Mercury and Selenium in Ringed and Bearded Seal Tissues From Arctic Canada

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ABSTRACT. Analyses for total mercury, methyl Schreber mercury and selenium, with age determinations for 390 ringed seals (*Phoca hispida* Shreber) and 64 bearded seals [*Erignathus barbatus* (Erxleben)] from 7 localities across the Canadian Arctic confirm (with up to 420 ppm) earlier reports of very high values for total mercury in liver. Concentrations in muscle were higher than 0.5 ppm in mature animals. There were no significant differences between localities.

Mercury and age show a strong positive correlation, and so do selenium and age; the concomitant correlation between mercury and selenium is striking, the elements occurring together in a ratio by atoms of close to 1:1. Rates of accumulation appear to be somewhat higher in bearded seals.

Methyl mercury in liver amounts to less than 5 percent of the total in ringed seals and to less than 1 percent in bearded seals. There appears to be a small increase with age of the fraction present as methyl mercury. This low proportion of methyl mercury in liver, together with some 75% in muscle is in contrast to reports of 89% methyl mercury in the blood of Inuit in Arctic Bay and remains to be explained.

RÉSUMÉ. Nous avons fait des analyses de concentration totale de mercure, de mercure méthyl et de sélénium sur des échantillons de foie et de muscle de 390 phoques annelés (*Phoca hispida*) et de 64 phoques barbus (*Erignathus barbatus*) en relation avec l'âge de chaque animal. Ces phoques qui ont été chassés dans sept régions différentes de l'arctique Canadien confirment nos rapports précédents que des taux de mercure très élevés (un maximum de 420 ppm) se rencontrent dans le foie. Les concentrations totales de mercure dans les muscles étaient plus que 0.5 ppm chez les phoques adultes. Il n'y avait pas de différences significatives entre chacune des régions.

Il y a corrélations positives entre les concentrations de mercure et de sélénium et l'âge. La corrélation entre le mercure et le sélénium est remarquable; ces éléments se trouvent toujours ensemble dans une proportion moléculaire de 1:1. Les taux d'accumulation semblent plus rapides chez les phoques barbus.

La proportion de mercure méthyl est de moins de 5% dans le foie des phoques annelés et moins de 1% dans le foie des phoques barbus. Il semble y avoir une légère augmentation de la concentration de mercure méthyl chez les animaux plus âgés. Cette faible proportion de mercure méthyl dans les foies et le taux de 75% dans les muscles n'expliquent pas la proportion élevée (89%) de mercure méthyl dans le sang des Inuit d'Arctic Bay.

INTRODUCTION

In recent years it has been shown conclusively that many species of mammals and birds, especially those feeding on marine organisms, can contain high levels of mercury, even though they are not exposed to any source of industrial contamination. We have shown this to be the case of the ringed seal

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Phoca hispida and the bearded seal *Erignathus barbatus* of the Amundsen Gulf region, Northwest Territories (Smith and Armstrong, 1975).

The present study was undertaken to determine if such levels of mercury were similar throughout the arctic regions occupied by the Canadian Inuit people. Several studies (Anon., 1972; Galster, 1976; Hendzel *et al.*, 1974; R. D. P. Eaton, National Health and Welfare, pers. comm., 1977) have shown that Inuit hair and blood samples have higher mercury contents than those of peoples living in most other areas in North America. It appears that this is caused mainly by their dependence on seals and whales as food. It was felt that documentation of mercury levels in marine mammals from as many localities as possible would be an important first step in evaluating the potential health hazard to our northern native peoples.

MATERIALS AND METHODS

Frozen liver and muscle samples were obtained from marine mammals hunted in the areas indicated in Figure 1. Tissues were analysed by the method of Armstrong and Uthe (1971) for total mercury, by that of Uthe *et al.* (1972) for methyl mercury and by that of Hoffman *et al.* (1968) for selenium. Results are given in ppm (micrograms Hg or Se per gram) on a wet weight basis.

