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^{210}Pb Flux in an Arctic Coastal Region[1]

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ABSTRACT. Seven marine coastal cores and one lake core were collected from a deltaic region in northern Alaska and analyzed for ^{210}Pb. The ^{210}Pb activity levels at the surface in all cores were less than the levels generally observed in coastal sediments of nonpolar regions. The attenuated flux of ^{210}Pb to this region, as deduced from analyses of snow deposits, is probably responsible for these reduced activity levels. Yet the calculated flux of this nuclide to the sediments (about 1 and 4 dpm cm^{-2} y^{-1} in the marine and lake sediments respectively) is in excess of that accountable from the atmospheric flux (0.08 dpm cm^{-2} y^{-1}). Presumably processes such as advection of ^{210}Pb contribute importantly to the sedimentary flux.

Key words: lead, cores, arctic coastal plain, radionuclide flux, air masses, lake sediments.

RÉSUMÉ. Sept carottes côtières marines et une carotte de lac furent recueillies d'une région deltaïque dans le nord de l'Alaska et analysées quant à leur teneur en ^{210}Pb. Les niveaux d'activité de ^{210}Pb à la surface de toutes les carottes étaient moins élevés que les niveaux ordinairement observés dans les sédiments côtiers des régions non polaires. Le flux atténué de ^{210}Pb à cette région tel que déterminé par l'analyse de dépôts de neige explique probablement ces niveaux d'activité réduite. Cependant, le flux calculé de ce nuclide aux sédiments (environ 1 et 4 dpm cm^{-2} y^{-1} dans les sédiments marins et lacustres, respectivement) excède la quantité attribuable au flux atmosphérique (0.08 dpm cm^{-2} y^{-1}). Les processus tel que l'advection du ^{210}Pb contribuent probablement de façon importante au flux sédimentaire.

Mots clés: plomb, carottes, plaine côtière arctique, flux de radionuclides, masses d'air, sédiments lacustres

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INTRODUCTION

The initial geochronological work with ^{210}Pb was on permanent snow fields (Goldberg, 1963), and the approach was subsequently extended to marine systems (Koide *et al.*, 1972). The commonly accepted basis for the procedure depends upon out-gassing of ^{222}Rn (half-life 3.8 days) to the atmosphere from its ^{226}Ra precursor (half-life 1622 years) in the regolith. The ^{210}Pb (half-life 22.3 years) is formed through a series of rapid decays. The half-lives of the intermediate decay products are brief by comparison with the average residence times of the aerosol. Thus, essentially all atoms of ^{222}Rn are transformed into ^{210}Pb in the atmosphere. The ^{210}Pb subsequently reaches to the earth's surface from the atmosphere with precipitation and dry fallout. Measurement of ^{210}Pb as a function of depth has been useful in establishing the chronology of recently deposited coastal marine sediments (Koide *et al.*, 1972; Koide *et al.*, 1973; Bruland, 1974) and in lake sediments (Krishnaswami *et al.*, 1971; Robins and Edgington, 1975; Von Damm *et al.*, 1979).

The relatively lower concentrations of ^{222}Rn in northern latitudes (Kirichenko, 1970; Anderson and Larson, 1974; Wilkening *et al.*, 1975) suggests that the flux of its longer-lived decay product, ^{210}Pb, may also be reduced in this environment. To evaluate this possibility, analyses of ^{210}Pb in sediments from Simpson Lagoon and east Harrison Bay, located in the Colville delta region of north arctic Alaska, were undertaken. With these values a mass sedimentation rate was estimated, and from this rate the ^{210}Pb flux was computed. The determination of this nuclide in snow provided a direct measurement of atmospheric flux to the environment. Finally, the values derived in this study are compared with results obtained in other geographic regimes, and reasons for the differences are suggested.

