Palaeolimnological investigation of atmospheric pollution in the Søndre Strømfjord region, southern West Greenland: accumulation rates and spatial patterns

Richard Bindler, N. John Anderson, Ingemar Renberg and Carola Malmquist

High-latitude ecosystems are inherently sensitive to natural environmental stress as a result of extreme seasonal variations in light and temperature, nutrient limitations, as well as other physical and chemical characteristics; consequently, these regions are quite vulnerable to the addition of pollutant stress. There is a poor understanding of spatial and temporal patterns of atmospheric pollution in the Arctic, because of the lack of monitoring stations and networks for current and past atmospheric deposition. Today, however, the Arctic is recognised as an important focus for long-range transport of contaminants, particularly from strong air flows which carry airborne pollutants from industrial regions at lower latitudes, e.g. heavy metals and persistent organic pollutants (POPs). A diverse range of anthropogenic pollutants has been shown to be present across much of the region (Aarkrog *et al.* 1997; AMAP 1998). Of particular importance are compounds, such as mercury and POPs, which present a risk to native fauna and also inhabitants. It is hypothesised for some volatile organic compounds, as well as possibly for mercury, that there may be a latitudinal fractionation that contributes to the continued mobilisation of these compounds from warmer to colder climates, where they are ultimately deposited and stored (Wania & Mackay 1993). Experimental data and limited field research support this 'cold-condensation' hypothesis, at least for some POPs (Blais *et al.* 1998).

Many areas of the Arctic, especially Greenland, are poorly represented in the scattered sampling programs that have so far been undertaken. Time-series data are even more limited, resulting in great uncertainty over the temporal trends of environmental contamination. A specific gap identified in arctic research is the lack of