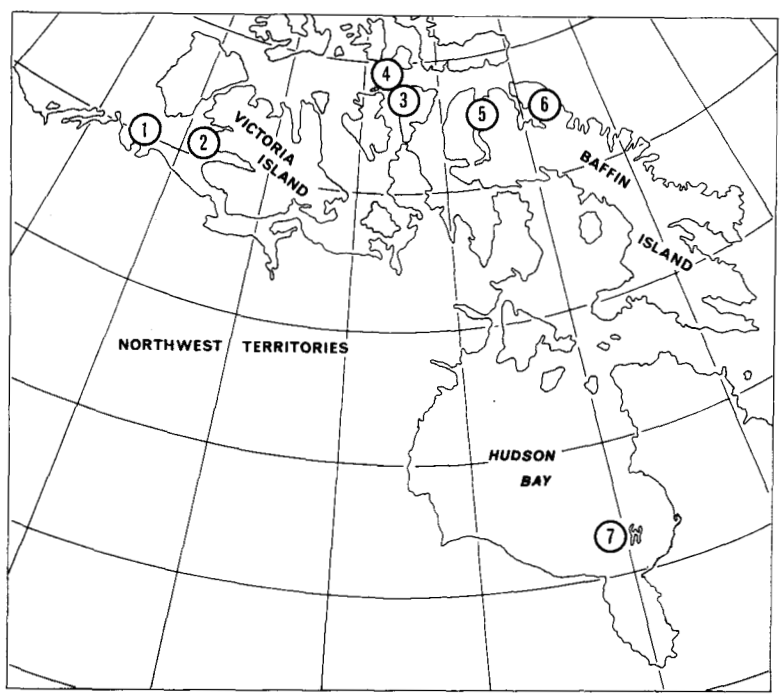

FIG. 1. Hunting localities from which ringed and bearded seal tissues were collected during this study: 1. Brown's Harbour; 2. Holman; 3. Aston Bay; 4. Barrow Strait; 5. Arctic Bay; 6. Pond Inlet; 7. Belcher Islands.

Age determination of ringed seals followed the method described by McLaren (1958a) and Smith (1973), while bearded seals were aged using both claws from foreflippers (McLaren, 1958b) and cementum lines in the upper canine teeth (Benjaminsen, 1973).

RESULTS

Ringed Seals

Total mercury and selenium contents are shown for the liver and muscle tissues of ringed seals in Table 1. Similar concentrations of total and methyl mercury in both tissues in adult animals were seen for all areas. For all such samples there were significant reductions of variation shown by the regressions of total mercury in liver and muscle on age. The differences in the mean mercury concentrations seen in the ringed seals (Table 1) are thus attributable to the different age structures of the samples. It is worth noting that the Arctic Bay sample, consisting entirely of first year ringed seals, is extremely low in mercury concentrations. In this sample only two out of 38 seals were not young of the year (not included in calculation of the mean) and both had significantly higher mercury values. To test further if there was a difference in the amount of total mercury in the Holman 1977, Pond Inlet 1976 and Barrow Strait 1976 samples, the regression coefficients were compared. No significant differences were found in the analysis of variance.

TABLE 1. Mercury, methyl mercury and selenium levels (ppm) in liver and muscle of ringed seals (*Phoca hispida*) from several different arctic localities.

	Total Hg (liver)			Total Hg (muscle)			Methyl Hg (liver)			Selenium (liver)			Mean age
Sample	Mean	S.D.	N	Mean	S.D.	N	Mean	S.D.	N	Mean	S.D.	N	(whole group)
Aston Bay (N.W. Somerset Is.) 1975	19.33	18.44	88	0.44	0.16	89				16.35	7.83	12	Not known
Barrow Strait 1976	16.14	13.84	27	0.91	0.38	27	0.89	0.45	10	9.44	6.66	10	10.20 yrs
Arctic Bay (N. Baffin I.) 1976	0.32	0.08	36	0.08	0.07	37							3 - 4 mos.
Pond Inlet (N.E. Baffin I.) 1976	3.76	3.42	33	0.31	0.17	33	0.50	0.24	8	4.13	2.67	8	5.17 yrs
Cape Parry (S.E. Beaufort Sea) 1972	1.00	1.16	13	0.23	0.11	13							1.27 yrs
Holman (W. Victoria I.) 1972-73	27.50	30.10	83	0.72	0.33	83	0.96	0.45	42	15.24	7.75	42	12.81 yrs
Holman (1977)	25.54	15.00	112				0.85	0.39	13	14.96	6.42	112	8.08 yrs

In order to generate the best predictive equations for total muscle and liver mercury, the Holman 1972, 1973, 1977 and Cape Parry 1972 samples, all from the Amundsen Gulf region, were combined. The regression equations for total mercury in liver and muscle against age were Hg = 13.9 + 1.14 age (Fs = 20.0, P <0.001) and Hg = 0.38 + 0.02 age (Fs = 37.33, P <0.001), respectively.