DESCRIPTION OF THE STUDY AREA

Simpson Lagoon, which is contiguous with the Colville Delta, lies between a chain of barrier islands and the Arctic Coastal Plain province of north Alaska (Fig. 1). The most prominent climatic features of this region are long, severely cold winters with ice cover that persists for 8 to 9 months of the year and cool summers for the remainder of the year. The lagoon is oriented parallel with the coastline. It is shallow (1-4 m deep), and about 2.4 km long. Sediment and water mass characteristics are greatly influenced by the protracted duration of the yearly ice cover and the subsequent spring discharge of the adjacent rivers (Colville and Kuparuk). At breakup, usually in early June, the river melt waters flow over and under the lagoon ice, and the fluvial overflows on sea ice are largely confined to the lagoon and the adjacent bay. During spring and summer the terrigenous debris in the form of sandy mud reaches the lagoon primarily through suspension, river overflow and/or ice-rafting. However, most of the mud of this debris does not immediately settle, owing to the turbulence prevailing, whereas the sand quickly deposits on the bottom. Most of the clay-size particles settle in the fall and winter months when wave and current actions have subsided considerably (Naidu and Mowatt, 1974).

MATERIALS AND METHODS

For the purposes of estimating the fluxes of ^{210}Pb into sediments and determining the sedimentation rates, seven core samples were recovered from aboard the R/V *Natchik* from the coastal area of the Simpson Lagoon and east Harrison Bay region of north arctic Alaska in August 1979 (locations provided in Table 1 and shown in Fig. 1). All cores were collected from

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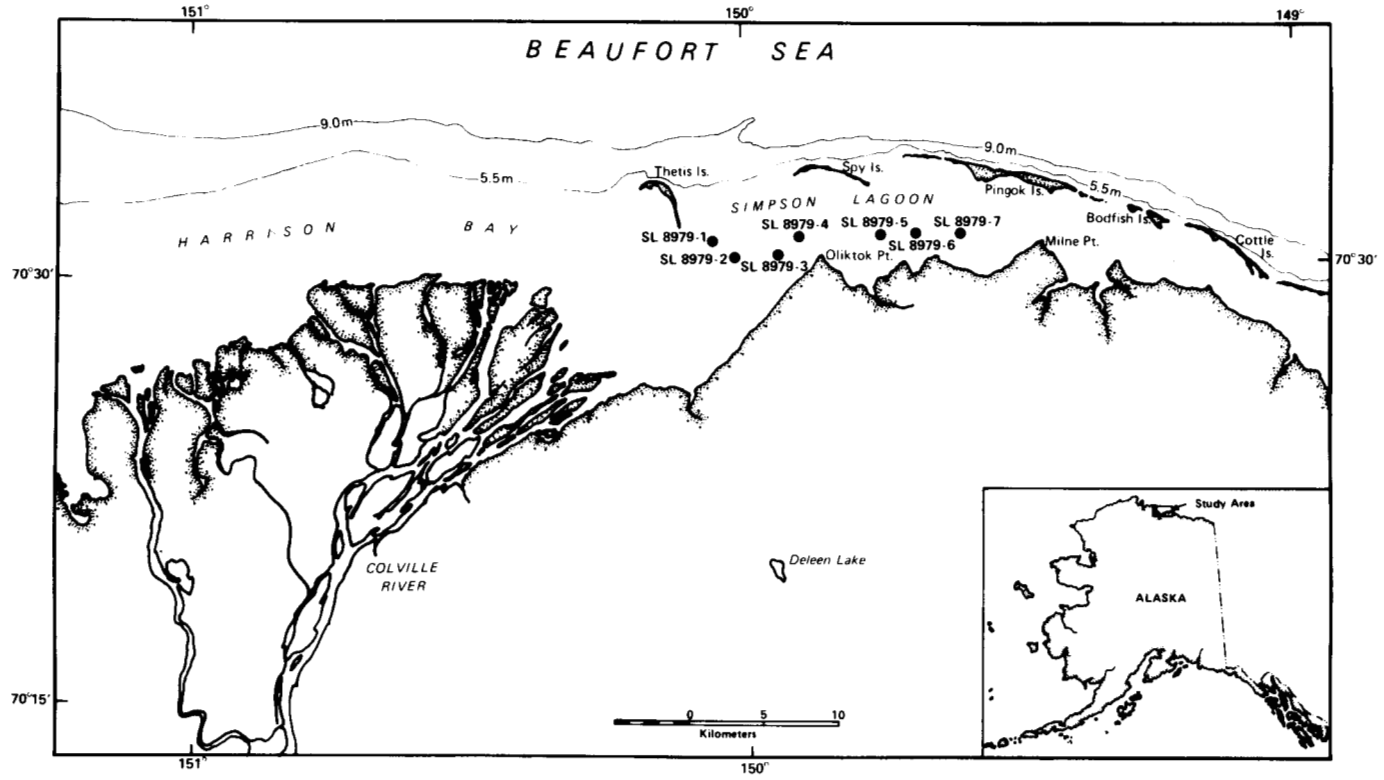