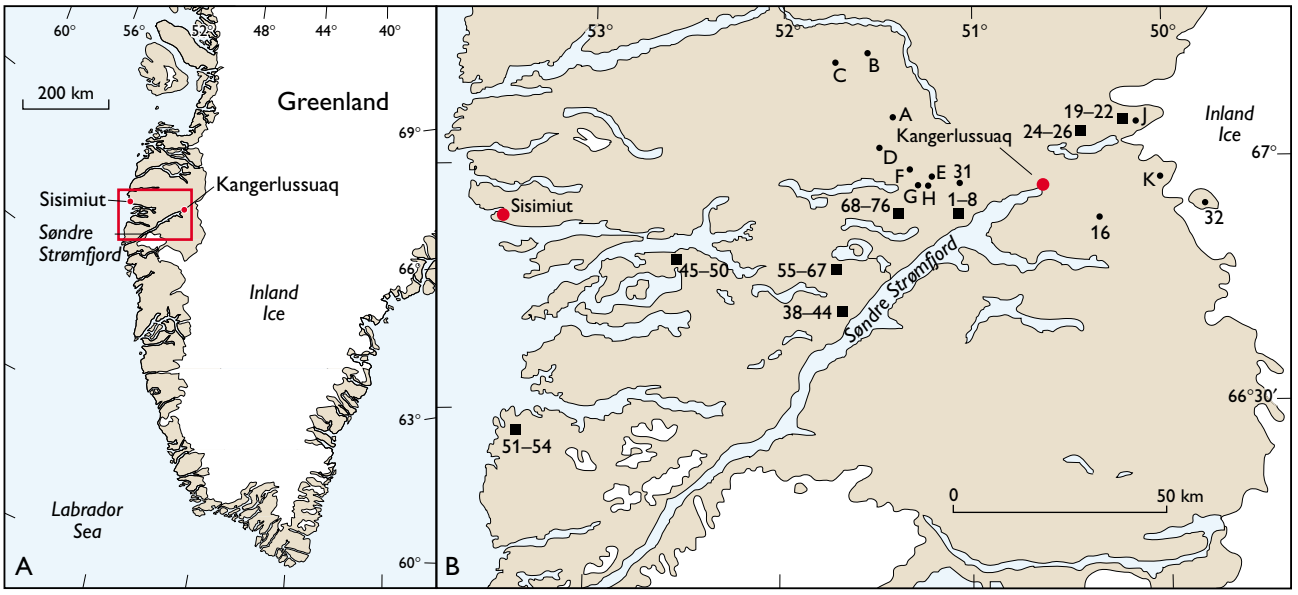

Fig. 1. **A**: Location of the Søndre Strømfjord region, southern West Greenland. **B**: The study lakes east and west of Kangerlussuaq cored in May 2000 are indicated by letters (**A–H** and **J**, **K**). Numbered lakes and groups of lakes refer to previously sampled lakes (Anderson *et al.* 1999), from which sediment samples were included in the analyses (• = individual lake; ■ = group of lakes).

Geology of Greenland Survey Bulletin 189, 48–53 (2001)

long-term retrospective time trends. A particular weakness in studies of time trends is the general assumption that natural, non-polluted conditions existed prior to the mid-1800s. Analyses of lake sediments and peat deposits in Europe (Renberg *et al.* 1994, 2000; Shotyk *et al.* 1998; Brännvall *et al.* 1999) and Greenland ice cores (Hong *et al.* 1994) have revealed an approximately 3500-year history for lead (Pb) pollution in the northern hemisphere, which contradicts the assumption of the mid-1800s representing 'background' levels.

In Greenland, studies of pollutants in the ice-free terrestrial margin have focused largely on synoptic surveys to identify and quantify pollutants in the contemporary environment. Temporal changes in pollutant deposition and accumulation in the terrestrial environment are not well established. Lake sediments provide an ideal archive to examine many anthropogenic environmental pollutants, as well as to assess the range of natural variability of compounds or elements such as lead and mercury. The Søndre Strømfjord region (Fig. 1) offers excellent opportunities for palaeolimnological research because of the lack of cultural disturbance and, more importantly, because of the extensive availability of data from limnological and palaeolimnological research programs that have been carried out in the region since 1996 (Anderson *et al.* 1999).

The long SW–NE-trending fjord around which this study is centred is known by both the Danish name, Søndre Strømfjord and the Greenlandic name, Kangerlussuaq. In this paper we follow the convention of using 'Søndre Strømfjord' for the fjord and 'Kangerlussuaq' for the airport at the head of the fjord (Fig. 1).

Main aims

The aims of the present project in the Søndre Strømfjord region of southern West Greenland (Fig. 1) are to:

1. Assess the onset of mercury and lead pollution and their accumulation rates in selected lake sediments.
2. Assess the spatial patterns and enrichment rates of mercury and lead pollution in sediments of lakes along a west to east transect in the Søndre Strømfjord region employing surface and bottom samples from short cores of recent sediments (*c.* 15–35 cm in length).
3. Supplement archived sediment samples with the collection of new lake sediment cores along an altitudinal transect (*c.* 150 to 950 m above sea level) and in lakes adjacent to the ice margin to assess the hypothesis of 'cold condensation', i.e. the progressive revolatilisation of organic pollutants from warmer to colder climates.
4. Establish which persistent organic pollutants (POPs) are present in recent lake sediments and assess temporal changes in their accumulation.

In the case of the first two aims, sediment samples collected in previous field campaigns during 1996–1999 can be used. Field work in 2000 was initiated specifically to collect fresh sediment samples for POP analyses and to address the 'cold condensation' hypothesis.