Although the sample sizes of ringed seals from each area were not sufficient to obtain significant reductions of variation in the regression of methyl mercury on age in all cases, the larger samples from the Holman area do show that this does occur (Holman 1977; MHg = 0.25 + 0.07 age; P of Fs <0.02).

Regressions of selenium on age for ringed seals were found to show significant positive correlations where the sample size was large enough: e.g. Holman 1977; Se = 8.82 + 0.70 age; P of Fs <0.003. Regressions of selenium in liver on total mercury in liver (ppm wet weight) were also highly significant for all ringed seal samples and when calculated to give ratios by atoms, always showed a 1:1 relationship. This had been demonstrated previously for this species (Smith and Armstrong, 1975) and for other marine mammals (Koeman *et al.* 1973).

Bearded Seals

Only two localities, the Holman area in Amundsen Gulf and the Belcher Island region of eastern Hudson Bay, were sampled systematically for the bearded seal. Comparisons of total mercury content in liver and muscle and methyl mercury in liver could indicate greater contamination in the animals from the western arctic, although the Holman sample was too small for valid statistical comparison. The difference in mean values for total mercury in liver, between the Holman and Belcher Island samples can probably be put down to the difference between the mean ages. Unaged bearded seal samples from the Barrow Strait area show mercury concentrations intermediate between the Holman and Belcher Island mean levels (Table 2).

TABLE 2. Mercury, methyl mercury and selenium levels (ppm) in liver and muscle of bearded seals (*Erignathus barbatus*) from different arctic localities.

Sample	Total Hg (Liver)			Total Hg (muscle)			Methyl Hg (liver)			Selenium (liver)			Mean age
	Mean	S.D.	N	Mean	S.D.	N	Mean	S.D.	N	Mean	S.D.	N	Years
Holman (W.Victoria Is.) 1973	143	170	6	0.53	0.35	3	0.30	0.26	6	34.42	33.23	6	8.5
Belcher Is. (E. Hudson Bay) 1974	26.18	26.13	56	0.09	0.04	55	0.12	0.04	10	20.83	13.47	10	4.9
Barrow Strait 1976 (adult)	79.2		1	0.20		1							
Barrow Strait 1976 (adolescent)	9.42		1	0.14		1							

For the Belcher Island sample the regression of total liver mercury on age was highly signficant (Hg = −1.79 + 5.11 age; Fs = 88.42, P <0.001). The regressions of total mercury in muscle and methyl mercury in the liver on age did not show any significant reduction in variation.

A highly significant positive correlation of selenium in liver with age and with total liver mercury was found (Se = −1.05 + 3.77 age; Fs = 23.52, P <0.001 and Se = 5.53 + 0.42 (Hg liver), Fs = 148.88, P <0.001, respectively). Again, the molar ratio of selenium and total mercury in liver was found to be almost unity (regression coefficient = 1.08, S.E. = 0.08).

Comparison between ringed and bearded seals reveal some differences with respect to mercury. Both show significant increases of total mercury in the liver with age and do not differ significantly in their regression coefficients. Both species also show an increase of selenium with age and high correlation of total mercury in the liver with the amount of selenium in the same organ. In both these cases the bearded seal regression coefficients are significantly larger, indicating a probable higher rate of accumulation in that species.

Comparison of the mean total mercury in muscle of ringed seals for Holman Island 1973 with that in bearded seals from the Belcher Islands shows a significantly higher concentration ($t = 14.07$, $P < 0.001$) in the ringed seal. Again, methyl mercury concentrations in liver were shown to be significantly higher in the Holman 1977 ringed seals than in the Belcher Island bearded seal sample ($t = 5.76$, $P < 0.001$). Methyl mercury, expressed as a percentage of total mercury in the liver, was significantly higher for the ringed seals in all comparisons made between the two species (Table 3). In these comparisons the mean age of the ringed seal samples was higher than that of the bearded seals; however, a regression of percent methyl mercury in livers of the Holman 1973 sample on age did not show any significant reduction in variation. A regression for this same sample of percent methyl mercury on total mercury in the liver did show a significant negative relationship (regression coefficient = -0.11; P of Fs < 0.03). A negative slope was also shown in the regression of percent methyl mercury on total liver mercury in the Belcher Island bearded seal sample but the reduction in variation was not quite significant, possibly because of the smaller sample size. Thus, there appears to be a real species difference in the percentage as well as the absolute amount of methyl mercury in the liver.

