

MERCURY IN VEGETATION AND LAKE SEDIMENTS FROM THE U. S. ARCTIC

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Abstract. Global atmospheric concentrations of mercury (Hg) appear to be increasing and with it the potential for ecosystem exposure and ecological effects. From 1990 to 1993 we examined U. S. arctic ecosystems over a broad spatial scale to develop baseline information on current concentrations of trace elements, heavy metals (including Hg), persistent organic compounds, and radionuclides in various components of the terrestrial and freshwater biosphere. Matrices reported here include, vegetation (lichens and mosses) and lake sediments. Total Hg in two lichen and two moss species from Alaska were generally low (0.02 - 0.112 µg/g dw), compared to reported values from other arctic locations and showed a statistically significant negative relationship between total Hg content and distance from the marine coastline. ²¹⁰Pb dated sediment cores indicated that average preindustrial total Hg accumulation rates were over four times greater in arctic Schrader lake than in subarctic Wonder Lake. Both lakes indicated a small increase (5 - 8%) in total Hg flux to the sediments during the last 145 years, much smaller than similar increases in total mercury for lakes in the north central U. S. The likely source of recent increases in Hg in these Alaskan ecosystems is long range atmospheric transport. While we can detect increases in mercury in lake sediments likely due to anthropogenic activities, values are low and there appears to be no immediate threat to terrestrial environments and inland freshwaters of arctic Alaska from long range atmospheric transport and deposition of Hg.

1. Introduction

The Arctic is not a remote, pristine ecological setting, but a remote region that is exposed to long range transport of airborne contaminants (Shaw and Khalil, 1989; Rahn and McCaffrey, 1980). Atmospheric contaminants pose a threat of uncertain magnitude to the function and structure of arctic ecosystems, as well as to arctic people that subsist on animals and plants (Barrie, 1986; Rahn *et al.*, 1989). In recent years the eight circumarctic nations have banded together under the Arctic Environmental Protection Strategy (1991) and implemented the international Arctic Monitoring and Assessment Program (AMAP Report, 1993). Along with chlorinated hydrocarbons, trace metals, in particular Hg, are of high interest due to their persistence in the environment and their known ability to adversely affect ecosystems, including humans (Schroeder, *et al.*, 1987; Clarkson, 1990). Environmental Hg may either be from natural sources (i.e. cinnabar deposits) or atmospheric emissions produced as a by-product of many forms of human industrial activities that include coal combustion, metal smelting, natural gas production and oil exploration (Mittra 1986, Nriagu and Pacyna 1988). Contaminants accumulating in the Arctic are generated within the Arctic as well as from more distant locations (Pacyna 1991). For some groups of contaminants, perhaps including mercury (Steinnes, this volume), the Arctic may function as an ultimate global sink due to the phenomenon of "global fractionation" (Ottar, 1981; Wania and Mackay, 1993).

Our understanding of the factors influencing the transport and mobility of various forms of Hg within temperate watersheds has improved in recent years (Bloom, 1989; Ramlal *et al.*, 1993; Engstrom *et al.*, 1994). This knowledge, however, may not translate directly to Hg dynamics in arctic environments, which differ in many significant ways from lower latitude ecosystems. Knowledge concerning concentrations and forms of Hg in arctic

ecosystems is rudimentary; many investigators addressing metals in the Arctic have just begun to measure Hg. The objectives of this study are to report our finding with respect to (1) the status and extent of Hg in lichens and mosses in the U. S. arctic and (2) the recent chronology of Hg flux to Arctic and subarctic lakes.

2. Materials and Methods

Environmental samples were collected as part of the U. S. Environmental Protection Agency's Arctic Contaminant Research Program (ACRP). The ACRP was designed to evaluate the current status, chronology and potential ecological effects of a broad suite of contaminants including semivolatile organic compounds, trace elements, heavy metals, and radionuclides (Landers *et al.*, 1992) in the U. S. arctic. The data reported here are from samples collected in Alaska from 1990 to 1993. Sample matrices include vegetation (lichens and mosses) and lake sediments. The ACRP was designed to be an integrated research program. Therefore, at several sites multiple sample matrices, with the addition of fish, ground squirrels, and soils, were collected to investigate relationships between various potential sinks or pools of contaminants within individual watersheds. The specific sampling and analytical approaches for the matrices reported here are described below.

