

# Recent environmental change and human impact on Svalbard: the lake-sediment geochemical record \*



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## **Abstract**

As part of a broader investigation into recent environmental change on Svalbard, the inorganic geochemical record of six lake-sediment cores was analysed. The major temporal trends in sediment elemental composition are driven by variations in two contrasting sediment components, both derived from catchment soils: (1) mineral matter, and (2) soil organic matter (SOM), enriched in Fe and Mn oxides and heavy metals.

Two environmental impacts are recorded in most or all of the lake sediment sequences. An up-core increase in organic matter can be partly attributed to diagenetic effects, but also requires an enhanced supply of SOM relative to mineral matter. In addition, the central and southern sites all show a ca. 1970 event characterised by an enhanced mineral matter accumulation rate. This requires either an enhanced allochthonous supply or an enhanced mobilisation of littoral sediments. In either case a regional-scale driving force, such as a shift in climate, is required.

At five of the lakes the sediment heavy metal concentration profiles can be explained entirely by natural factors. However, at Tenndammen (U), situated close to the Svalbard's largest settlement at Longyearbyen, possible anthropogenic Pb enrichment is found. Comparison of observed and expected heavy metal profiles (based on Greenland ice-core data) shows that the lakes are generally too insensitive to have recorded a long-transported heavy metal pollution signal.

## **Introduction**

Growing concern about the state of Arctic environments, still amongst the most pristine in the world, has generated a need to establish base-lines for ecosystem disturbance. The establishment of such base-lines is not, however, straightforward. In Svalbard, for example, the relatively long history of coal mining (since the late nineteenth century) makes it necessary to consider the state of the environment not just at present, but also prior to recent human activities (Rose et al. 2004). With the continuing potential for fossil fuel exploitation on Svalbard (Holte et al. 1996), the establishment of base-line data for Svalbard is doubly important.

One of the principal concerns is the impact of long-transported atmospheric pollution, and possible contamination of Arctic ecosystems. Rose et al. (2004) show that significant amounts of spheroidal carbonaceous particles (SCPs) and anthropogenic organic pollutants are present in the sediment of the studied lakes. However, the picture for heavy metal pollution is made more complex by the importance of natural sources. The natural fluxes of heavy metals to lakes are controlled by biogeochemical cycling in the catchment, which is subject to alteration by both natural change and human disturbance. To understand the lake sediment record of heavy metal fluxes, it is therefore necessary to understand temporal changes in the catchment.

It is known that heavy metal fluxes to the Arctic have increased in modern times: there are marked increases in heavy metal concentration and accumulation rates in glacial ice (Boutron et al. 1995; Hong et al. 1996) since the Industrial Revolution. However, such increases are small, and it is not certain that a measurable impact will be seen in lake ecosystems. In a comparison between recent and pre-industrial lake sediments in Svalbard, Rognerud et al. (1998) found that only Hg was significantly enriched above natural sediment

concentrations. Pb concentrations were found to be marginally higher in the core tops, but not to any significant degree. It is clear from this that any heavy metal enrichment of recent lake sediments in Svalbard will be very weak, and that great care must be taken to allow for natural fluctuations.

