onward. The second, winter 2008 newsletter was completed behind schedule and printed in April 2009 and distributed in May 2009. With many other IRA duties throughout the year, the newsletter was completed late. However, this was the longest and most information driven of the newsletters with detailed articles about all the organizations, activities and current research.

The second project that was initiated and still on-going is updating the *Health and Environment page on the IRC website*. This project was not accepted in the NCP budget, but was able to be implemented by funds from ArcticNet and Nasivvik. In December 2008, meetings were set up with IRC's Information Technology and Communications department to discuss the how to use the website software and the range of updates that would be possible with our network in Inuvik. It was agreed by IT and the communications department that the HERC/IRA would be able to update the page on an on-going basis after installing the software on a laptop computer. It was also recommended that the HERC/IRA purchase a new more 'multi-media' friendly computer for this and other communication tasks. In March 2009, the IT department purchased this new equipment for the HERC/IRA. Now that we have the plan for the website and have purchased up to date equipment implementation of the plan will take place during summer 2009 when there is more free time to work on special projects. Peggy Jay, IRC's Communications Officer and Maya Daher, IRC's IT Manager will help in any way that they can. The HERC/IRA has set a deadline of April 30<sup>th</sup> to have final content ready to be inserted online.

The third and final project undertaken by the HERC/IRA was the annual community tour that each employee of the IRC Community Development Division is expected to carry out each year for their programs. The purpose to visit each Inuvialuit community is to consult with people about what community programs are currently being offered

to them. The HERC/IRA held the *1<sup>st</sup> annual IRC Health and Environment Workshop and Feast Community Tour* from November 2008-March 2009. The tour started off in Paulatuk on November 5-7, 2008, and then went on to Sachs Harbour from January 25-27, 2009, Aklavik from February 23-25, Inuvik on March 3<sup>rd</sup>, and Tuktoyaktuk from March 4-5<sup>th</sup>. The final workshop was held in Ulukhaktok from March 16-19, 2009.

Each workshop and feast featured most of the same events, prizes and food with the exception of one or two communities such as Paulatuk that eat fish and geese more often than reindeer and musk ox which we were promoting and serving during the event. On the day of each event, the HERC/IRA worked with the community helpers to ensure that everything would be on time. Secondly, the HERC/IRA set up the venue, unpacked food, prizes, set up tables and handed out information packages (see appendix 1) which were placed on each table. When people arrived they were asked to review their packages, which contained three information sheets on ArcticNet, Nasivvik and Northern Contaminants Program, a 'what do you know about contaminants questionnaire, an 'Education and Training Signup sheet' and a Health and Environment Research Survey'. Then people were asked to fill out as many of the forms as they could and hand them into the HERC/IRA. After people had received the information packages the HERC/IRA proceeded to do a twenty minute presentation called 'IRC Health and Environment Workshop' which was focused on explaining to Inuvialuit what the HERC/IRA does for the community through IRC, what organizations we work with, and how we can help interested Inuvialuit to become involved in research and science in the region. The HERC/IRA then set aside time for a question and answer period of about ten minutes. When all the questions were answered the HERC/IRA gave to go ahead for people to begin serving themselves for dinner. The last thing on the agenda was to go over and review the contaminants questions. Anyone with a correct answer to any of the questions was able to have first choice in prizes. After the review, people were given numbers and their names taken to win the remaining prizes offered to the community. All the remaining food, information packages and prizes were given to community organizations and people/families that

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may have been in need such as the old folk's home and the community corporations for example.

***UPDATE: Other IRA events and projects***

**Inuit Youth Resiliency and Suicide Prevention Working Group**

In August 2008, the HERC/IRA was requested to join the Inuit Youth Resiliency and Suicide Prevention Working Group. The role of the working group was to gather thoughts and ideas from Inuit youth from across Canada and those who are involved in youth programs in the North to have them featured on a live television show to be aired on APTN in May of 2009. The goal was to reach the main audience, which was set by the group, to be youth ages 13-30. In addition to that, a secondary audience was identified and determined unilaterally to be family and immediate family members of the youth in the Inuit communities. The main role of the HERC/IRA was to first provide names and summaries of youth programs offered in the ISR that help to combat suicide, and encourage youth resiliency. The HERC/IRA came up with a list of programs to possibly be featured on the show of which include the Inuvik Youth Centre, FJMC' student mentoring program and the IDC Arctic Youth Expedition Program, but only one was able to be chosen for the actual show. The show is aimed at setting a good example for Inuit youth to show them that there are ways to be an active community player, be positive in life, and accomplish your goals through programs, self made community led programs and other opportunities that they have the option to take advantage of in their respective communities and regions.

**Nasivvik Board of Directors**

In December 2008, the HERC/IRA was appointed to the Nasivvik BOD as an IRA representative, and attended the first BOD meeting in Ottawa, Ontario in April 2009.

**NWT RCC**

The HERC/IRA attended three in-person meetings during the 2008-2009 fiscal years. The first was held in October, the second in December 2008 and the third 'Social and Cultural Review' in January 2009 in Yellowknife, NWT. The HERC also attended a minimum of three teleconferences throughout the year.

**Inuit Health Survey**

In August 2008, The HERC/IRA became involved volunteering time and resources to the second portion of the Inuit Health Survey. This portion of the study was interested in collecting country food samples that will be analyzed alongside the results from human samples taken in summer 2008. During the November IRC board meeting, the HERC/IRA brought this project to the attention of the board and was given permission to begin helping the project coordinator, Linda Van Pelt with this work. Sample collection began in December 2008 and continued until March 2009. The HERC/IRA ensured community participation by first asking regional HTC offices if they would like contribute any samples and be compensated. Three HTC offices were interested in the opportunity including ones in Inuvik, Sachs Harbour and Paulatuk. We later received samples from the Tuktoyaktuk area. Each participant was paid \$50 a sample regardless of the type of samples sent in, be it caribou, fish or birds. The project collection ended in March 31<sup>st</sup>, 2009 but analysis will continue into the summer. All samples were sent to DFO labs in Winnipeg in April 2009.

**Healthy Foods North**

Healthy Food North is a revolutionary new project that first came into the ISR in 2006 to change the eating and exercise habits of people in the North. A program similar to this one was successfully completed in other places around the world that are facing the same preventative health problems like acute obesity and diabetes epidemics. The program promotes the consumption of country foods using the 'family model', where family meals are prepared together and eaten together to create unity in changing the lifestyles of whole entire families, rather than just a few demographics like elders and youth. The program also tries to reduce factors that lead to preventable diseases that are prevalent in northern communities such as diabetes, strokes, cancer and obesity through education and outreach initiatives. The program makes use of 'familiar faces' in the community to speak to people about how to buy healthier food and cut down high sugar intake and processed foods, increase vitamin intake like vitamins and minerals, increase physical activity, and get regular checkups from health care professionals. HFN worked with and received support from various organizations and



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businesses in the ISR to accomplish the programs objectives such as NorthMart, Stanton, IRC, and Gwich'in Tribal Council to get more nutritious foods into the stores at lower competitive prices, and to educate people about why switching a few things in their diet could mean a longer, healthier life in the long run. Overall, this program was very well received in the ISR and people are well aware when they go into the grocery store what is healthier to eat for their families. Stores have volunteered to stock the foods and promote them on their shelves, and since then even they have noticed that there is now a demand for products that never used to leave the shelf, such as whole wheat bread versus white bread.

### **Inuit Nipingit Committee**

The Inuit Nipingit Committee started off this year as NICER (National Inuit Committee of Ethics Research). This committee was brought together by NAHO (National Aboriginal Health Organization) to review the Tri-Council Policy Statement with Inuit and national organizations from across Canada. The Inuvialuit HERC/IRA and Larry Gordon are the representatives for the Western Arctic. To date, we have reviewed the policy statement, began working on communication materials and are now in the process of looking for renewable funding sources to keep the committee going in the future.

### **IRA Review**

An IRA review was conducted by an outside consultant by Inuit Tapiriit Kanatami on behalf of all three funding organizations to find out what lessons have been learned concerning the IRA position. An official meeting was held in Ottawa from April 8-9, 2009 and was attended by all funding bodies, support organizations and most importantly had representation from all IRA's (all Inuit regions in Canada). The report can be obtained by any of the above organizations.