Temporal changes in atmospheric pollutants

Analysis of lake sediment cores is a useful method of documenting the historical loading of atmospheric pollutants to remote areas, such as mercury, lead and organic pollutants. These pollutants derive from the combined effects of direct deposition to the lake surface and run-off from the catchment area. Palaeolimnological researchers have used lake sediments to reconstruct site-specific as well as regional pollution histories in order to determine the onset of atmospheric pollution, the rates

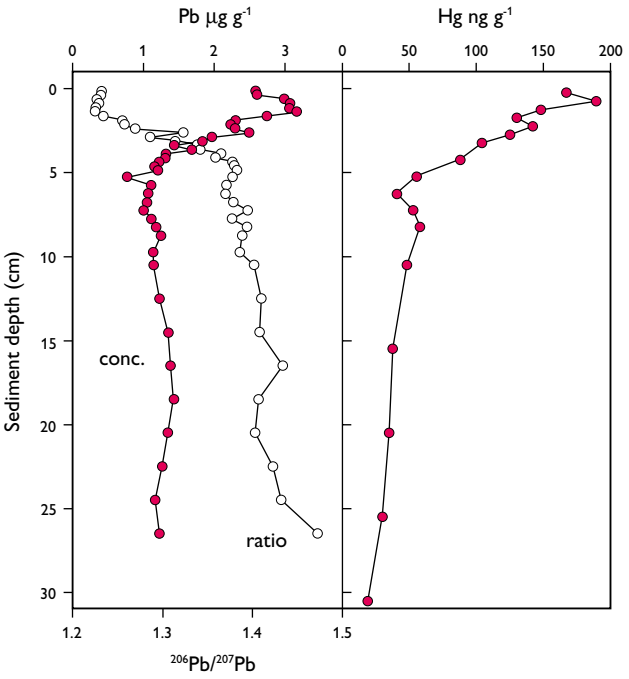

Fig. 2. Profiles of lead (Pb concentrations and $^{206}Pb/^{207}Pb$) and mercury (Hg) in sediment cores from Lake 32 (Nunatak lake) (Bindler *et al.* 2001a, b).

of increase of individual pollutants and spatial patterns of deposition. Landers *et al.* (1998) summarised the most reliable mercury reconstructions from lake sediment cores from arctic and boreal ecosystems to evaluate spatial patterns at high latitudes on a broad scale.

Analyses of mercury and lead in ^{210}Pb-dated cores from a few lakes in the Søndre Strømfjord area (Lakes 16, 32, 53 and 70) show patterns generally consistent with data from elsewhere in the Arctic and the boreal zones. Cores from all four lakes show clear evidence of pollution from industrial sources, with enhanced lead and mercury concentrations and accumulation rates in near surface sediments relative to deeper sediments (Fig. 2). In the case of mercury, analysed using cold vapour atomic fluorescence spectrometry (CVAFS), the sediment records indicate increased mercury accumulations of about two to three times that determined for background sediment layers (Bindler *et al.* 2001b). The current and historic mercury accumulation rates in the Greenland lakes are in the same range as those reported for lakes from the Canadian Arctic, where comparable lakes had mercury accumulation rates of 1–5 μg m^{-2} yr^{-1} in deeper sediments, which increased to 5–8 μg m^{-2} yr^{-1} in surface sediments (Landers *et al.* 1998). The Greenland sediments indicate an initial increase in the mercury accumulation rate during the mid-1800s (Bindler *et al.* 2001a), which is generally consistent with the onset of mercury pollution in North America and northern Europe. In one lake (Lake 32; Fig. 2), located on a nunatak *c.* 5 km within the ice margin, there is an indication of a slightly earlier mercury pollution signal, prior to 1800, based on extrapolation of the ^{210}Pb chronology (Bindler *et al.* 2001b).

For lead, analyses of the sediment cores in lakes around Søndre Strømfjord also included stable lead isotopes (^{204}Pb, ^{206}Pb, ^{207}Pb and ^{208}Pb). Lead isotopes make it possible to infer the provenance of the pollution lead in the environment, based on the fact that lead ores have characteristic lead isotopic compositions that are typically distinct from the isotopic compositions of soils and bedrock. Consequently, isotope analyses have become an effective method in environmental studies to quantify pollution lead contributions and to infer specific sources for the pollution lead. For the sediment cores from the Søndre Strømfjord region, there is a large decline in the ^{206}Pb/^{207}Pb ratio coincident with increased lead concentrations upwards in the cores (Fig. 2) clearly indicating that the increased concentrations are a direct result of an increasing input of pollution lead to these lakes (Bindler *et al.* 2001b). Based on an isotope mixing model, the lead pollution can be demonstrated to derive from western Europe and Russia, and not from North America (Bindler *et al.* 2001b).