FIG. 1. The area of study showing locations of the core samples in the Simpson Lagoon, east Harrison Bay and Deleen Lake.

TABLE 1. Locations and water depths (m) at which samples were collected

Core no.	Latitude (N)	Longitude (W)	Water depth (m)
SL8979-1	70°32′	150°07′	3.9
SL8979-2	70°31′	150°01′	3.6
SL8979-3	70°31′	149°57′	3.3
SL8979-4	70°32′	149°53′	3.0
SL8979-5	70°32′	149°45′	2.7
SL8979-6	70°32′	149°40′	2.6
SL8979-7	70°32′	149°35′	2.6

waters deeper than 2 m (the depth to which the lagoon and bay waters generally freeze) to avoid possible perturbation of surficial sediment by ice gouging. The longest core obtainable was 33 cm. The sediment core sample from the coastal lake (Fig. 1) was obtained from a 3.5 m depth of water. For estimation of the atmospheric flux of ^{210}Pb the snow deposits, about 30 cm deep, were gathered on 15 January 1978 from the frozen surface of the mouth of the Colville and Kuparuk rivers and analyzed in May 1980. This time lapse between collection and analysis of the snow sample insured secular equilibria between Po and Pb isotopes.

Cores were sectioned into 1 cm segments and each segment was split into two portions. One fraction (1-2 g) was reserved for the ^{210}Pb analysis; the other was used for size distribution analysis by the usual sieve-pipetting method.

Various methods are available for ^{210}Pb analysis. The one employed is similar to that described by Nittrouer *et al.* (1979) and depends upon its secular equilibrium with ^{210}Po. Sediment samples were dried at 110°C overnight, and a weighed aliquot (1-2 g) was ashed out at 400-450°C overnight and then spiked with ^{208}Po tracer. It should be noted that whether ^{208}Po tracer was introduced before or after the dry ash step, the results were consistent. Further analyses agreed with samples that were oxidized by the wet ashing technique. Thus, a differential loss between ^{208}Po and ^{210}Po by $PoCl_2$ volatilization was not discernible. Snow samples were melted and acidified with conc. nitric acid to a final concentration of 0.6 N. One-liter aliquots were spiked with ^{208}Po and then slowly evaporated to dryness. Sediment and snow samples were further processed in the following manner: The residue was treated with about 30 ml each of conc. hydrochloric acid, aqua regia, and conc. hydrochloric acid, in that order. With each treatment, the liquid was evaporated to dryness at simmering temperatures. The final residue was prepared for autodeposition by its dissolution in 0.3 N hydrochloric acid. The alpha particles emitted by ^{208,210}Po deposited on silver disc were pulse-height analyzed with a surface barrier detector, usually for an overnight period.

RESULTS

The range of values for the ^{210}Pb disintegration rate, dpm/g, extended from 0.34 in core 8579-3 to 5.44 for the Deleen Lake core. The error, as determined in replicate analyses, was 3 and 10% at the higher and lower disintegration rates respectively.