### **What was found?**

Many new lessons have been learned throughout the year about the best ways to communicate, educate, and increase the number of people in the ISR that are interested in science and research. It was found that **at the individual level** people; like to receive regionally relevant information about research, they would like the opportunity

to submit and contribute to publications, they like to see a familiar face hosting the community meetings and like when one or more organizations team up to expand an event (rather than having more smaller events), they like to have traditional style feasts, enjoy when prizes are abundant, they were happy to be aware that IRC had a Health and Environment position aimed at educating community members, and youth are interested in this work, they just need a medium that will allow them to be current and involved (ex. Website, newsletter). It was found that **at the community level**; you must be aware of other competing events happening in the community (bingo, fundraisers, community meetings, weddings, funerals, etc), you must be very clear on what you expect from community helpers and what they can expect from you, you must be able to ensure your supplies (food, prizes, etc.) are sent ahead of time if you are travelling on a scheduled flight in small planes, you must be prepared for unexpected delays and miscommunications, you must always have a plan B to save money or time when in coastal communities, you must make maximum effort to advertise your presence in the community if you expect a good turn out by posting advertisements on (CBC/community radio, television, and post advertisements on local rolling channels, and make them present in high traffic areas in communities such as [Community Corps., Hamlets, HTC's NorthMart, Stanton, Post Office, Co-op]. Lastly, **at the regional level** the region prefers people to hire locally when possible, outsiders should be accustomed to use local businesses to support the community economy, use local translators and facilities such as the handi bus, for example to transport elders and kids to the meetings, always give left over's from the event to people/organizations that may be in need and who would benefit from the donation, visit the IRA in the region so that the region can be made aware of the visit and can prepare ahead of time, and make an effort to stay in the community and visit around with families to get to know them. If any message should be taken from these regional suggestions, I would recommend billeting with a family when possible for the more personal experience.

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### What was concluded?

- Newsletters were a success. It is proposed for 09-10 to have more editions and more opportunities for people in the communities to be involved
- There is a need to create a regional contaminants and research working group consisting of regional organizations that will effectively review and make recommendations on research projects.
- Website will be updated this summer 2009 after deliberations and discussions with IRC
- Proposed community visits will take place in the summer 2009 and beginning of spring 2010

**ArcticNet:** created research monitoring process, organized ARI licenses, assisted Jaime and Pitsey coordinate the BOD meeting in December 2008, presented at the BOD, created an abstract and poster for Arctic Change, attended Arctic Change and Student Day, distributed information on ArcticNet during community visits, submitted annual report, and had correspondence with the following researchers, Keith Levesque, Lisa Losetol, Sonja Ostertag, Emilie Cameron, Andrew Stuhl, Kathleen Paerewyck, Breanne Reinfort, Klaus Gantner, Catherine Gagnon, Mark Andrachuk, Sangita Sharma, Gary Stern, Dawn Bazley, Elie Dolgin, and Robert Powell.

**Nasivvik:** Participated in Nasivvik proposal review, sent out opportunity from Susie 'Be a food scientist for the day', joined Nasivvik BOD as an IRA representative, sent call for complimentary funding, reviewed documents and attended the Nasivvik BOD meeting in Ottawa in April 2009.

**Northern Contaminants Program:** Submitted NCP 2008-2009 Proposal, completed two editions of Health and Environment Newsletter, developed website creation strategy, wrote NCP synopsis 08-09 report, reconnected with NWT RCC and attended most teleconferences and in-person meetings, completed all community visits (6), attended NCP Results Workshop, submitted 2009-2010 proposal, and wrote 09-10 synopsis report.

**Other IRA activities:** Attended Canadian Arctic Summit (May 2008), attended Weathering Uncertainty: Climate Change Symposium Iqaluit (June), had IRA teleconference (May), hosted and chaired IRA teleconference (June), began receiving

all ARI applications (May-July), had correspondence with AHRN for their new PhD and Masters scholarships, got involved with the Inuvialuit database (Ross Goodwin, U of Calgary), attended IRA teleconference (October 2008), joined the Youth Resiliency Working Group, began helping Inuit Health Survey (Linda Van Pelt) and completed sample collection in March, joined NICER (Inuit Nipingit) committee and attended 1<sup>st</sup> in-person meeting, attended climate change forum in Ottawa, submitted to HC a climate change and adaptation proposal, attended the 'Inuit Knowledge Centre' teleconference, submitted abstract for the International Congress on Circumpolar Health which was accepted in March, attended Inuit Nipingit Committee meeting in Ottawa in February, and accepted to be a panelist on the Youth Resiliency TV show on APTN.

### Results

- There is a need to better coordinate with frontline workers involved with research in the ISR through a working group as proposed for the 2009/2010 year
- We need to have more information about sources of funding communities can use to study problems or concerns of importance to them. Funding organizations need to give more time for communities to respond to the calls
- There is a need for a community liaison in each ISR community to take care of Health and Environment work that the HERC cannot always deal with from Inuvik, esp. concerning HTC, CC and Hamlet offices

### Discussion and Conclusions

This work will continue into the 2009-2010 fiscal year. This work is significant because the IRA position is finally being standardized and established. After the IRA review the key challenges and benefits of the IRA position have been clearly laid out and show that the benefits of having a position like this in Inuit regions had helped and will continue to build capacity in the North for research, science and for the younger generation to benefit from the work being done now. There is a need to further establish these processes, via IRA manual, network building and continuation of projects undertaken by Inuit Research Advisors.

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## **Expected Project Completion Date**

April 31, 2009

## **Acknowledgements**

All community helpers, IRC helpers

## **References**

None.

## **Appendix 1**

PDF “Health and Environment Workshop Information Package,” contact the NCP secretariat for more information

# Building Capacity Through Education: Development and Delivery of Community Level and Northern College Module Courses on Contaminants, Wildlife and Health: Year 1

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Cindy Dickson, Council of Yukon First Nations  
Erika Solski, INAC, Iqaluit  
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## **Abstract**

In 2008, the Northern Contaminants Program (NCP) of Indian and Northern Affairs Canada (INAC) initiated and funded a collaborative educational venture with Yukon College and Trent University (Indigenous Environmental Studies Program). In an effort to expand and strengthen the education, communications and outreach capacity of the NCP, two Yukon College paraprofessional courses were developed and delivered through

## **Résumé**

En 2008, le Programme de lutte contre les contaminants dans le Nord (PLCN) d'Affaires indiennes et du Nord Canada (AINC) a lancé et financé un projet d'éducation concerté avec le Collège du Yukon et l'Université Trent (programme d'études environnementales autochtones). Pour élargir et renforcer le volet éducation, communications et diffusion des connaissances du PLCN, deux cours paraprofessionnels du Collège du Yukon ont été

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distributed learning (distance education). A total of 19 students from 7 Yukon communities participated in the two courses through videoconference. One course was on Northern Contaminants and Wildlife and one on Northern Contaminants and Community Health. In addition, a university transfer level module on contaminants was developed as part of an existing environmental science course at Yukon College. The module and paraprofessional courses were designed to be transferrable to the other 2 northern colleges (with appropriate regional modifications) – Aurora College (N.W.T.) and Arctic College (Nunavut). This project will continue in 2009/10.

## Key Messages

- Yukon College and Trent University have partnered to establish modules of course material for the existing Renewable Resources diploma program at Yukon College as well as two courses at the paraprofessional level for front line people working in areas that require a basic knowledge of northern contaminants (e.g. existing wildlife managers, lands and environment representatives, Community Health Workers, etc).
- This initial partnership has expanded to include the other two northern colleges (Aurora and Nunavut Arctic College). With this pan-northern scope and distance delivery through videoconference and internet (interactive course websites), the team hope to develop sustainable mechanisms through which to transfer and teach important aspects of environmental and environmental health sciences incorporating the latest Northern Contaminants Program generated research and information.

mis au point et offerts par apprentissage à distance. Au total, 19 étudiants de sept collectivités du Yukon ont suivi ces deux cours par vidéoconférence. L'un des cours portait sur les répercussions des contaminants sur la faune septentrionale et l'autre, sur les répercussions des contaminants sur la santé des collectivités septentrionales. Qui plus est, un module de passage à l'université traitant des contaminants a été élaboré dans le cadre du cours sur les sciences de l'environnement déjà donné au Collège du Yukon. Ce module de même que les cours paraprofessionnels ont été conçus pour être transférables aux deux autres collèges du Nord (sous réserve de modifications pertinentes compte tenu de la région) – soit le Collège Aurora (T. N.-O.) et le Collège de l'Arctique du Nunavut. Ce projet se poursuivra en 2009-2010.