Spatial patterns of Hg and Pb pollution

Analyses of sediment samples from earlier field campaigns in the Søndre Strømfjord region (individual lakes and lake groups numbered in Fig. 1) revealed some trends on the spatial pattern of atmospheric pollution. These samples represented the top (0–1 cm) and bottom (a 1 cm interval at *c.* 15–35 cm depth) of gravity cores from lakes north of Søndre Strømfjord. As expected from the steep gradient for annual precipitation, with the highest annual precipitation at the coast and declining rates inland, the unsupported ^{210}Pb fluxes and inventories in the lake sediments were higher at the coast and lower inland (Bindler *et al.* 2001b). Predictably, pollution lead inventories followed a similar pattern: higher lead concentrations and a stronger influence on the isotope composition by pollution sources on the surface lake sediments nearer to the coast together with larger pollution lead inventories in whole cores (i.e. the total amount of pollution lead accumulated in the sediment core). The close relationship between the gradient for ^{210}Pb fluxes and pollution lead inventories supports the inference that the differences between sites is largely due to variations in rainfall.

In contrast, the mercury analyses of the lake sediments, both cores and the top/bottom sample pairs, revealed an unexpected reverse trend. While the sediments generally revealed a two- to three-fold enrichment for mercury in surface sediments relative to the deeper sample, there was a tendency towards greater enrichments in the vicinity of the ice margin (Bindler *et al.* 2001a). The lakes within about 20 km of the ice-sheet margin had higher mercury concentration enrichments, from three to as much as ten times greater than the background concentration. This region corresponds approximately with the area most influenced by the meteorology of the ice margin, e.g. the katabatic winds that flow down off the ice and over the adjacent tundra, and suggests the possibility of a 'cold condensation' effect on mercury contents.

Field work in 2000

During May 2000, sediment cores were collected from eleven lakes in two areas around Kangerlussuaq. Sediment cores were collected with a gravity corer. One

Fig. 3. Sampling at Lake H in May 2000. The lake is at *c.* 150 m a.s.l., well below the snowline.

area (Lakes A–H on Fig. 1; ~ 67°10′N, 57°30′W) constituted a 35 km long transect of eight lakes along an elevation gradient from *c.* 150 m to 950 m above sea level. A second group of three lakes (a new core from Lake 21 and new Lakes J and K) were each located within a few km of the ice-sheet margin, east of Kangerlussuaq. One lake (Lake K, 67°02.6′N, 50°09.9′W) is situated immediately above an ice tongue whose meltwater drains into the head of Søndre Strømfjord.

At each lake, two to three short sediment cores were collected and sectioned in the field. One core was sectioned into 1 cm intervals and stored in glass containers for the analysis of persistent organic pollutants. A second core was sectioned into a surface sediment sample (0–1 cm), a bulk sample (9–10 cm), and a bottom 2 cm interval taken near the base of the core (the cores were *c.* 20–30 cm in length). In most lakes a third core was collected and sectioned into 0.5 cm slices from 0–10 cm, and 1 cm slices down to the base of the core, for detailed stratigraphic analyses of mercury and lead. To make assessments of bulk pollution, the background concentrations (the concentration at depth of the compound of interest, e.g. mercury) were subtracted from the concentrations in the surface and bulk samples. Mercury pollution inventories (mass per unit area) can then be compared between sites, and their differences assessed in light of the altitude of the lakes.

The eight lakes west of Kangerlussuaq (A–H) were selected as an altitudinal substitution for a climatic gradient, and to limit regional variations in geology and precipitation as much as possible. The use of this altitude-for-climate substitution can be illustrated by the situation in May 2000; the snowline was at *c.* 500 m above sea level, such that one lake was situated at the snowline, two well above and the remaining five lakes below the snowline (Fig. 3). Two of the lower elevation lakes were found to have laminated sediments and high conductivities, similar to other studied lakes in the area at the head of the fjord (Anderson *et al.* 1999).