2.1 VEGETATION

We used lichens (*Cetraria cucullata* and *Masonhalea richardsonii*) and mosses (*Hylocomium splendens* and *Racomitrium lanuginosum*) to study the status and extent of our target contaminants (Ford *et al.*, 1992) in 23 arctic locations (Figure 1). Sites were selected across arctic Alaska and were located in habitats that were common to the surrounding landscape. Lichens and mosses are non-vascular plants and generally lack the capacity for active uptake from soil solutions. Mineral nutrition is primarily derived from atmospheric sources; thus, lichens and mosses are considered to be good choices for defining exposure of terrestrial habitats to atmospherically derived substances, especially in remote regions with logistic constraints (Hale, 1981). Not all species were present at all sampling locations; at each site we collected as many species as were available. To minimize exposure to the samples from other than atmospheric sources of Hg, vegetation sampling sites were selected to be geomorphologically "independent" whenever possible, meaning that sampling sites were located on knolls, saddles, and hilltops with little or no drainage into the sampling sites.

Vegetation samples for Hg analysis were collected over areas of ca. 50 to 200 m². Collections were made using plastic or stainless steel forceps and powderless latex surgical gloves into washed (distilled, deionized water) plastic containers or Kapak™ metalized polyester bags. Samples were picked clean of visually obvious debris and transferred to pre-cleaned polyester mesh bags and dried to ambient moisture conditions in the field. Drying sites were remote from roads, generators and other potential point sources of Hg. Dried vegetation samples were placed into individual Ziploc™ or Kapak™ bags and kept cool in insulated containers and in the dark until they could be shipped refrigerated via overnight courier to the analytical laboratory. Field storage of samples sometimes lasted for up to three weeks due to the remote field locations.

In the laboratory, samples were either oven dried at 35°C or transferred to sealed Spex™ jars in a Class 100 laminar flow hood and then freeze dried and homogenized with a Spexmixer™ mill or Spex™ ball mill. 1992 samples were digested with nitric/perchloric acid (Baker™ Instra-analyzed ≤ 0.01 ug/g total Hg) in an acid-cleaned Teflon™ bomb gently

warmed for 6-8 h followed by oven heating at 130^o C for 4 h. 1990 samples were digested by nitric/perchloric acid digestion followed by hydrofluoric acid digestion (all acids were Fisher™ <0.004 ug/g total Hg). Samples were analyzed with an atomic absorption spectrophotometer using cold vapor atomic absorption (CVAA). Method detection limits (MDL) for Hg were 0.04 μg/g dw (1990) and 0.001 μg/g dw (1992).

Accuracy, precision, contamination, and recovery were assessed using standard reference materials (SRMs), field and laboratory duplicates, reagent blanks and spiked blanks and matrices. SRMs were either NIST 1572 (orchard leaves), NIST 1547 (peach leaves) or NIES 9 (Sargasso). Blanks were always negligible relative to sample values. Performance on quality control samples regularly fell within ± 20%, a reasonable envelope when operating close to the method detection limit. Duplicate analyses usually varied by no more than ±7%.