## Messages clés

- Le Collège du Yukon et l'Université Trent ont élaboré, en partenariat, des modules de cours faisant partie du programme menant à un diplôme et portant sur les ressources renouvelables du Collège du Yukon ainsi que deux cours paraprofessionnels destinés aux intervenants de première ligne qui doivent posséder des connaissances de base sur les contaminants présents dans le Nord, compte tenu du secteur dans lequel ils travaillent (p. ex. gestionnaires de la faune, représentants territoriaux ou environnementaux, agents de santé communautaire, etc.).
- Le partenariat initial s'est élargi et les deux autres collèges du Nord s'y sont joints (le Collège Aurora et le Collège de l'Arctique du Nunavut). Grâce à l'enseignement à distance par vidéoconférence et Internet (sites Web des cours en mode interactif) offert dans les régions septentrionales, l'équipe espère élaborer des méthodes viables de transfert et d'enseignement des aspects importants des sciences environnementales et des sciences de la santé environnementale intégrant la recherche et l'information la plus récente découlant du Programme de lutte contre les contaminants dans le Nord.

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## Objectives

- Develop and pilot the delivery of a module that incorporates recent NCP research for both Yukon College's Renewable Resources Program and for Trent University's Indigenous Environmental Studies Program;
- Develop and pilot the delivery of two paraprofessional courses that provide information on northern contaminants for individuals residing in rural Yukon who are in the health or environment fields;
- Through engaging a Steering Committee comprised of representatives from the three Northern Colleges, ensure development of courses in Year 1 of the project that will allow for adoption and implementation in other regions in Year 2 at Aurora College and Nunavut Arctic College.

## Introduction

Since its inception, the Education and Communications sub-program of the Northern Contaminants Program has endeavored to find ways to effectively communicate the findings of northern contaminants research and build capacity for the understanding of this information in northern communities (Furgal et al., 2003). Currently, the three northern colleges (Yukon, Aurora, and Nunavut Arctic College) have some form of Environmental Technology or Renewable Resources program through which students are educated and very often go on to fill environment related positions in the region afterwards. The Northern Contaminants Program has yet to formally connect to and support the training of these individuals who are already motivated and interested in the environmental and environmental health fields at this critical time in their higher education prior to seeking employment. Further, with the rapid progress being made in the implementation of northern information technology infrastructure, the feasibility of communicating, exchanging knowledge and learning 'at distance' in the "comfort of one's own community" rather than having to travel to a workshop or college, or University in the south is quickly increasing. As one example, Yukon College has now delivered a "Math for Water Operators" training program in many of the Territory's First

Nations Communities through an online distributed learning format that is now present in these communities. They are now looking for other opportunities to explore and develop 'distributed learning' formats for a diversity of subject matter of interest to students, and professionals and community residents working throughout the Territory.

While changes occur in the northern workplace (e.g. growth in environmental and research oriented positions with resurgence in northern science and development) the NCP continues to generate significant amounts of world-class science to advance our understanding of contaminants, pathways, trends and their effects in northern ecosystems and on northern populations. Some of this leading edge science has made its way, albeit passively (i.e. without a focused specific effort of the program in many cases) into the classrooms of Universities and some high schools, often in the south. However, little sustained and concerted effort has been made to translate this material into learning modules for those that are perhaps some of the most motivated and interested to learn about this information in the north: northern college students and community workers.

It is for these reasons that Yukon College and Trent University have partnered to establish module course material for the existing Renewable Resources program at Yukon College as well as two courses at the paraprofessional level for front line people working in areas that require a basic knowledge of northern contaminants (e.g. existing wildlife managers, lands and environment representatives, CHRs, etc). In partnership with the other northern colleges for the adaptation and expanded use of this material in the two other Territories, this effort introduces a more sustainable mechanism through which to transfer and teach important aspects of environmental and environmental health sciences using the latest northern contaminants research and information.

The partnership between the Northern Colleges and Trent University is critical to provide the foundation not only for course development and delivery but also to offer the opportunity of University level transfer accreditation to students taking northern college courses as well as those then wanting to pursue University level education.



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## Activities in 2008/2009

This past year two paraprofessional courses were prepared and pilot tested in the Yukon. The first course, on wildlife and contaminants ran in the fall of 2008 and included 15 students from 7 different communities. The course lasted 7 weeks and received excellent feedback from students. As the sample testimonials show students very much appreciated the course and are requesting more research and learning opportunities of this kind.

*"Wow! There is so much to love about this course!... I would love it if we got to do some testing on our water, soils, plants, animals and even in our home and ourselves....I now want to learn more and teach others and help my community members....I would love my classmates to help the community to help the animals and all living things....I would love a summer job as a student to start making changes or assisting with contaminants testing and monitoring."*

Christine Blanchard, Dawson City

*"In our class, I liked the opportunity for active participation....I enjoyed doing the assignments... there was lots of good information....Enjoyed the classes but also reviewing the PowerPoint presentations on the website afterwards...the video technology was new to me but now I think it is amazing and so convenient that I didn't have to go to Whitehorse....The material we covered was useful and brought an awareness of the seriousness of the global problem of dealing with contaminants....The most valuable aspect of the course to me was becoming more aware of the contaminants issue and the stakeholders involved. I didn't realize the amount of contaminants in the north; now, I can do my part as an individual by continuing to go "green" and be more proactive in my community....I believe there was a good balance of information...it was good to have guest speakers come in \*\*"*

Shirley Lord, Tagish

*\*\*Elder Randal Tetlich from Old Crow, Mary Gamberg and Lewis Rifkind (Yukon Conservation Society) accepted to be guest speakers in this course in fall 2008*

*"What I liked most about the class was local stories and things that hit close to home....I plan to continue with the next course offered on contaminants and community health"*

Dawn Baker, Carmacks

The course inspired many of the students to want to learn more about the topic and the research taking place on their land. Below is a poem written by one student, Shirley Rose, about her perspectives on the issue, as inspired by her participation in the course:

### ***Cry in the Land***

*Listen closely to what we say,  
For generations past and even today.  
People of the north, Indigenous to this land,  
We've lived close to nature sharing all that we can.  
Affected from industry and politicians from afar,  
Forever touched, forever changed Who we really are...  
Did we ask for this destruction that so freely came?  
No, we gave only our land in exchange for disease  
and pain, Now, please hear, our Cry in the Land!*

*Our Ancestors have told us time and again,  
To look after the plants, the wildlife and the land.  
Are we to continue in silence, allow companies to gain  
Their glory and riches, their shallow right to fame.  
We have rallied here for the future looks so bleak,  
To call on our Ancestors to help us because we  
really feel weak.*

*We have no powers to control the pollution coming  
our way  
Are we to remain defenseless and defeated, no,  
let's stand united today  
Please hear our Cry in the land!*

*Let's work together to protect, preserve and enhance  
Our global village that truly is not here by chance.  
Let's take the P.O.P.'s, the metals, the radionuclides,  
Increase the awareness, stop the damage and  
destruction worldwide.*

*Let's think of future generations of all in the  
Circle of life, living in harmony with the four  
leggeds, the winged ones, all that swim in the sea,  
Creepy crawlies, the plants and two-legged and all  
that we see.*

*We must never take for granted the air that we breathe*

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*The lakes & streams we fish in, the plant medicines we receive.*

*Please hear our Cry in the land!*

*We call on Healers of long ago to circle round and show us the way,  
To live without polluting each and every day,  
To walk on Mother Earth in a Sacred way,  
To revere and  
respect Her for all Her gifts She's given us today.  
Renew our strength and help us understand.*

*We cannot continue to abuse the sacredness of this land,  
Tools we must gather, knowledge and wisdom to stop unhealthy attitudes of today,  
Help us say no to the riches that consume us in a negative way.  
Please hear our Cry in the land!*

*The pollutants are issues at hand that continue to grow,  
They destroy the environment and Circle of life as we know.  
Abolish the contaminants that want to destroy the family of man.  
Together today, for our children, our future,  
Let's take a stand.*

*Written by: Shirley Lord – Gwich'in Nation,  
Tagish, Yukon (Student, Contaminants and  
Wildlife Paraprofessional Course, Fall 2008)*

The Community Health and Contaminants paraprofessional course was offered in March 2009 and, although enrollment was lower (4 students in 3 communities), the feedback from the course was similarly very positive.