The sediments from the second group of lakes (21, J and K) east of Kangerlussuaq were sampled to further assess the 'ice-margin' effect (Bindler *et al.* 2001a). Lake K is adjacent to the margin of the Inland Ice, while Lakes 21 and J are within 5 km. Studies of meteorological conditions at the Inland Ice margin during the Greenland Ice Margin Experiment (GIMEX) in June and July 1991 observed that the energy balance at the margin was strongly influenced by the radiative differences of the ice and the adjacent tundra (Fig. 4; Duynkerke & van den Broeke 1994). The temperature gradient from the ice to tundra in the summer produces strong katabatic winds that flow down off the ice sheet and out over the tundra. The region most strongly affected by this thermal forcing coincides geographically with the lakes that showed the highest mercury enrichments. Detailed stratigraphic analyses from the new study lakes will complement the analyses of archived samples.

Initial results of high-resolution gas chromatography/mass spectrometry (HR GC/MS) analyses of the lake sediments indicate low concentrations for polychlorinated biphenyls (PCBs) (*c.* 0.5–3 nanogram g^{-1} for total PCBs; individual compounds are measured at picogram g^{-1} levels) that are consistent with other regions of the Arctic (e.g. northern Canada). However, analy-

Fig. 4. The region along the ice margin (as here north-west of Kangerlussuaq) is strongly affected by the radiative differences of the glacier and the adjacent tundra (Duynkerke & van den Broeke 1994), which may influence processes that affect mercury accumulation and retention in lakes in this region. Air temperatures above the surface of the Inland Ice in the background of the photograph are just above freezing in the summer, whereas temperatures above the adjacent tundra (foreground) may reach 15ºC. It is this temperature gradient that periodically gives rise to strong katabatic winds flowing from east (the ice) to west (the land). As described in the text, these strong environmental (temperature, radiation) gradients from ice to land may influence mercury accumulation and retention in the lakes on the tundra. The lake in the middle foreground is *c.* 250 m across.

ses of some sediments, particularly from the lakes with laminated sediments and high-water conductivities, have been complicated by the high sulphur contents which necessitated pre-treatments. Additional improvements in these procedures have contributed to a lowering of the detection limit for persistent organic pollutants in sediment samples and reductions in sample size, which permits better temporal resolution in sediment cores. Assessment of analyses of the samples collected in May 2000 are in progress, which includes estimations of accumulation rates and cumulative inventories of PCBs and polybrominated diphenyl ethers (PBDEs), a class of widely used flame retardants. The results of the sampling in Greenland in May 2000 will be compared with those of a similar project using lake sediment cores taken in the northern Swedish mountains.

Conclusions

Ongoing research on the limnology and palaeolimnology of lakes in the Søndre Strømfjord region, which is focused largely on aspects related to climate change, has provided a solid foundation for additional studies on environmental change, such as assessments of pollution inputs. In addition to contributing to the general understanding of the distribution of pollutants in the Arctic, both in terms of the rates of deposition and their spatial distribution, analyses of lake sediments in the Søndre Strømfjord region also provide a potential for assessing climatic processes that may interact with pollutant deposition and accumulation; such as the 'cold condensation' hypothesis, which contributes to the continued mobilisation of volatile compounds to the Arctic.

Acknowledgements

The Danish and Swedish Natural Science Research Councils are gratefully acknowledged for financial support. For other help we would like to thank Simon Patrick (University College London, UK), Melanie Leng (Natural Environment Research Council, UK), Peter Appleby (Environmental Radioactivity Research Centre, University of Liverpool, UK), Ove Emteryd (Department of Forest Ecology, Swedish University of Agricultural Sciences, Umeå, Sweden), Mats Tysklind (Department of Environmental Chemistry, Umeå University, Sweden), John Munthe and Elsemarie Lord (Swedish Environmental Research Institute, Gothenberg, Sweden) and the Kangerlussuaq International Science Support (KISS) facility.