The recent rate of sedimentation was determined from the slope of the least-squares regression of depth in the core on the logarithm of ^{210}Pb. In a number of cores, usually at a depth of 15-16 cm, and extending to the bottom, a divergence in the ^{210}Pb values from linearity was evident. These values were not included in the computation of the sedimentation rates. Table 2 presents a summary of the number and range of segments included in the least-squares fit, the slope, intercept, correlation coefficient and the tabular coefficient at the 99% confi-

TABLE 2. Sedimentation rates and ^{210}Pb flux of the lake, bay and lagoon cores, including linear coefficients, number and range of linear segments and correlation coefficients

Sediment core	Number of linear segments (n)	Linear range (cm)	Intercept (*a*)	Slope (*b*)	Correlation coefficient	Tabular (r) at 99% level	Sedimentation rate cm y^{-1}	Mass sedimentation rate g cm^{-2} y^{-1} (r)	^{210}Pb flux dpm cm^{-2} y^{-1} (F)
SL8979-1	30	0-33	1.77	–0.019	0.75	0.46	$1.64^{+0.15}_{-0.13}$	2.05	3.63
SL8979-2	14	0-14	1.85	–0.038	0.75	0.66	$0.82^{+0.13}_{-0.10}$	1.03	0.91
SL8979-3	20	0-20	1.56	–0.067	0.98	0.56	$0.46^{+0.03}_{-0.02}$	0.58	0.90
SL8979-4	11	0-11	—	—	0.26	0.74	—	—	—
SL8979-5	16	0-16	1.82	–0.054	0.91	0.62	$0.58^{+0.04}_{-0.04}$	0.73	1.93
SL8979-6	15	0-15	1.24	–0.042	0.75	0.64	$0.74^{+0.07}_{-0.06}$	0.93	1.15
SL8979-7	15	0-15	1.57	–0.060	0.89	0.64	$0.52^{+0.05}_{-0.04}$	0.65	1.02
Deleen Lake	21	0-21	4.86	–0.035	0.60	0.55	$0.89^{+0.15}_{-0.12}$	0.80	3.89

dence level. The sedimentation rate and associated error were calculated from the slope and the standard error of the slope (Snedecor, 1950).

Included in Table 2 are r, the mass sedimentation rate (g cm^{-2} y^{-1}), and F, the ^{210}Pb flux at the sediment surface (dpm cm^{-2} y^{-1}), derived from

$r = W(1- Q)\rho$, and

$F = A_o r$

where:

W is the linear sedimentation rate (cm y^{-1})

Q is the fractional water content

ρ is the mean density of solids (g cm^{-3})

A_o is the ^{210}Pb activity at the surface dpm/g

The fractional water content for the marine sediments and Deleen Lake was taken as 0.5 and the mean density for the respective sediments was estimated to be 2.5 and 1.8 g cm^{-3} respectively.

With the exception of core SL8979-4 ^{210}Pb concentrations, the correlation coefficient exceeded the tabular correlation coefficient at the 99% confidence level. This result clearly indicates the statistical significance of the calculated *a* (intercept) and *b* (slope) constants. The sedimentation rates in the marine cores ranged from 0.5 to 1.6 cm y^{-1}. Their intercept values extended from 1.24 to 1.85 dpm/g. A sedimentation rate of 0.9 cm y^{-1} was measured for the core from Deleen Lake. Plots of the data for the cores appear in Figure 2. The solid line in each covers the region in the core over which linearity obtained.

The ^{210}Pb flux centered around 1.3 dpm cm^{-2} y^{-1}, except for the core taken off the mouth of the Colville River and from Deleen Lake. In these two cases, the value was about 3 times greater (Table 2).

The concentrations of ^{210}Pb in the snow on the Colville and Kuparuk rivers were 5.5 ± 0.3 and 8.4 ± 0.4 dpm/kg respectively.