As well, in Year 1 of this project, the structure and outline for the college level course module (to be proposed as the first component of the transfer credit arrangements being established between Yukon College and Trent University through the creation of an articulation agreement) was completed.

Additionally, one steering committee meeting was held in Yellowknife in conjunction with the NCP Annual results Workshop and representatives from each of the northern colleges and contaminants committees were in attendance.

## Discussion and Conclusions

The first year of this new project was very successful. The project has now been renewed for Year 2 during which the following is planned:

- Expand the existing half courses (7 weeks) at the paraprofessional level into full courses (14 weeks) and deliver them in 09-10 in the Yukon;
- Complete the adaptation of existing paraprofessional material and development of other needed material for courses to be offered (if possible) in the other two other territorial regions in 2010-11;
- Develop a proposal in association with the Yukon paraprofessional courses to submit to the NCP in 2010-11 for the expansion of these courses to include contaminants project experiential learning opportunities for students in cooperation with researchers working in the Yukon under the NCP;
- Finalize the Yukon College – Trent University articulation agreement;
- Complete the University Transfer credit module on contaminants and wildlife and pilot this module within the Yukon College Renewable Resources Program;
- Explore the possibility of adding a college level health and contaminants module within the existing health education program at Yukon College (we are not currently requesting funding to develop and pilot this module at this time as arrangements are being negotiated with the current health instructor etc; if negotiations are finalized this summer a mid-year funding request will be submitted to the NCP, o funding will be requested for this in a future funding year of the program);
- Explore, via Steering Committee meetings, adaptation and adoption of the modules and delivery by the other northern colleges in 2010-11.

The current courses are fully accredited by Yukon College and developed and delivered as distance education courses using distance technology such as video conferencing and interactive web technology.

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Currently they include:

- One course, Contaminants and Human Health, is specifically designed to introduce the fundamentals of northern contaminants while exploring the nature of exposure and effects of northern contaminants on human health. An exploration of fundamentals in Arctic human health provides some context to the effects of contaminants on human health. This course is targeted for Community Health Representatives and others who are working as rural front line community health workers;
- One course, Contaminants and Wildlife, is specifically designed to explore the levels, trends and possible effects of northern contaminants on local wildlife populations. An exploration of the basics of wildlife health and ecology and possible impacts of other forms of environmental change (e.g. climate change) provides some context for understanding the effects of contaminants on wildlife health. This course is specifically targeted for individuals working as front line workers in rural land management departments. This course includes a full-day on-site lab component.

College modules are being developed with the intention that they become transfer credit courses for those individuals wanting to apply these credits to further education at Trent University in one of the applicable programs (Environmental Resource Studies/ Science; Indigenous Environmental Studies/ Science). By involving representatives from Aurora College and Nunavut Arctic College on the Steering Committee, we are establishing a northern college partnership that has not existed among these individuals previously on this topic and will provide the opportunity for future collaborations on curriculum development and knowledge exchange across the colleges of the north as well as between colleges and supporting institutions to the south.

Yukon College will deliver the wildlife module as a core component of the Renewable Resources program and will deliver an expanded (full course version) of the two paraprofessional courses via distributed learning in 2010-11 to those planning to work or already working in either the health area or the lands and environment area or to other interested community members.

To ensure that this initiative remains relevant to individuals across the north, the steering committee will continue work into the second year of the project and involve three NCP Regional Contaminant Committee (RCC) co-chairs, representatives from Yukon College, Aurora College, Nunavut Arctic College, and Trent University. This committee will continue to advise and oversee the program and will explore opportunities where transfer agreements can be made among the three northern colleges to strengthen opportunities for communicating results stemming from northern contaminant research.

This is an ongoing project that will be complete, in its current form, in 2010.

## References

- Furgal, C., Kalhok, S., Loring, E., Smith, S. 2003. Knowledge in Action: Northern Contaminants Program Structures, Processes and Products. Canadian Arctic Contaminants Assessment Report II. Indian and Northern Affairs Canada, Ottawa, ON. ISBN: 0-662-33470-1.

# Nunatsiavut Inuit Research Advisor

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## ◆ **Project leader(s):**

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Mary Denniston, Northern Contaminants Researcher, NG

## **Abstract**

The Inuit Research Advisor (IRA) program continues to serve as the first step in a more coordinated approach to community involvement and coordination of Arctic science in Nunatsiavut. Together with IRAs in the other Inuit regions of Canada, the Nunatsiavut IRA works towards achieving a new way of knowledge sharing and engagement of Inuit in Arctic science in the region. In addition to NCP support, the program is co-funded by ArcticNet and the Nasivvik Centre for Inuit Health and Changing Environments.

## **Key Messages**

- In 2008-09 the IRA undertook various tasks in liaison with the Northern Contaminants Program (NCP), ArcticNet, Nasivvik Centre, and Nunatsiavut Government (NG) in the areas of research promotion and coordination, public education and information.
- The IRA has also served as liaison for partners such as Inuit Tapirit Kanatami (ITK), Inuit Circumpolar Council (ICC), Nunatsiavut

## **Résumé**

Le conseiller en recherche inuite (CRI) continue à faire office de première étape de l'approche mieux coordonnée ayant pour objectif de favoriser la participation des collectivités et de coordonner les sciences de l'Arctique au Nunatsiavut. De concert avec les CRI des autres régions inuites du Canada, le CRI du Nunatsiavut s'efforce de promouvoir une nouvelle façon de diffuser les connaissances et de mobiliser les Inuits en ce qui touche aux sciences de l'Arctique dans la région. Le financement des activités est conjointement assuré par le Programme de lutte contre les contaminants dans le Nord (PLCN), ArcticNet et le Centre Nasivvik pour la santé des Inuits et les changements environnementaux.

## **Messages clés**

- En 2008-2009, le CRI a exécuté diverses tâches en collaboration avec le PLCN, ArcticNet, le Centre Nasivvik et le gouvernement du Nunatsiavut (GN) dans les secteurs de la promotion et de la coordination de la recherche, de l'éducation du public et de l'information.
- Le CRI assure également la liaison avec des partenaires comme Inuit Tapirit Kanatami (ITK), le Conseil circumpolaire inuit (CCI),

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Nuluak, Nunatsiavut Inuit Community Governments, International Polar Year (IPY), researchers, students, and other organizations.

- The IRA oversaw the management of the Nunatsiavut Government Research Office, serving as the first point of contact for all researchers conducting work in Nunatsiavut and requiring contact with or assistance from the Nunatsiavut Government. The IRA is the Chair and administrator of the Nunatsiavut Research Advisory Committee;
- The IRA served as liaison, contact and assistant to research projects taking place in Nunatsiavut. This assistance ranged from linking the researchers with appropriate individuals and/or organizations such as NG departments and Inuit Community Governments in Nunatsiavut to providing input on research proposals and plans.
- The IRA is a member of the newly formed NCP funded Nunatsiavut Contaminants Committee, now called the Nunatsiavut Health and Environment Review Committee (NHERC).
- The IRA has undertaken diverse tasks for the host organization Nunatsiavut Government (NG) ranging from attending NG workshops, researcher workshops and collaborating with many researchers and organizations.

Nunatsiavut Nuluak, les administrations des collectivités inuites du Nunatsiavut, l'Année polaire internationale (API), les chercheurs, les étudiants et d'autres organismes.