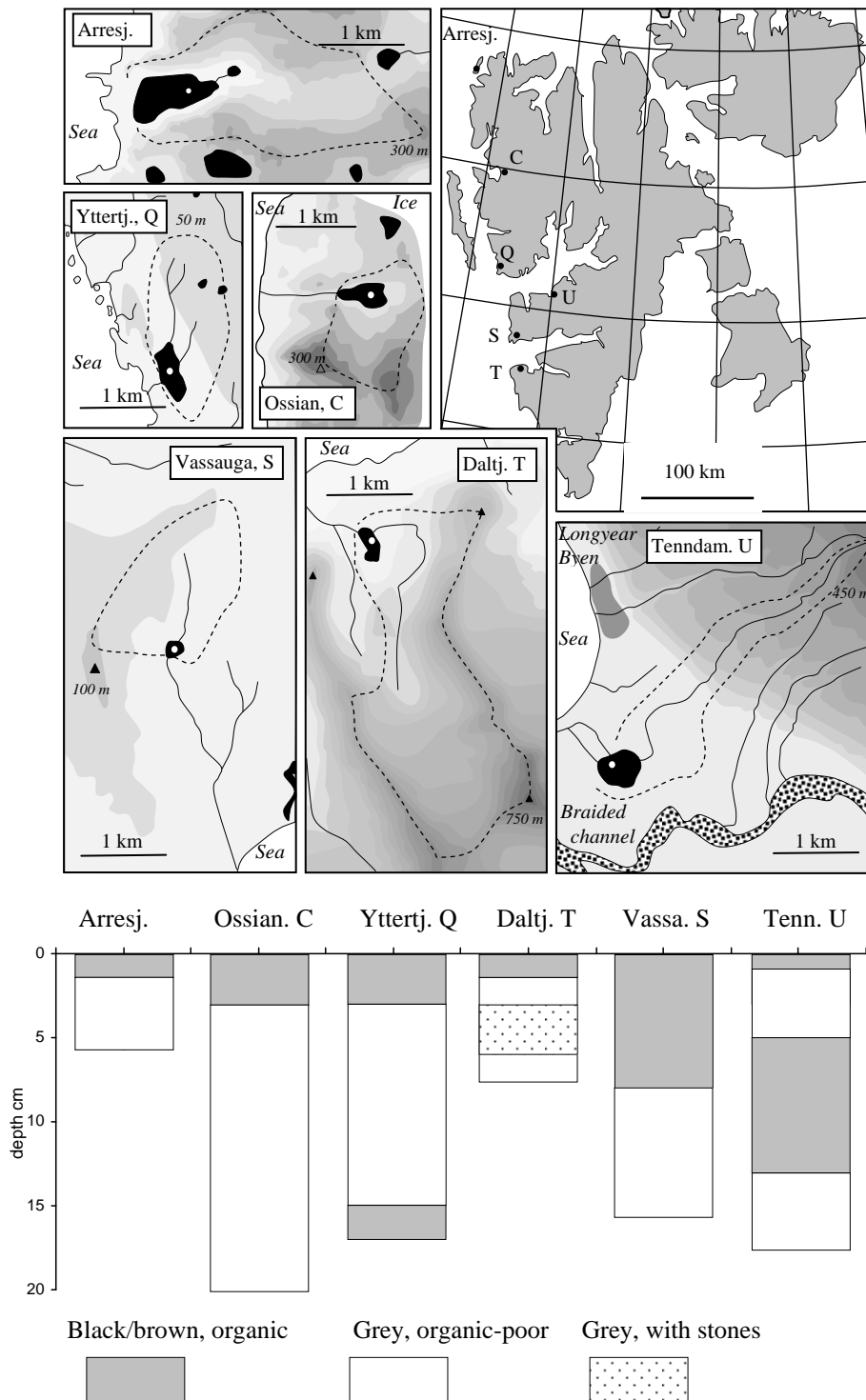


Figure 1. Site maps and basic core descriptions. These site maps show the lakes and streams (solid lines), topography (50 m contour intervals, with the highest contour interval labelled), approximate catchment boundaries (dashed line), and approximate coring location (white circle), always at the deepest point in the lake.

It has long been established that lake sediment sequences in the industrialised parts of Europe and North America can provide a quantitative record of atmospheric pollutants (e.g.