The stratigraphic variations in the contents (wt.%) of gravel, sand, silt, and clay in the Simpson Lagoon and Harrison Bay cores have been reported elsewhere (Naidu, 1980). The sediment textures are variable both between and within cores. Textural analysis of the individual cores indicates that the lagoonal sediments are generally sandy silts with minor amounts

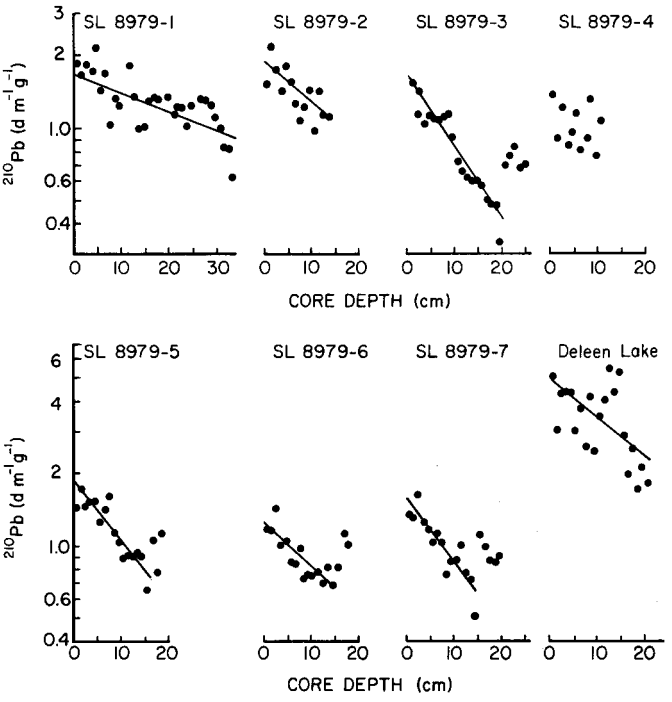

FIG. 2. Profiles of ^{210}Pb activities for the 8 sediment cores investigated. See Figure 1 for core locations.

of clays. The sediment from Deleen Lake was essentially muddy, with an organic content approximately 50%, as determined by weight loss upon thermal ashing.

DISCUSSION

Variations of the ^{210}Pb values from the best-fit line for the coastal cores are substantially greater than usually observed and in excess of the error associated with the radiometric assay. These fluctuations are, in part, attributable to the degree of coarseness of sediments along the length of the cores. Relatively lower activity levels associated with the coarser sandy particles have been reported a number of times (Nittrouer *et al.*, 1979; Kalesha *et al.*, 1980; Smith and Walton, 1980) and reflect the

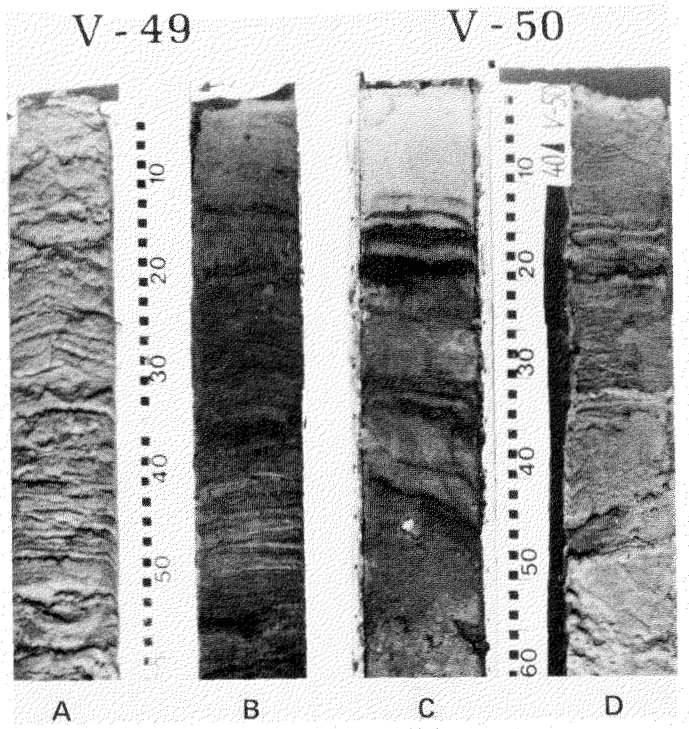

FIG. 3. Vibro core samples from the Simpson Lagoon area displaying preservation of original laminations. The cores were split longitudinally into two halves. One half was resin mounted (A and D) to heighten the structures, and the other is shown in the original state (B and C).