- Le CRI a supervisé la gestion du bureau de la recherche du gouvernement du Nunatsiavut, en faisant office de point de contact initial pour tous les chercheurs effectuant de la recherche au Nunatsiavut et ayant besoin de communiquer avec le gouvernement du Nunatsiavut ou d'obtenir son aide. Le CRI est le président et l'administrateur du comité consultatif de la recherche du Nunatsiavut.
- Le CRI a joué le rôle d'agent de liaison, de contact et d'assistant pour ce qui est des projets de recherche menés au Nunatsiavut. Entre autres, il a mis les chercheurs en contact avec les personnes ou organisations pertinentes, par exemple les ministères du GN et les administrations des collectivités inuites du Nunatsiavut, et il a fait des suggestions quant aux propositions et plans de recherche.
- Le CRI fait partie du Comité des contaminants du Nunatsiavut, nouveau comité financé par le PLCN et désormais appelé comité d'examen de la santé et de l'environnement du Nunatsiavut (*Nunatsiavut Health and Environment Review Committee* ou NHERC).
- Le CRI a accompli diverses tâches pour l'organisme d'accueil, le gouvernement du Nunatsiavut (GN), entre autres, il a assisté à des ateliers donnés par le GN ou des chercheurs et a collaboré avec un grand nombre de chercheurs et d'organismes.

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## Objectives

- Provide liaison support for and promote research in Nunatsiavut
- Promote more community-based research in the region
- Assist in the development of local capacity for research in Nunatsiavut
- Provide information regarding research in Nunatsiavut and opportunities for local involvement

- Liaise with national organizations and other Inuit regions in matters related to Arctic science and research

## Activities in 2008/2009

- Managed the Nunatsiavut Government Research Office and served as Chair to the NG Research Advisory Committee, making contact with virtually all researchers, students and organizations visiting the Labrador Inuit Land Claim Area.



- Participated in Nunatsiavut Contaminants Committee (NCC) now the Nunatsiavut Health and Environment Review Committee (NHERC) meetings on March 13-14 (Goose Bay) Sept 19 (teleconference), November 13 (Hopedale) and March 10, 2009 (Postville). Undertook socio-cultural review of NCP proposals in March 2009.
- Attended annual NCP Results Workshop, Yellowknife, September 23-25 2008
- Attended ArcticNet – Arctic Change 2008 Conference, December 9-12 2008, Quebec City.
- Participated in preparation of a regional science meeting to promote and provide information on NCP, ArcticNet, Nasivvik and other research initiatives in Nunatsiavut.
- Participated in teleconferences with other regional IRAs
- Maintained the Nunatsiavut Research list recording contact with all researchers visiting the region. Provided this list to all funding organizations.
- Kept Nunatsiavut Government Research Advisory Committee informed of all NCP, ArcticNet and Nasivvik activities and opportunities
- Promoted NCP, ArcticNet, Nasivvik activities and opportunities at the Nunatsiavut community and regional level through word of mouth and the distribution of promotional materials
- Assisted in the preparation of the Nunatsiavut NCP Newsletter (Avativut)
- Participated in IRA Program Review.
- Joined the National Inuit Committee on Ethics and Research (NICER), attended the NICER meeting and the Pre-Tri council ethics meeting in Ottawa February 2009. Provided input into the design for a poster on research ethics.
- Assisted in the development of a proposal to the Arctic Research Infrastructure Fund (ARIF, INAC) for the establishment of a Nunatsiavut Research Centre in Nain.
- See attached Research Contact list for all contacts the IRA made with researchers during the 2008-09 fiscal year.

## Results

The IRA program in Nunatsiavut continues to provide a coordinated process by which Inuit and researchers can become connected for more effective and meaningful research in the disciplines of environmental science, contaminants and human health.

## Expected Project Completion Date

This is an ongoing project.

## Attachment

2008-09 Nunatsiavut IRA Research Contact List

## 2008-09 Nunatsiavut IRA Research Contact List

1. Julie Bull, Dr. Malcolm Murray, University of Prince Edward Island, Dr. Fern Brunger, Memorial University of Newfoundland, January 2007–May 2008, Interpreting the Tri-Council Policy Statement in Community- Based Research.
2. Beatrix Arendt, PhD Candidate, July and August of 2008, Excavations on Anniowaktok Island near Hopedale.
3. Jacquie Bastick, Environmental Sciences Group, July, 2008–August 2008, Establishing Marine Ecological Integrity Monitoring Measures for Nachvak and Saglek Fiords, Northern Labrador.
4. Dr. Trevor Bell, Department of Geography, Memorial University of Newfoundland, July, 2008–August, 2008; July, 2009–August, 2009, Recent Glacier Change in the Torngat Mountains, northern Labrador.
5. Sam Bentley (lead investigator) and Elisabeth Kahlmeyer (student), Ken Reimer, Tom Sheldon, Environmental Sciences Group, July 2008–August 2011, Marine Records of Riverine Water and Sediment Discharge in Fjords of Nunatsiavut.
6. Culp, Joseph M., Environment Canada, Water S&T Directorate, July–August 2008, The ARCTIC Freshwater BIODiversity Research and Assessment NETwork: ARCTIC-BIONET, Saglek and Nachvak Fiord in the Torngat Mountains National Park and the Koroc River watershed in Nunavik.