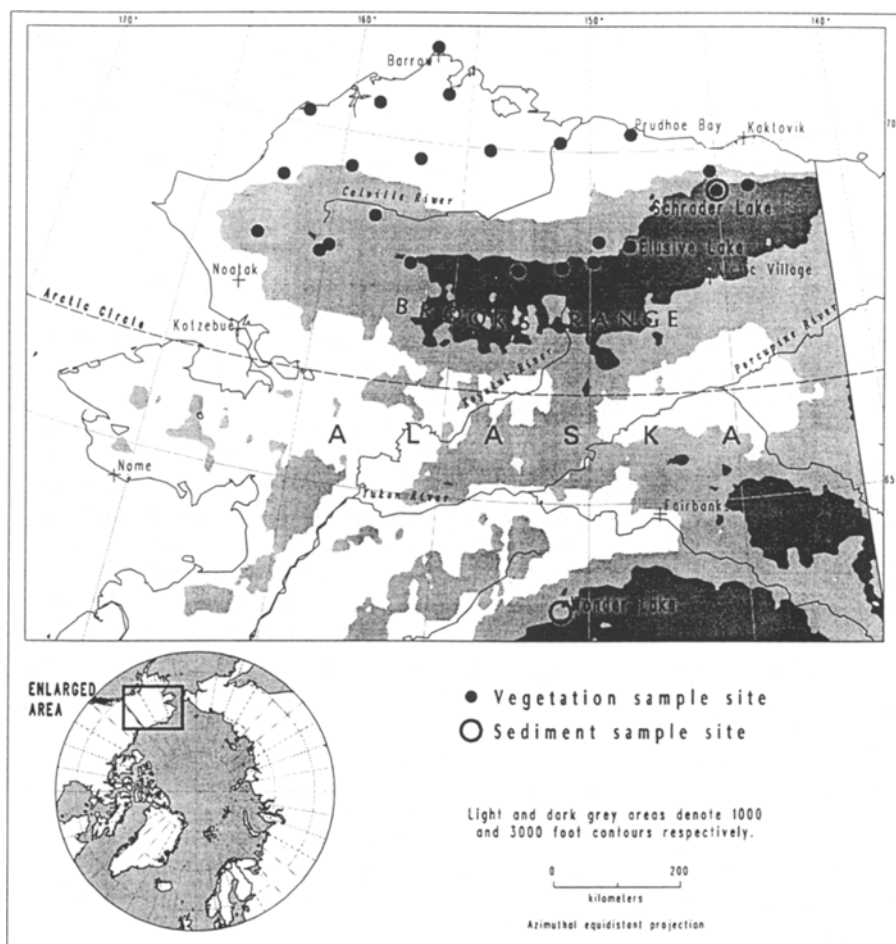


Fig 1. Map of Northern Alaska showing the locations of vegetation and lake sites sampled by the U. S. EPA Arctic Contaminants Research Program during 1990 to 1993.

2.2 SEDIMENT

Lake sediment was obtained from the deepest basin of subarctic Wonder Lake (63° 28' N, 150° 52' W, Denali National Park and Preserve) and arctic Schrader Lake (69° 22' N; 144° 60' W, Arctic National Wildlife Refuge) (Figure 1) through the ice in the spring (April/May) of 1991. Both lakes are large glacial systems with gradual bathymetric characteristics suggesting stable sediments and regular sediment accumulations through the past 150 years (Wilding, 1940; Hobbie, 1961, 1962; Werner *et al.*, 1990). There are minimal current influences of man in the watersheds of the two lakes and access is limited.

We used a stainless steel gravity coring device with Plexiglas™ core liner tubes (internal diameter of 12.7 cm). Although physical disturbance associated with gravity coring may result in erroneous data and misinterpretations (Crusius and Anderson, 1991; Cumming *et al.*, 1993), the condition of our retrieved cores showed no apparent physical disturbance resulting from the coring operation as evidenced by an intact sediment water interface, undisturbed surficial worm and insect tubes, and horizontal uniformity of sediment texture and color. Cores were extruded and sectioned at either 1 cm or 0.5 cm intervals from 0 to 10 cm (yielding approximately 125 cm³ sediment per 1 cm interval), and at 2-cm intervals from 20 to 40 cm or the maximum depth of the core (yielding approximately 250 cm³ per interval). Sediment samples were placed into commercially certified pre-cleaned glass jars with Teflon™ lid liners and kept near 4° C, but not frozen, in the field (< 4 weeks) until they could be refrigerated.

Sediment intervals were analyzed for total water and carbon content, radionuclides, and total Hg. Radiometric analysis of ²¹⁰Pb, ¹³⁷Cs, and ⁷Be were first used in a constant rate of supply (CRS) model (Robbins, 1978) to assign approximate dates of deposition to the individual sediment intervals (Gubala *et al.* 1994). Based upon this information an analytical strategy was prepared regarding the selection of sediment slices for analysis.