lesser capacity of these particles to scavenge ^{210}Pb from the water column. Yet a calculation of ^{210}Pb in the core segments on a sand-free basis did not result in improvement in the least-squares fit. Another source of variability may relate to fluctuations in the sedimentation rate according to an alternate model proposed by Appleby and Oldfield (1978) and further discussed by Robbins (1978). The north Alaskan arctic lagoonal sediment is derived primarily from land erosion during spring discharge. Since this event may vary significantly in magnitude from year to year, it could account to some extent for the degree of scatter about the line of regression. Nonetheless, for six of the seven cores the data correlated significantly with the linear equation.

Sedimentation rates were derived without correction for the quantity of background-supported ^{210}Pb. Thus, the rates presented represent upper limits for sedimentation. Furthermore, the appearance of undisturbed laminations as displayed along the length of some representative cores (Fig. 3) taken from Simpson Lagoon indicates that the sedimentation rates are uninfluenced by reworking of surface sediments (Benninger *et al.*, 1979).

A deviation from exponential decay occurred at a depth of 15-16 cm for cores SL8979-5, 6 and 7 and at 21 cm in core SL8979-3. An event that occurred two to three decades ago may account for the elevated ^{210}Pb values. Whether the ^{210}Pb in this underlying layer was originally in excess or decayed from ^{226}Ra could not be established due to insufficient core length.

A comparison on the intercept values (Table 2) reveals that a substantially higher activity level prevails for the sediment from the lake. This difference may be partially explained by sediment composition. In contrast with the sandy muds characteristic of the lagoonal sediments, the debris descending through the water column of the lake is largely organic and as such scavenges ^{210}Pb more effectively (Lewis, 1977). Further, the lake serves as a reservoir for atmospherically deposited ^{210}Pb, whereas no such confinement exists in the coastal waters. Some of the sea ice is mobilized seaward out of the lagoons subsequent to its breakup in July (Naidu and Mowatt, 1974). Accordingly, the ^{210}Pb accumulation, beginning with the formation of the ice since the previous September, is transported away from the site of deposition and thereby could effect a partial depletion of ^{210}Pb in the littoral environment.

It is apparent that the surface marine sediments in the Arctic carry a lesser concentration of ^{210}Pb than those of more temperate regions. For comparable sedimentation rates, representative values fall between 5 dpm/g in Narrangansett Bay (Goldberg *et al.*, 1977) to 50 dpm/g in Santa Barbara Basin (Koide *et al.*, 1972) and Baja California (Koide *et al.*, 1973). Although scavenging efficiency and ice movement may account for much of the difference in ^{210}Pb content between Deleen Lake and bay and lagoon sediments, the difference noted between the Arctic and other marine sediments may be influenced by an additional element. This is indicated by a comparison of the data from Deleen Lake with that of other lakes.

The scatter in the data from Deleen Lake suggests some post-depositional sediment disturbance. The sediment was largely organic, of fluffy consistency, and therefore subject to mixing. Thus, the intercept value, 4.86 dpm/g, is probably too low. However, it seems doubtful that a value as high as 16 dpm/g, which is the lowest of surface values recorded for a lake at temperate latitudes (Koide *et al.*, 1973) would be attained were the sediment undisturbed. More representative surface values of ^{210}Pb for sedimentation rates comparable to that of Deleen Lake range from 45 dpm/g for a Sierra Nevada subalpine pond (Shirhata *et al.*, 1980) to 85 dpm/g for Trout Lake in Wisconsin (Koide *et al.*, 1973).