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7. J. Brian Dempson, Milton Shears, Fisheries and Oceans Canada, June 23–September, 2008; Nain, Climate Variability & Change Effects on Charr in the Arctic, Voisey's Bay – Anaktalik Bay area, Nain – Tikkoatokak area, Okak area, Hebron – Saglek – Nachvak area.
  8. Vincent Brodeur, M.Sc., Wildlife Biologist, Ministère des Ressources naturelles et de la Faune Direction de l'aménagement de la faune du Nord-du-Québec, Joëlle Taillon, bio., Ph. D. candidate, Département de Biologie, Université Laval, Sampling and capture of caribou in the Nunatsiavut portion of the George-River caribou calving ground in June 2008, Nord-du-Québec caribou (*Rangifer tarandus*) management plan (2004-2010).
  9. David Bruce, Director, Rural and Small Town Programme, Mount Allison University, May 1, 2008 and July 31, 2008, A Study of the Atlantic Aboriginal Post-Secondary Labour Force;
  10. Gérald Chaput, Biologist, Fisheries and Oceans Canada, August 10 and 23, 2008, Pelagic ecosystem survey of the northwest Atlantic with emphasis on Atlantic salmon.
  11. Professor Kirk Dombrowski, Associate Professor of Anthropology, John Jay College, CUNY, and CUNY Graduate School, Nain, June 2008–August 2009, Resource Development and Emerging Informal Structures in Northern Labrador.
  12. Dr. Grace Egeland, McGill University, The Inuit Health Survey: Health in Transition and Resiliency.
  13. Dr. Marlene Evans, Environment Canada, Nain, July 15–September 15, 2008, The project will continue in 2009/2010 with a graduate student, Temporal trends and spatial variations in persistent organic pollutants and metals in sea-run char from the Canadian Arctic.
  14. Laura Fleming, Department of Geography, University of Guelph, Climate Change Adaptation Management of Wildlife Resources, Hopedale, February 17<sup>th</sup>, 2008 – June, 2008, Nain.
  15. Gerlis Fugmann, Nain July 2 –August 1, 2008; Sept 2009, Circumpolar Concepts of Autonomy. Autochthon Development Perspectives of Inuit Societies. (Preliminary title);
  16. Dr. Christopher M. Furgal, Department of Indigenous Studies/Environmental Resource Studies, Trent University, Susie Bernier, Nasivvik Centre, Philippe Max Rouja, January 2008, Integrated Research Program on Arctic Marine Fat and Lipids: The Urqsuk Program.
  17. Scott Gilliland, Canadian Wildlife Service, 10-12 days between 25 July and 10 August 2008, Ecology of Surf Scoters Moulting in Northern Labrador;
  18. Dr. Scott McEwen, University of Guelph, Dr. Victoria Edge, Public Health Agency of Canada, Sherilee Harper, University of Guelph, Nain and Makkovik. September 2007-2008, An investigation of potential impacts of climate change on surface drinking water quality and infectious gastrointestinal illnesses in Nunatsiavut, Canada;
  19. Hermanutz, Luise, A.; Dept. of Biology, Memorial University of Newfoundland, July 26, 2008 and ongoing for 2-3 years, CiCAT – Climate Change Impacts on Canadian Arctic Tundra Ecosystems: Interdisciplinary and Multi-scale Assessments – ArcticNet – Impacts of vegetation change in the Canadian Arctic: local and regional assessments;
  20. Sylvain Leblanc, Canada Centre for Remote Sensing, Natural Resources Canada, 2008/08/09 – 2008/08/21, Map and change detection in Canada's arctic tundra ecosystems;
  21. Rebecca Jeffery, Wildlife Division, NL Government, June 9-13<sup>th</sup>, 2008, George River Calf Work, Hebron Fjord, LISA Lands;
  22. Rebecca Jeffery, Wildlife Division, NL Government, April 9, 2008, June 19-20<sup>th</sup>, 2008, Mealy Mountain Caribou Calving Survey – George's Island Classification, LIL;
  23. Rebecca Jeffery, Wildlife Division, NL Government, March 17-28, 2008, Mealy Mountain Caribou Monitoring Project – George's Island Re-collaring;
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24. Andria Jones DVM, PhD, 2008 – Engaging Northern communities in the monitoring of country food safety, Nain and first sampling phase begin in Hopedale,
  25. Colin Jones, Wildlife Biologist, LGL Limited, 27 May, 2008, Harlequin Duck Population Surveys in Labrador and northern Newfoundland
  26. Dr. Colin Laroque, Mariana Trindade. MSc., Mount Allison University, Nain/Okak, Cabot Lake, Anaktalik, Tasisuak Lake, Kingurutik Lake, July 2008, Creating an extensive tree-ring sampling network in Labrador using living trees;
  27. Ken Reimer, Tom Sheldon, Environmental Sciences Group and Marina Biasutti, Nunatsiavut Government, August 2008, Foraging behaviour of ringed seals in Saglek Bay, Labrador.
  28. Dr. Suzanne Mills, Dr. Angela Robinson, Sir Wilfred Grenfell College, Memorial University, April 2009 – March 2010, Effecting change in long-standing institutions: Aboriginal employment and labour unions in northern resource development projects,
  29. Nunatsiavut Nuluak (NN) Project (ArcticNet project 3.7). Project co-leads Nunatsiavut Government (NG), and the Environmental Sciences Group (ESG).
  30. Aliya Pardhan, University of Guelph, An Investigation of the Potential Impacts of Climate Change on Infectious Gastroenteritis in the Canadian Arctic.
  31. Peter Phillips, Department of Political Studies, 2007-2008, Translating Science: Genomics and Health Systems, Applying PPP Models to Manage Traditional Knowledge.
  32. Reinhard Pienitz, Thomas Richerol, Laval University, Ken Reimer, Tom Sheldon Environmental Sciences Group, Marina Biasutti, Nunatsiavut Government, Saglek Bay, July-August 2008, Paleoceanographic and paleolimnologic study of the impacts of natural and anthropogenic perturbations on fiord and lake ecosystems in northern Labrador.
  33. J. Bruce Pollard, Gamebird Management Biologist, Environment Canada/Canadian Wildlife Service, July 2008, North Atlantic Population Canada Goose Banding.
  34. Ken Reimer, Tom Sheldon, Tanya Brown, Environmental Sciences Group, Marina Biasutti-Brown, Nunatsiavut Government, July 15 to October 15, 2008; Stable isotope and fatty acid food web model of an Arctic coastal nearshore environment: evaluating the effects of climate change and modernization on trophic transfer and bioaccumulation in northern Labrador.
  35. Ken Reimer, Tom Sheldon, Tanya Brown, Environmental Sciences Group, Marina Biasutti-Brown, NG, July to October, 2008; Variability of primary production, zooplankton, and oceanographic conditions across a latitudinal gradient in Northern Labrador.
  36. Lori Ann Roness, August 2008–August 2009, A Study Examining Partnership Arrangements between Aboriginal and Non-Aboriginal Businesses.
  37. Lori Ann Roness, June 18, 2008. 'A Study Assessing the Effectiveness of Labour Force Participation Strategies'. The expected completion date is June 19, 2009.
  38. Marina Sherkina-Lieber, Ph.D. student, Dept. of Linguistics, University of Toronto, Receptive knowledge of Inuttitut, Approximately 3 weeks in late April/May 2008;
  39. Steven Siciliano, University of Saskatchewan, Speciation and In vitro Bioaccessibility of Mercury from Traditional Country foods, Start Date: Jan 2008, End Date: Sept 2009;
  40. Dr. Chris Southcott, Lakehead University, Mapping the Social Economy in Northern Canada, February 2008 through August 2008;
  41. Joan O'Brien, Department of Fisheries and Oceans Canada, Oceans Division, Community-Based Coastal Resource Inventory for Northern Labrador, 2007/08 to fiscal year 2009/10;
  42. Carrie Gillon, Memorial University, The Syntax and Semantics of Bare Nouns in Inuttitut;

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43. Peter Whitridge & James Woollett, Memorial University of Newfoundland, Torngat Mountains National Park Reserve of Canada, Komaktorvik Archaeology Project, July and August 2008.
  44. Valerie Malazdrewicz, Health Canada, Inuit Oral Health Survey, Nov. 10-21 2008.
  45. Catharyn Andersen, Aspects of Behaviour relating to Inuttitut in Nain, Nunatsiavut, May 2009.
  46. Marc Choquette, Healthy Living in Schools and Substance Abuse among Youth (Part 2), Nain and Postville.
  47. Vincent Brodeur, Request to NL Government from Quebec for sampling of the George River Herd, fall 2008.
  48. Yang Mao and Y. Anita Li, MD, MSc, The Prevalence of Human Papillomavirus and Its Impact on Cervical Dysplasia in Labrador, Happy Valley-Goose Bay, Nain, Hopedale, North West River, two years from Nov 2008–Dec 2009.
  49. Michael Ungar, Ph.D. and Linda Liebenberg, Ph.D., Dalhousie University, Pathways to Resilience Research Program, Inuit Communities of Labrador, five-year program.
  50. Maria M'Lot, Research Associate, Centre for Indigenous Environmental Resources, Remote Communities Quality of Life Project, Nain and/or Hopedale community workshops September 2008–March 2009.
  51. Millie Spence, Port au Choix National Historic Site, Parks Canada; Inuit perspective on Reconstructing Dorset house structure, 3 days Nain March 2009.
  52. Paul Pigott, Master of Arts program at Memorial University, sikkuginnait Kanuittusuatuinnanik Inuttut ice typology, Contact Inuttut speakers in Nain, Hopedale, Makkovik and Rigolet, April 4-18 2009, Summer 2009, freeze-up time 2009, ongoing until final results winter 2010.
  53. Peter Whitridge, Memorial University of Newfoundland and James Woollett, Université Laval, Komaktorvik Archaeology Project, 2007 and 2008.
  54. James Woollett, Université Laval and Peter Whitridge, Memorial University of Newfoundland, Landscape History of Inuit Settlement in Nain Bay, July–Aug, 2008.
  55. Mary Gamberg, Gamberg Consulting, Whitehorse, Lorna Skinner, Contaminants and Remediation Directorate, Indian and Northern Affairs Canada, Mitch Campbell, Department of Environment, Government of Nunavut, Arctic Caribou and Moose Contaminant Monitoring Program 2007/8.
  56. Isabella Pain, Voisey's Bay Nickel Company. Socio Economic Monitoring for Impacts of the Voisey's Bay Project on the Inuit Communities in Nunatsiavut and on the Innu Communities, Nain, Hopedale, Postville, Makkovik, and Rigolet, start in fall 2006 and will be ongoing over the next 2 years.
  57. Daniel Martin, Ph.D., Cathleen Knotsch, M.A., Benoit Levesque, MD, "Drinking Water Quality and Climate Change in Labrador: A Pilot Project for Two Inuit Communities", was continued to include the other Inuit Communities in Nunatsiavut – "Drinking water quality in Nunatsiavut and Nunavik: adaptation strategies in an evolving climate change context".