For total Hg analysis, 1 to 5 g of wet sediment was totally dissolved in a sealed Teflon™ pressure vessel using a sequential, two-step digestion process with high purity nitric, perchloric, and hydrofluoric acids. Hg concentrations in the extract were then quantified using CVAA. Analysis of internal standard solutions, matrix recovery spikes, and standard reference materials were used to provide calibration and quality assurance for each analytical batch. Replicate analyses were conducted on 10% of the samples to assess precision of the analytical technique. Duplicate analyses varied by no more than 5%.

3. Results and Discussion

3.1 TOTAL Hg IN LICHENS AND MOSSES

Hg was present in all vegetation samples, but generally in low concentrations. Among the four species sampled, median concentrations of total Hg were similar, ranging from a low of 0.043 µg/g dw in *C. cucullata* to 0.056 µg/g dw in *M. richardsonii*. These median values were very close to the analytical method detection limit (MDL) of 0.04 µg/g dry wt. The "feather moss," *H. splendens*, that is used as a biomonitoring tool in Nordic countries (Rühling *et al.*, 1987, 1992; Steinnes *et al.* (in press)), had a median value of 0.055 µg/g dw; not significantly different from the median total Hg concentration in *M. richardsonii*. The highest concentration found among our four target sampling species, was 0.112 µg/g dw, in *R. lanuginosum* and *H. splendens*, with slightly higher concentrations

(0.146 to 0.156 $\mu\text{g/g dw}$) observed in the non-target taxon *Umbilicaria hyperborea*. Site specific studies show that even these low levels of Hg in our target monitoring species were higher than Hg concentrations in blueberries (*Vaccinium uliginosum*) and in a ground squirrel forage food (*Dryas octapetala*) (Ford *et al.*, in press).

While lichens and mosses are widely used in biomonitoring efforts worldwide, particularly in the Arctic (Thomas, 1986; Rühling, 1987, 1992), analyses of total Hg are rare. Steinnes *et al.* (1993) report total Hg concentrations of 0.15 to 0.32 $\mu\text{g/g dw}$ for 11 samples *H. splendens* collected from throughout Norway as part of a laboratory methods intercomparison. The highest concentration of total Hg in our *H. splendens* from the Alaskan arctic (0.112 $\mu\text{g/g dw}$) is lower than the lowest Norwegian value (Steinnes *et al.* 1993). A bulk sample of *R. lanuginosum* from Svalbard had a total Hg content of 0.167 $\mu\text{g/g dw}$; *Cetraria nivalis* from the same arctic location had a concentration of 0.113 $\mu\text{g/g dw}$ (Drbal *et al.*, 1992). Both of these samples had higher total Hg than any of our lichen and moss samples. Total Hg concentration in the epiphytic lichen, *Parmelia sulcata*, is reported for two surveys, 1982-83 and 1986-87, conducted in the Netherlands (Sloof and Wolterbeek, 1992); concentrations ranged from 0.1 to 3.7 $\mu\text{g/g dw}$ in 1982-83, and 0.1 to 3.6 $\mu\text{g/g dw}$ in 1986-87. Means were 0.4 and 0.5 $\mu\text{g/g dw}$ in 1982-83 and 1986-87, respectively. Our minima concentrations were as low as the minima reported for the Netherlands but our mean concentrations were an order of magnitude less.

Even though total Hg in lichen and moss samples from this study were generally low, sampling sites closer to the ocean appeared to have higher concentrations of total Hg (Table I). Distance from the coast was determined by measuring the shortest distance from each sample site to the marine coastline. For 3 of the 4 species the relationship between total Hg and distance to the coast was significant at the 0.05 level. The strongest relationship was for the lichen *M. richardsonii* which demonstrated a very significant regression ($P < .001$) explaining over 40% of the variation in the data. The poorest relationship was that for *R. lanuginosum*, for which only 14% of the variation in the data was explained.