TABLE 3. Percentage of methyl mercury in liver tissues of ringed and bearded seals.

Sample	Percentage methyl mercury			Mean age of sample
	$\bar{X}$	S.D.	N	Years
Ringed seal (Holman 1973)	5.84	12.29	42	14.33
Ringed seal (Holman 1977)	3.29	1.16	13	9.38
Bearded seal (Holman 1974)	0.39	0.32	6	8.50
Bearded seal (Belcher Is. 1974)	0.58	0.71	10	5.41

DISCUSSION

Top carnivores throughout the oceans of the world accumulate high concentrations of mercury. Often these animals are far removed from areas of industrial mercury contamination. It is generally accepted that, on a global

scale, industrial sources do not account for more than one-half of the annual input of mercury to the oceans (Krehl, 1972). At the same time there are good indications that there has been a world-wide increase in the input of mercury from natural sources into freshwater systems, caused by the gradually changing pH of rainwater since the Industrial Revolution (Jernelov *et al.*, 1975). Increased input of either sort to the oceans is insufficient to make any significant change in concentration in the waters.

Our studies of ringed and bearded seals in arctic Canada have shown them to be similar as regards mercury contamination to other marine mammals samples in areas which are not directly influenced by increased mercury loads from industrial dumping. In the ringed seals from this study there does not appear to be any significant difference in mercury levels between areas as widely separated as Holman in the western arctic and Pond Inlet on eastern Baffin Island. There is a slight indication that the bearded seal, a more sedentary species with benthic feeding habits, might have higher mercury contents in the western Canadian arctic than in Hudson Bay. There are indications from other studies of pinnipeds done in western Alaska that mercury contents in their tissues are much lower (Galstar, 1971). This is also reflected by the mercury content of polar bear tissues (Lentfer, 1976) and in the tissues of Inuit people which are much lower in the western Alaskan localities (Galster, 1971) than in the Canadian arctic. Since there do not appear to be any sources of direct industrial mercury pollution in these arctic areas, it would appear that regional differences must be due either to as yet unknown differences in mercury in the environment or to differences in diet. For areas in the United States higher mercury values in humans from areas containing high natural mercury-bearing ores have been demonstrated (Gabica *et al.*, 1975). Unfortunately, little is known of either the subsea geology, the mercury content of sea water or of other arctic marine organisms for most of the vast arctic marine areas.

Both ringed and bearded seals accumulate high levels of naturally occurring mercury in their livers. In this respect they are similar to most other marine mammals and other long-lived top marine carnivores such as tuna and several species of billfish (Miller *et al.*, 1972). In no instance, in the large sample of ringed and bearded seals that we have examined, was there any indication of mercury intoxication, although complete chemical and pathological examinations of the seals were not possible. Only one case in all the literature dealing with mercury in seals suggests that it might have had some toxic effects (Helminen *et al.*, 1968) and this was from an area of heavy industrial mercury dumping. It appears likely that marine mammals in general have developed mechanisms enabling them to cope with the substantial amount of mercury that naturally occurs in their diets.

Both species in this study accumulate large amounts of mercury in the liver. This is mostly in a non-methylated form, although the large proportion of mercury in the fish they consume is methylated (Westoo, 1969). There appears, therefore, to be a mechanism for demethylating mercury, which may be in the liver. The demethylation of mercury can be considered a form of

detoxification since methyl mercury is known to be most easily transferred across the blood-brain barrier, thus causing the central nervous system disorders associated with mercury poisoning. The conversion of methyl mercury to inorganic mercury in the liver also probably facilitates mercury excretion possibly via the bile. It is known from experimental studies that inorganic mercury is preferentially excreted through the faeces (Norseth and Clarkson, 1970), at least in rats and man.