# Beluga Communication Package for Inuvialuit Settlement Region (ISR): Everything you wanted to know about beluga but were afraid to ask

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## ◆ Project team members:

Rebecca Pokiak, Tuktoyaktuk NT, Nellie Pokiak, Tuktoyaktuk NT, Shelia Nasogaluak, Resource Manager (FJMC), Peter Ross (DFO), Gary Stern (DFO/U of Manitoba), Sonja Ostertag (UNBC), Stephen Raverty (BC animal health), Steven Ferguson (DFO), Ole Nielsen (DFO), Gregg Tomy (DFO), Robie Macdonald (DFO), Richard Binder (JS/NWTRCC), Laurie Chan (UNBC), Scot Nickels (ITK).

## Abstract

The traditional hunt for belugas is an important cultural and nutritional event in the Inuvialuit Settlement Region (ISR). The beluga whale remains a vital part in the healthy diet for many Inuvialuit. Therefore it is critical that any scientific information communicated back to Inuvialuit is done so in a cultural appropriate and contextual manner to reflect overall picture of beluga health. Beluga research is being conducted on many fronts to understand the health of the beluga population

## Résumé

La chasse traditionnelle des bélugas constitue un important événement culturel et nutritionnel dans la région désignée des Inuvialuit (RDI). Le béluga demeure une composante vitale d'un régime alimentaire sain pour un grand nombre d'Inuvialuit. Par conséquent, il est essentiel que toute information scientifique communiquée aux Inuvialuit soit transmise dans le respect de leur culture et du contexte afin de présenter une vue d'ensemble de la santé des bélugas. Les recherches sur les bélugas

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and the health of the ocean they inhabit. Local knowledge of the beluga is being gathered and shared in the traditional Inuvialuit ways. Bringing these two knowledge systems together in the form of a contextualized communication package is the goal of this study. Foremost to the success of this project is the need to train a local youth to help bridge the two knowledge systems and develop and communicate beluga information back to the community. Our program also addresses the need to invest in the capacity of communities to be knowledgeable in both local views of beluga health and scientific findings. Further, this would allow individuals to receive and understand this information within their community when needed, rather wait for a scientist to report back. This partnership approach in communication and research will make research in the arctic more responsible to the needs and issues of Inuit communities.

## Key Messages

- Developing local capacity to communicate scientific results.
- Combining local knowledge and observations into scientific results
- Contextualizing all scientific information about the overall health of beluga whales into one communication package

couvrent de nombreux aspects afin de mieux comprendre l'état de santé de la population des bélugas ainsi que de l'océan qui constitue leur habitat. Par ailleurs, nous recueillons les connaissances de la population locale sur les bélugas qui sont diffusées selon les traditions des Inuvialuit. L'objectif de la présente étude consiste à réunir ces deux systèmes de connaissances sous forme d'une trousse d'information contextualisée. Pour assurer le succès de ce projet, il est essentiel de former la jeunesse locale afin de jeter un pont entre les deux systèmes de connaissances et de préparer l'information sur les bélugas pour la communiquer à la collectivité. Notre programme tient également compte de la nécessité d'investir dans la capacité des collectivités afin d'être mieux informés sur les points de vue de la population locale et sur les conclusions scientifiques concernant la santé des bélugas. De plus, cela permettrait aux personnes de recevoir et de comprendre cette information au sein de leur collectivité selon leurs besoins au lieu de devoir attendre les résultats des rapports scientifiques. Cette forme de partenariat en matière de communication et de recherche permettra de s'assurer que la recherche dans l'Arctique répond mieux aux besoins et aux enjeux des collectivités inuit.

## Messages clés

- Créer une capacité locale dans le but de communiquer les résultats scientifiques.
- Combiner les connaissances et les observations locales avec les résultats scientifiques.
- Mettre en contexte toute l'information scientifique concernant l'état de santé général des bélugas et la regrouper dans une seule trousse de communication.

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## Objectives

- a. Work closely with beluga research scientists, Inuit organizations and key frontline workers to synthesize findings and develop a regional and community strategy that integrates all scientific programs (NCP, ArcticNet and IPY) while meeting, incorporating and providing information to frontline workers and ultimately

the development of community communication that will be made available for scientists and frontline workers and policy makers.

- b. Increase the cohesiveness among western arctic beluga programs funded within and outside of NCP that will enable all project participants to be fully aware of all on going western arctic beluga studies.

- c. Collaborate more closely with ITK/ICC, FJMC and traditional knowledge programs (Trent University) to incorporate regional insight and community knowledge on the health and well being of beluga whale along side western science.
- d. Work closely with a community – science liaison from Tuktoyaktuk (Rebecca Pokiak) to take the lead in asking community members what information they would like to receive from scientists.

## Introduction

The Beaufort Sea beluga whale population is hunted by several communities in the Inuvialuit Settlement Region (ISR), in particular Tuktoyaktuk, Inuvik, Akalvik, and Paulatuk. Currently, there several research programs investigating contaminant trends and health in this beluga population, some are supported by NCP others by funders such as IPY and ArcticNet. These research programs operate in partnership with communities and hunters who provide tissue samples and data for analyses. Reporting science findings back to the communities remains an imperative part of working with communities. In efforts to effectively communicate all research findings we are working closely with the science programs to succinctly summarize the science in a holistic manner. Strengthening communication may strengthen relationships between science and communities, increase local participation and reduce any communication redundancy. Our goal is to develop a contextualized beluga information package that integrates both western science and Inuit knowledge to help communicate

beluga findings in the Inuvialuit Settlement Region (ISR). This innovated coordinated approach to communicate contaminant findings with climate change impacts and health implications from one species (beluga) is an attempt to incorporate regional desires to communicate information in context. If successful, this approach may provide NCP with a new method for researchers and regional frontline workers to work together in the development of communication tools to be used when presenting information back to communities.

## Activities in 2008/2009

### *Field Related Activities 2008*

We worked closely with science programs to integrate and present them under one umbrella to communities for approval. The idea of working closely together was presented to FJMC (Feb 2008 meeting) who strongly supported the collaborative effort. Since our sampling efforts were to take place on Hendrickson Island we focused our communication with the community of Tuktoyaktuk, as hunters from other ISR communities to not visit Hendrickson Island.

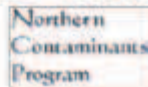
Once all approvals were received the research program was advertised using a poster posted at the Northern, Kitty Hall and the THTC office in Tuktoyaktuk. The poster listed science personal on the island and described their research. It also invited hunters to visit the island for snacks and discussions. While on Hendrickson Island we were told the poster was a good communication tool to advertise what we were doing and hunters were not surprised to find so many scientists taking samples from their whale.

## Hendrickson Island Beluga Sampling Team 2008

Program	Field Personal	Operations
FJMC Whale Monitoring	Dale Panalutluk	Record morpho, Jaw (age)
NCP Stern, Tomy	Pokiak Family	Tissue sampling chemicals
NCP Ross	Loseto, Noel, Ross	Tissue sampling health
CFL Ferguson/Raverty	Raverty	Tissue sampling disease
ArcticNet Chan/Ostertag	Ostertag	Tissue sampling neurotox
FJMC Loseto/Ross	Pokiak R., Felix, R.	Host family
Nasivvik Ostertag/Loseto	K. Felix, R. Walker	Mentoring/training



# Beluga Research on Hendrickson Island



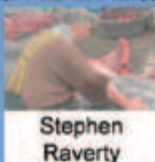
## RESEARCH

### Scientists on Hendrickson Island

- Lisa is a researcher at the University of Victoria
- Stephen is a vet at the Animal Health Centre in BC
- Sonja is doing her PhD in Prince George, BC



Lisa Loseto



Stephen Raverty



Sonja Ostertag

### Sampling and Research Goals

- Sample blubber, blood, brains and reproductive tracts from harvested whales



Beluga brain



Beluga blubber

- Study illness, nutritional stress and health effects related to contaminants in belugas

## COMMUNITY



### We will:

- Mentor youth in scientific research methods
- Provide activities for children and youth on Hendrickson
- Discuss our research with hunters and other beneficiaries
- Learn about Inuvialuit culture, language and knowledge
- Return research results to Tuktoyaktuk and Inuvik



July 5th to 25th: Activities for youth and children, tea and cookies and information about our research will be available for all visitors to Hendrickson Island

Poster design: S. Ostertag, 2008.