Table I
Regressions Statistics: Total Hg vs Distance from the Coast

Species	intercept	slope	Std. Error of the slope	R Square	Significance	No. of Observations.
<i>C. cucullata</i>	0.059	-0.0001	0.00005	0.29	.002	30
<i>H. splendens</i>	0.077	-0.0002	0.00005	0.35	<.001	36
<i>M. richardsonii</i>	0.068	-0.0002	0.00005	0.43	.001	20
<i>R. lanuginosum</i>	0.062	-0.0001	0.00007	0.14	.066	25

We offer four hypotheses regarding the potential source or cause of coastal Hg in arctic Alaska: ocean water containing ionic forms of Hg (e.g. HgCl_4^{-2} , $\text{HgCl}_3\text{Br}^{-2}$, and HgCl_3^{-}) distributed inland via windborne marine aerosols; lithologic sources associated with the coastal plain; industrial emissions resulting from the petroleum and natural gas industries located near Prudhoe Bay and Barrow, respectively; and temperature-mediated accumulation due to colder temperatures near the coast (Steinnes and Andersson, 1991).

Bering Sea water has been reported to contain 7.0 ± 3.3 ng/L of total Hg (Fowler, 1990). It is well documented for coastal regions that, in general, there is a strong marine chemical "signal" that can be detected inland from the coastlines some 10 - 100 km (Mairs, 1967; Sullivan, *et al.*, 1988; Eilers, *et al.*, 1993). Thus, marine systems may provide a

consistent total Hg signal within the study area that is negatively associated with distance from the coast. The geology of the coastal plain of the Alaskan arctic differs significantly from the interior Brooks Mountain Range and foothills, possibly resulting in elevated Hg in vegetation. The causal factors responsible for the slightly elevated, but highly significant, total Hg concentrations in coastal vegetation may be a combination of these explanations. The temperature-mediated Hg accumulation hypothesis that seems tenable for the Norwegian situation (Steinnes and Andersson, 1991) appears not to apply to Alaska. Mean winter air temperature for arctic Alaska is warmer toward the west coast, decreasing to the east (National Foreign Assessment Center (U. S.) 1981). A similar, but less pronounced pattern is observed for mean summer temperatures. Large scale industrial activities in arctic Alaska are limited to oil and gas exploration and pumping, and localized mining. These activities have some atmospheric emissions and are mostly located around Prudhoe Bay and Barrow. If Hg was associated with these activities the relationship would not likely affect the westernmost coastal sites (Figure 1) which show the negative relationship with distance from the coast. Therefore, we suspect that the most likely hypotheses explaining these relationships are marine aerosol sources and/or geologic differences between coastal and interior sampling locations.

3.2 TOTAL Hg IN SEDIMENT

A primary objective of the stratigraphic studies in the two Alaskan lakes was to compare the concentrations of total Hg in recent vs preindustrial sediments. We also wanted to compare results for these lakes to those reported from other Arctic and lower latitude lakes in order to evaluate the relative status of Hg flux to the watersheds. Cores from the two lakes were similar with regard to the distribution of radionuclides used for dating. The entire inventories of ^{137}Cs and unsupported ^{210}Pb were found in the top 5 to 7 cm of the cores' and ^{210}Pb decreased at a uniform rate with sediment depth (Gubala *et al.*, in press). This suggests that the rate of sediment accumulation in the last 150 years has been constant and that the beginning of the 6 cm sediment interval in both cores is roughly equivalent to 145 years before present, or about 1845. Sediment deposited below the 5 cm interval is considered to represent pre-industrial conditions with regard to Hg loading. After correcting for focusing factors of 1.4 and 1.5, the average rates of sediment accumulation over the past 145 years are approximately 11 and 55 $\text{g}/\text{m}^2/\text{yr}$ for the Wonder Lake and Schrader Lake cores, respectively (Gubala *et al.*, in press). Figure 2 shows the accumulation rate of total Hg for both Wonder and Schrader Lakes.