The lower concentrations of ^{210}Pb in arctic sediment may partly result from a weakened atmospheric flux at northern latitudes. The measurement of this nuclide in surface waters of the oceans suggests that the flux at higher latitudes is substantially less than in temperate zones (Schell, 1977). The measurement of ^{222}Rn further supports the consideration of a reduced flux in the Arctic. The average ^{222}Rn flux is variously given as 0.7 atom cm^{-2} sec^{-1} (Israel, 1951) and 0.75 atom cm^{-2} sec^{-1} (Wilkening *et al.*, 1975). Fluxes measured in a northern region of the USSR (65°N, 40°W) by Kirichenko (1970), the Yukon Basin (65°N, 150°W) by Anderson and Larson (1974) and the Tanana Basin (65°N, 147°W) by Wilkening *et al.* (1975) are only 0.18, 0.33 and 0.37 atom cm^{-2} sec^{-1} respectively. ^{210}Pb data acquired for Arctic Lake sediments further indicate low atmospheric ^{210}Pb flux (Kipphut, 1978; Cornwell, 1983) in northern latitudes.

Our analysis of snow samples for ^{210}Pb affirms the flux concept. The samples represented the accumulation of about 4 months of precipitation. Information available from the Lonely DEW Station and the weather station at Point Barrow, which are respectively situated 130 and 260 km to the west of the study area, indicated that snow equivalent to 5.5 cm of water was deposited over this period (James L. Wise, pers. comm.). The average for the two snow samples, 6.9 dpm/kg, leads to a flux of 0.08 dpm cm^{-2} y^{-1} for the average annual precipitation of 12 cm y^{-1} in this region of the Arctic. It is understood that the calculation of a flux from data acquired over such limited duration could be uncertain by as much as a factor of two (Turekian *et al.*, 1977). However, this value contrasts sharply with a ^{210}Pb atmospheric flux of 1.0 ± 0.2 dpm y^{-1} in the

northeastern United States (Benninger, 1978), a region that has received concentrated study.

It is interesting to note that the ^{210}Pb atmospheric flux calculated for this region of the Arctic agrees with fluxes derived from permanent snow-field data from Greenland. At Camp Century (77°N, 61°W; 1885 m elevation) at South Dome (63°N, 44°W; 2700 m elevation), and at another Greenland site (77°N, 56°W; 2000 m elevation) fluxes of 0.09 dpm cm^{-2} y^{-1} (Crozas and Langway, 1966), 0.06 dpm^{-1} cm^{-2} y^{-1} (Koide *et al.*, 1979) and 0.11 dpm cm^{-2} y^{-1} (Windom, 1969) were measured. However, whether or not the Greenland values reflect the flux at sea level is not certain. It has been suggested (Turekian *et al.*, 1977) that the low fluxes could be a sign of effective scavenging of ^{210}Pb from air masses prior to their arrival at these altitudes.

The analysis of snows indicates that the ^{210}Pb atmospheric flux to this environment is appreciably depressed. However, it should be noted that, as in other environments, the ^{210}Pb flux to the sediment, 1-3 dpm cm^{-2} y^{-1} (Table 2), is substantially greater than that accountable from atmospheric input. As previously suggested (Carpenter *et al.*, 1981), advection of sea water containing dissolved ^{210}Pb produced *in situ* from ^{226}Ra may account for this enrichment. Accordingly, either a less dominant advective influence or a lower concentration of ^{210}Pb precursors, or combination of these factors, may contribute to the paucity of ^{210}Pb in these arctic sediments.

The possibility that the lower concentrations of ^{210}Pb in arctic sediments are simply due to dilution by relatively inert material such as sand-size particles appears unlikely because the sediments are generally muddy. It is to be further noted that the arctic sediments in question have relatively high amounts of smectite (Naidu, 1982), a clay mineral type with a high potential for ^{210}Pb adsorption.

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