The beluga sampling program took on a holistic field approach on Hendrickson Island, July 2008. The integration of field efforts enhanced communication and awareness among science and community sampling programs. All participants worked closely together to sample whales in partnership with the hunters. The strong coordinated presence at the field camp helped facilitate communication with community members who wanted to learn more about the program and participate with the communications program. Thus, a well planned field season helped to pave new avenues and stronger bridges for the beluga communications program.

Through FJMC funding (L. Loseto, P. Ross) a host family (Rebecca Pokiak, Robin Felix and their toddler) was hired who also participated in sampling. We engaged the host family along with the northern students in discussions surrounding communication of the program. Given the interest

expressed by Rebecca Pokiak we invited her to participate in the communication program as a project lead.

### Fall Communication Events (Southern and Northern)

Participants at the Hendrickson Island camp (Loseto, Ostertag, Noel, Pokiak, Nasivvik students) prepared a field summary report for the THTC that was presented by R. Pokiak to the THTC. The THTC appreciated having a report in the fall, shortly after the field season, and having it presented by a community member who could respond to any concerns and questions. Additionally they were happy to hear about Rebecca's involvement in the communications program and that science would be attempting to cohesively present findings to the community.

At the annual NCP meeting in September L. Loseto presented an overview presentation of all the science programs (NCP funded and non-NCP funded) taking place at Hendrickson Island.

In efforts to bridge the traditional knowledge and community aspect Nellie Pokiak was asked to co-present and to share the work she and Frank Pokiak have been carrying out at Hendrickson Island and how there family incorporates traditional knowledge and science at the camp. Nellie Pokiak also presented a poster demonstrating all camp activities and won a poster award.

R. Pokiak was invited to attend and learn more about NCP and the communications program. At the meeting a small side meeting was held to discuss next steps. R. Pokiak, N. Pokiak, E. Loring, L. Loseto and James Pokiak (THTC chair) were present. From here it was decided R. Pokiak would be a delegate for the next THTC meeting where she would present the a) science report, b) be available for questions, c) propose plans for next summer and finally d) introduce herself as a project partner with the NCP beluga communication program.

Given the initial successes of the communications program and the well received presentation at NCP, L. Loseto and N. Pokiak along with Shelia Nasogaluak and R. Pokiak presented a talk at the Arctic Change Conference in Quebec City QC in December. The talk titled: *Integrating Science and Traditional Knowledge in the Inuvialuit Settlement Region: Perspectives from a Beluga Community Based Monitoring Program*, presented four perspectives of the beluga sampling program. It highlighted how science and community work together to better understand the health of the beluga whale. E. Loring assisted in the group co-ordination for the presentation. While preparing for the presentation we learned a lot about one another's perspectives and discussed future directions the program should take to enhance science-community collaboration.



Photo: Nellie Pokiak and her poster at the Northern Contaminants Meeting on the beluga sampling camp at Hendrickson Island 2008.





Photo: Oral presentation at the Arctic Change Conference in Quebec QC, December 11-13 2008. Speaking: Nellie Pokiak, sitting: Rebecca Pokiak, Sheila Nasogaluak, and Lisa Loseto.

A communications meeting was advertized and held at the Arctic Change Conference. Attendees included L. Loseto, E. Loring, R. Pokiak, S. Ostertag, B. Reinfort.

### ***Science Communication Events with Community Stakeholders***

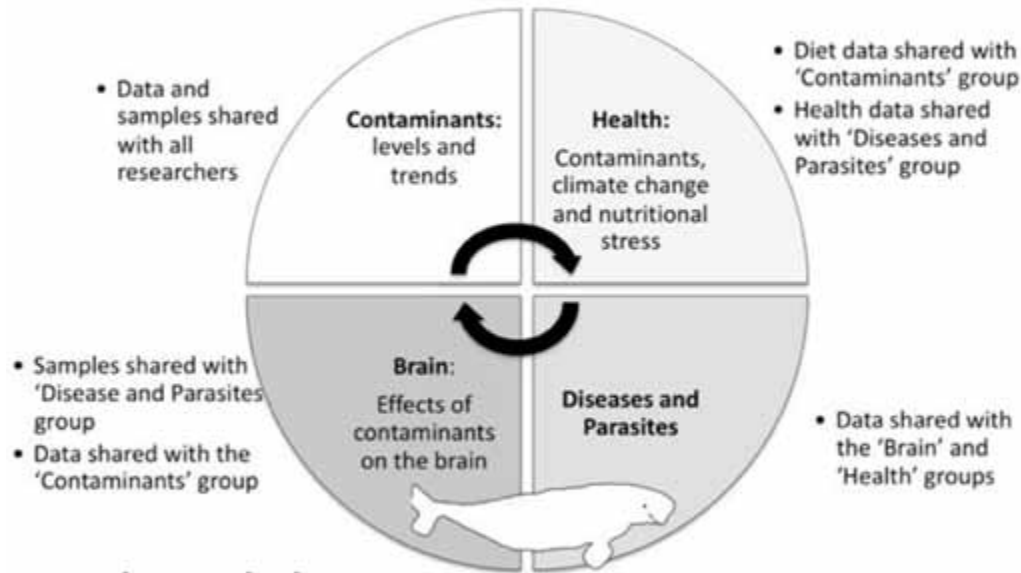
In January 2009 L. Loseto presented a talk to FJMC updating them on last year's success as well as presenting outcomes from the communications program. The board was happy with the inclusion of local members in the field, science and communication aspects and hope to see the program carry on.

Due to the logistics and the lack of final results to ready to present, a teleconference rather than a presentation with the THTC was organized with all beluga scientist research teams on February 17 2009. Template "Fact Sheets" were designed to assist communications by: a) presenting the science in a concise and cohesive manner, b) direct address HTC/ community concerns. To achieve these goals L. Loseto and R. Pokiak selected 7 questions

that would be of interest to the board members, and would require the scientists to be concise and directly answer questions to fit on to page. The template listed the research focus and the team members followed by the following questions:

- 1) *List your research questions and how belugas are sampled to answer them?*
- 2) *What is the value of for Inuvialuit, national and international levels?*
- 3) *What is the time line for your research program?*
- 4) *List 3 major findings to date. When will further results be completed?*
- 5) *Do you plan to continue sample collections next year or in the future, if so please provide a brief description?*
- 6) *Does your research involve community participation at some level?*
- 7) *Would TEK information surrounding beluga observations be of value to your research?*

# Beluga Whale Research in the ISR



## Researchers Involved:

**Contaminants:** G. Stern, G. Tomy; **Health:** P. Ross, L. Loseto, M. Noel;

**Brain:** S. Ostertag, L. Chan; **Diseases and Parasites:** S. Raverty, O. Nielsen

In addition to the fact sheets, all beluga science teams were requested to prepare their 2009 proposal to the Tuktoyaktuk HTC (THTC), together as one document. In the past they were prepared separately, this enhanced team cohesiveness and awareness of one another's programs. Finally, to help tie all the programs together S. Ostertag created a poster showing how all programs link together.

All materials were provided to the THTC before the meeting to be reviewed before the teleconference. R. Pokiak was present at the THTC for the call and assisted with organizing the calling (each science team was called individually from THTC boardroom). Prior to each call R. Pokiak had members look over the fact sheets, and following the call allowed a few moments for the board to comment. R. Pokiak prepared a summary report. Overall the report stated the THTC board was happy with the materials, meeting format and R. Pokiak's participation. They requested all science personal be present next year and to work with Rebecca before hand to develop their communi-

cation. Having Rebecca constantly available as the "expert" for beluga information provided a sense of ownership for the community and for the THTC.

## Conclusion

The Northern Contaminants Program continues to try and find new innovated tools to communicate scientific information back to communities. This project is an attempt to find a new solution for communicating a lot of scientific information back to a community and region that are more cognisant of community needs and engagement. With many research programs and scientists working on beluga whales in the ISR it important programs communicate with one another and develop an effective communication strategy with community partners. Our organized holistic approach that involves community members appears to have helped elevate such concerns. The program continues to evolve and evaluate new tools of communication. We hope stronger communication will lessen the gap between a working relationship between traditional and scientific knowledge.