Accumulation rates of total Hg were lower in Wonder Lake, ranging from a high of 1.64 $\mu\text{g}/\text{m}^2/\text{yr}$ at the 2 cm interval to a low of 1.07 $\mu\text{g}/\text{m}^2/\text{yr}$ at the 5 cm interval. These flux estimates are less than one half the background or pre-industrial estimates reported for lakes in the north central USA (3.7 $\mu\text{g}/\text{m}^2/\text{yr}$, Swain *et al.*, 1992) and arctic and subarctic Canadian lakes (mean 4.15 $\mu\text{g}/\text{m}^2/\text{yr}$, range 0.7 to 7.4 $\mu\text{g}/\text{m}^2/\text{yr}$, Lockhart *et al.*, this volume). Schrader Lake total Hg accumulation rates were higher (7.48 $\mu\text{g}/\text{m}^2/\text{yr}$) in the 1 centimeter interval. The lowest flux rate for Schrader Lake, 5.17 $\mu\text{g}/\text{m}^2/\text{yr}$, occurred at the 17 cm depth. The lowest Hg flux for Schrader Lake occurred at a sediment depth dated to be prior to 1845 while for Wonder Lake the lowest flux was for the period from about 1845 to 1885.

We compared mean total Hg flux rates between pre-industrialization (prior to c. 1845, depth intervals 6 to 22 cm) and post-industrialization (depth intervals 1 to 5) to create a flux ratio or enrichment factor. The Wonder Lake flux ratio was 1.08 and Schrader lake was

1.05. Both ratios suggest a slight (5 to 8%) increase in total Hg flux to the lake sediments during the last 145 years. Based upon the variance of the pre-1845 total Hg accumulation rates, recent flux ratios become statistically significant at the 95% confidence level (one-tailed Student's t test) at 1.15 (Wonder) and 1.08 (Schrader). Flux ratios for increments 1, 2 and 3 cm in Wonder Lake were 1.21, 1.21 and 1.19 relative to the downcore average mercury flux of $1.36 \mu\text{g}/\text{m}^2/\text{yr}$. Flux ratios for increments 1 and 2 cm in Schrader Lake were 1.24 and 1.10 relative to the downcore average flux of $6.05 \mu\text{g}/\text{m}^2/\text{yr}$. Each of these five increments is statistically different ($P \leq .05$) from the downcore background (pre-industrial) flux. The average increase in total Hg accumulation over pre-industrial background during the last 40 years (increment 1) is remarkably similar between the two lakes, 21 and 24% in Wonder and Schrader Lakes, respectively. This is true even though the average preindustrial Hg flux in the two lakes differed by over a factor of 4. These small but similar increases in Hg flux to the two lakes in the surficial sediments indicates that there is a consistent, regional scale increase in the rate of total Hg atmospheric deposition. Since the formidable elevational barrier of the Brooks Range lies prominently between these two lakes (Figure 1) which are located over 600 km apart in a North - South direction, the consistent rate of total Hg flux in the surficial sediments is even more interesting.