The presence of selenium in a 1:1 atomic ratio with mercury in the liver, and the fact that selenium is 40 times as abundant as mercury in marine fish tissues (Taylor, 1976) indicates a biochemical process binding these two elements in the seal. Parizek and Ostadalova (1967), Ganther *et al.* (1972) and Parizek *et al.* (1974) showed experimentally that selenium could modify the toxic effect of methyl mercury. Other compounds or elements could also be involved. More recently, Stillings *et al.* (1974) showed that cysteine had a similar and additive effect. Bromine has also been implicated as an essential element in relation to mercury and selenium in studies comparing natural, as opposed to premature births in the California sea lion *Zalophus californianus* (Martin *et al.* 1976).

The mechanism of detoxification by selenium is not understood. It appears that when selenium is ingested along with mercury some mechanism operates in seals which causes both metals to combine and become immobilized in the liver. Stillings *et al.* (1974) showed that in rats cysteine and selenium did not inhibit toxicity by increasing the elimination of mercury from the body. It appeared to reduce methyl mercury excretion by the kidney by demethylating mercury and increasing its storage in other organs, notably the liver.

There are no definite indications from the large sample of ringed seals in this study that a breakdown in the detoxification mechanism at high liver mercury levels as suggested by Koeman *et al.* (1972) and Roberts *et al.* (1976) might occur. In this study three seals, showing an unusually high proportion of methyl mercury in the liver, were very old animals. The shift to high methyl mercury concentrations cannot, however, be caused in these cases by an overburden of mercury because their overall mercury levels were lower than the mean levels in the sample. It was also seen in the Holman 1977 sample that the regression for percent methyl mercury on total mercury in the liver showed a significant negative correlation. Other factors, such as age or disease (Buhler *et al.*, 1975) might also be implicated in causing a breakdown in both the detoxification and excretion mechanisms in some instances.

Several studies conducted on Inuit in different Canadian (Table 4) and American arctic (Galster, 1976) localities show that there are somewhat elevated levels of mercury in their hair and blood compared with the rest of the North American population. It is generally accepted that this is caused by the Inuit dependence on marine mammals for food (Smith and Armstrong, 1975; Galster, 1976). It comes as a surprise then that for the Arctic Bay Inuit an average of 88.8% of the mercury in the blood is of the methylated form when only 6% of the mercury in the heavily-laden ringed seal liver and 75% of the very low mercury content of ringed seal muscle is methylated mercury.

TABLE 4. Mercury contents (ppb) of Inuit blood and hair from several different arctic localities.

Source	Locality	Hair						Blood					
		Total Mercury			Methyl Mercury			Total Mercury			Methyl Mercury		
		$\bar{X}$	S.D.	N	$\bar{X}$	S.D.	N	$\bar{X}$	S.D.	N	$\bar{X}$	N	$\bar{X}$ percentage
Anon. (1972)	Holman							34.01	11.86	23			
Hendzel et al. (1976)	Igloolik	15.35		134									
National Health and Welfare (R.D.P. Eaton, pers. comm. 1977)	Inuvik	7.58		56									
National Health and Welfare (R.D.P. Eaton, pers. comm. 1977)	Arctic Bay	8.42	4.9	51				23.30	12.39	45	27.1	33	88.8

Smith and Armstrong (1975) showed that there were no other large sources of mercury in the Inuit diet in the Holman area and it is believed that the Arctic Bay natives eat essentially the same food items. Rowland *et al.* (1975) and Rowland *et al.* (1977) demonstrated that there were bacteria in the gut of rats and humans which could methylate inorganic mercury. His calculations revealed that the human gut could methylate 400 mg of mercury a day which would likely be almost completely absorbed into the blood. This mechanism might then explain the high percentage of methyl mercury in human blood which is obtained from the large proportion of inorganic mercury in seal liver and muscle which the Inuit eat.

Methyl mercury is present in the diet of many groups of native Canadians. Some have elevated blood-mercury levels (Bernstein, 1974; Barbeau *et al.*, 1976; Charlebois, 1977) but so far there has been no clear evidence of methyl mercury poisoning. Clear evidence is of course very difficult to come by since diagnosis is not straightforward, particularly where other diseases are common, so that it would be unwise to assume that there is no danger.

It is possible that people traditionally dependent on marine mammals for food have evolved an adaptation to their diet. There is an opportunity here for research. Better understanding of the subject of methyl mercury in diet may be vital to peoples whose way of life is changing rapidly.

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