References

Aarkrog, A. *et al.* 1997: AMAP Greenland 1994–1996, 788 pp. Copenhagen: Ministry of Environment and Energy, Danish Environmental Protection Agency (Arctic Monitoring and Assessment Programme).

AMAP 1998: Arctic pollution issues, AMAP assessment report, 859 pp. Oslo: Arctic Monitoring and Assessment Programme.

Anderson, N.J., Bennike, O., Christoffersen, K., Jeppesen, E., Markager, S., Miller, G. & Renberg, I. 1999: Limnological and palaeolimnological studies of lakes in south-western Greenland. Geology of Greenland Survey Bulletin **183**, 68–74.

Bindler, R., Renberg, I., Appleby, P.B., Anderson, N.J. & Rose, N.L. in 2001a: Mercury accumulation rates and spatial patterns in lake sediments from West Greenland: a coast to ice margin transect. Environmental Science and Technology **35,** 1736–1741.

Bindler, R., Renberg, I., Anderson, N.J., Appleby, P.G., Emteryd, O. & Boyle, J. 2001b: Pb isotope ratios of lake sediments in West Greenland: inferences on pollution sources. Atmospheric Environment **35,** 4675–4685.

Blais, J.M., Schindler, D.W., Muir, D.C.G., Kimpe, L.E., Donald, D.B. & Rosenberg, B. 1998: Accumulation of persistent organochlorine compounds in mountains of western Canada. Nature **395**, 585–588.

Brännvall, M.-L., Bindler, R., Renberg, I., Emteryd, O., Bartnicki, J. & Billström, K. 1999: The Medieval metal industry was the cradle of modern large-scale atmospheric lead pollution in northern Europe. Environmental Science and Technology **33**, 4391–4395.

Duynkerke, P.G. & van den Broeke, M.R. 1994: Surface energy balance and katabatic flow over glacier and tundra during GIMEX-91. Global & Planetary Change **9**, 17–28.

Hong, S.M., Candelone, J.P., Patterson, C.C. & Boutron, C.F. 1994: Greenland ice evidence of hemispheric lead pollution 2 millennia ago by Greek and Roman civilizations. Science **265**, 1841–1843.

Landers, D.H., Gubala, C., Verta, M., Lucotte, M., Johansson, K., Vlasova, T. & Lockhart, W.L. 1998: Using lake sediment mercury flux ratios to evaluate the regional and continental dimensions of mercury deposition in Arctic and boreal ecosystems. Atmospheric Environment **32**, 919–928.

Renberg, I., Wik-Persson, M. & Emteryd, O. 1994: Pre-industrial atmospheric lead contamination detected in Swedish lake sediments. Nature **368**, 323–326.

Renberg, I., Brännvall, M.-L., Bindler, R. & Emteryd, O. 2000: Atmospheric lead pollution history during four millennia (2000 BC to 2000 AD) in Sweden. Ambio **29**, 150–156.

Shotyk, W., Weiss, D., Appleby, P.G., Cheburkin, A.K., Frei, R., Gloor, M., Kramers, J.D., Reese, S. & van der Kamp, W.O. 1998: History of atmospheric lead deposition since 12,370 ^{14}C yr BP from a peat bog, Jura mountains, Switzerland. Science **281**, 1635–1640.

Wania, F. & Mackay, D. 1993: Global fractionation and cold condensation of low volatility organochlorine compounds in polar regions. Ambio **22**, 10–18.

Authors' addresses

R.B., I.R. & C.M., *Department of Ecology and Environmental Science, Umeå University, SE-901 87 Umeå, Sweden*. E-mail: *rich.bindler@eg.umu.se*

N.J.A., *Institute of Geography, University of Copenhagen, Øster Voldgade 10, DK-1350 Copenhagen K, Denmark.*