Mercury Accumulation Rates

$\mu\text{g}/\text{m}^2/\text{yr}$

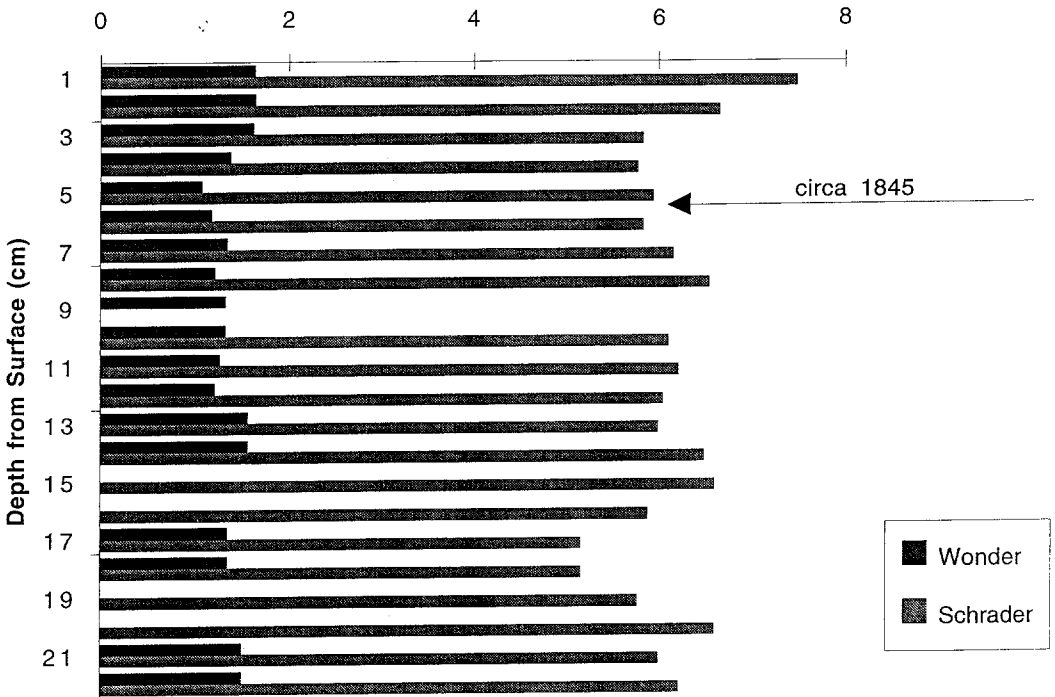


Fig. 2. Depth profiles of total Hg accumulation rates in lake sediment cores from Schrader and Wonder lakes in Alaska.

The flux ratios of total Hg to 7 lakes reported by Swain *et al.* (1992) ranged from 3.22 to 4.88, representing a three to five fold increase in total flux of Hg for these North American, mid-latitude (45°- 49° N) lakes. Lockhart *et al.* (this volume) reports flux ratios for Canadian arctic and subarctic lakes ranging from a low of 1.1 in Lac Belot to a high of 7.0 in Hawk Lake (mean of 8 lakes = 2.98). Our reported increase in total Hg flux in recent decades to the Alaskan lakes is consistent with, although of considerably lower magnitude than, increases in total Hg flux reported for arctic and temporal lakes in other countries (Fig 3). Swedish forest lakes demonstrate a flux ratio from about 2 in the most northerly, arctic lakes, to about 5 in the lakes located in southern Sweden (Johansson, 1983). Finnish lakes show a similar latitudinal gradient (Verta *et al.* 1990) with ratios ranging from 6.3 (range 0.7 to 15.7) in southern Finland to 2.9 (range 1.0 - 5.6) in northern Finland.

Increased Hg flux to lakes is very often associated with acidic deposition (Verta *et al.*, 1990; Grieb *et al.*, 1990; Winfrey and Rudd, 1990; Haines *et al.*, 1992). The two Alaskan lakes have circumneutral pH and acid neutralizing capacity greater than 100 µeq/L (Landers and Gubala unpublished) and are located in a geographic setting remote from any large regional sources of acidic precursors. These factors suggest that acidic deposition is not currently a significant environmental risk to these systems. We hypothesize that the slight increase in Hg loadings to the two Alaskan lakes is more closely associated with an increase in the global or at least northern hemisphere background concentrations of Hg due to increases in global air contamination by trace metals (Nriagu and Pacyna, 1988). There is ample evidence for transpolar transport of metals associated with industrial activities in Europe and Asia (Landsberger *et al.*, 1992). Although Hg concentrations were not reported by Landsberger *et al.* (1992), Hg is a known by-product of coal combustion and smelting activities that are common in Eurasia and is probably associated with and transported to the Arctic in the gas phase by the northern hemisphere air masses described. Hg, due to its potential for revolatilization after deposition may be affected by a "global fractionation" process (Wania and Mackay, 1993) resulting in the colder arctic regions acting as a sink for Hg that reaches these most northern destinations (Steinnes, this volume).

The transfer of Hg from terrestrial to aquatic systems depends on several factors. Large watershed to lake area ratio, low pH, low ionic strength, and high dissolved organic carbon all favor the flux of Hg into lake basins (Bodaly *et al.*, 1993; Engstrom *et al.*, 1994). Within-lake re-mobilization and bioaccumulation of Hg is also be affected by sediment redox conditions, primary and secondary production, basin morphometry, water temperature, and the concentrations of Zn and Se (Björnberg *et al.*, 1988; Winfrey and Rudd, 1990; Bodaly *et al.*, 1993). Hence, it is difficult to judge, *a priori*, the degree to which a relatively small amount of Hg deposited within the watershed of a freshwater system will appear in the associated food web.

An important consideration in the Arctic with respect to ecosystem Hg is the vast expanses of landscape containing shallow, freshwater thermokarst lakes and wetland systems moderately high in organic compounds. The aggregate Hg in these systems may be considerable and a portion could be mobilized if future climate change results in permafrost melting and a general increase in arctic temperatures. It is currently unclear if these systems function in the same way as more temperate freshwater systems with regard to Hg cycling. Therefore, the effect of warming on Hg in arctic ecosystems under a global warming scenario remains undetermined.

Flux Ratios of Total Mercury to Lake Sediments in the Northern Hemisphere

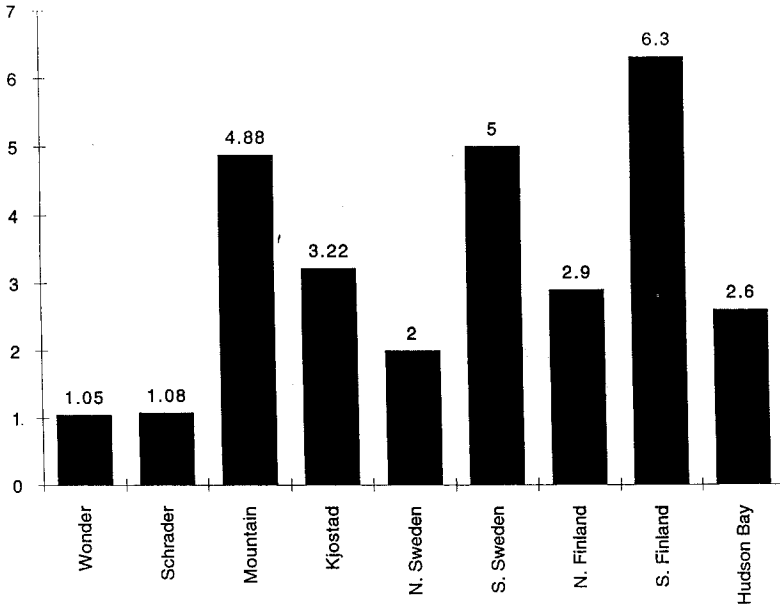


Fig. 3. Total Hg is compared for industrial vs. pre-industrial eras using flux ratios (enrichment factors). Total Hg flux ratios in lakes from throughout the northern hemisphere show a consistent decrease in flux ratios with increasing latitude. Flux ratios are printed at the top of each bar. Wonder and Schrader (this study); Mountain and Kjostad (Swain *et al.*, 1992); Sweden (Johansson, 1983); Finland (Verta *et al.* 1990); Hudson Bay (Hermanson 1993).

4. Conclusions

Total Hg concentrations in lichen and moss sampled in arctic Alaska are relative low when compared to results from similar samples collected in Norway. We found a significant, negative relationship between total Hg in vegetation and distance from the coast in Alaska. Marine aerosols and or the effect of coastal geology seem the most likely explanations of this spatial trend. There is evidence of a relatively small increase (21 to 24%) in anthropogenic contamination of Hg in arctic and subarctic Alaskan lakes in the last 40 years based on sediment analysis. The source of this increased flux to aquatic systems is probably from long range atmospheric transport of emissions from industrialized sources in Eurasia and N. America. Since global background Hg concentrations do appear to be increasing, select ecosystems in arctic Alaska should be monitored to detect changes in the rate of Hg loading.

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