Crececius and Piper 1973; Hamilton-Taylor 1979; Rippey et al. 1982; Norton and Kahl 1991). It has also been shown that lake-sediment metal concentrations can provide unbiased estimates of the atmospheric metal deposition fluxes (Boyle and Birks 1999). However, such techniques are more problematic in regions where the atmospheric contamination signal is small compared with both the natural flux from the catchment and within-lake recycling fluxes. Within-lake cycling, which can lead to short-lived heavy metal enrichment of the surface sediment (e.g. Morfett et al. 1988; Boyle et al. 1998; Boyle 2001a), is a particularly severe problem at sites with low sediment accumulation rates. If the sedimentation rate is so low that the pollution signal is confined to the top 1 - 2 sampling intervals, then recycling contributions cannot be distinguished from pollution in the absence of isotope data. This situation is illustrated by a study from arctic Canada (Cornwell 1986), which shows enhanced heavy metals concentrations in the top two sampling intervals. A diagenetic interpretation is proposed, but with only two data points both within-lake cycling and atmospheric contamination are plausible explanations. This problem can be avoided, at least under favourable circumstances, if the sediment is sampled at finer intervals. For example, a fine resolution palaeolimnological assessment of heavy metal pollution in Lake Baikal successfully isolated a weak anthropogenic signal from a number a natural contributions (Boyle et al. 1998). A fine resolution study in Greenland also finds a pollution signal recorded in the total Pb concentration, in this case confirmed by the Pb stable isotope signature (Bindler et al. 2001).

Sediment heavy metal concentration profiles were measured for six lakes located along the western margin of Svalbard (Figure 1). A full geochemical analysis was undertaken to look at possible impacts of changing catchment sources, diagenesis, and atmospheric contamination. Atmospheric contamination is evaluated by comparison with spheroidal carbonaceous particle concentration profiles (Rose et al. 2004), and with data from Greenland ice cores (Boutron et al. 1995; Hong et al. 1996).

Table 1. Site and catchment details

Lake name and code	Lat.	Long.	Catchm. area km <sup>2</sup>	Lake area km <sup>2</sup>	Catchm./lake area ratio	Depth (max.) m	Depth (mean) m	Water pH
Ossian Sarsfjellet (C)	78° 57'	12° 28'	1.8 ± 0.2	0.13	13.8	26	11	8.0
Ytertjørna (Q)	78° 13'	12° 56'	1.5 ± 0.6	0.14	10.7	2.6	1.2	7.9
Vassauga (S)	77° 45'	13° 57'	1.8 ± 0.7	0.037	49	1.3	0.8	8.2
Daltjørna (T)	77° 33'	14° 13'	4.9 ± 0.5	0.054	91	10.5	7.4	8.1
Tenndammen (U)	78° 06'	15° 02'	2.1 ± 0.2	0.15	14	2.5	1.5	7.1
Arresjøen (Arsj)	79° 40'	10° 51'	3.1 ± 0.5	0.34	9.1	29	18	6.2

(Abbreviations: Lat. = latitude; Long. = longitude; Catchm. = catchment; Max. = maximum)

Table 2. Catchment geology

Lake name and code	Geology; Geological map 1:500 000, Svalbard, sheets 3G and 1G. Norsk Polarinstitut.
Ossian Sarsfjellet (C)	Lower Hecla Hoek, Late Precambrian, gneiss/migmatite complex.
Ytertjørna (Q)	Wedel Jarlsberg Land Supergroup, Daudmannsodden Gr., late Proterozoic basement complex.
Vassauga (S)	Bellsund Gr., late Precambrian, diamictite + Billefjorden Gr., early Carboniferous; fluvial clastic rocks + recent beach deposits
Daltjørna (T)	Bellsund Gr., late Precambrian, diamictite
Tenndammen (U)	Recent marine deposits + Van Mijenfjorden Gr., Palaeocene-Eocene, sandstone, shale and coal seams.
Arresjøen (Arsj)	Lower Hecla Hoek, Late Precambrian, gneiss/migmatite complex.

(Abbreviations: Gr. = Group)

## Methods

### *Site descriptions and catchment analysis*

All the lakes except Arresjøen were sampled in July and August 1995 using a Glew (1989) corer. Arresjøen (Arsj) was sampled using a Glew corer in July 1993. All cores were divided in the field and stored in polythene bags. Daltjørna (T) and Tendammen (U) were divided into 1 cm intervals, while the remainder were divided into 0.25 cm intervals.

The sediments of the lakes mainly comprise more or less organic fine-silty clay with some sand, and very high water concentrations (typically > 90%). Such material does not lend itself to detailed lithological description. Basic field descriptions of the sediment are shown in Figure 1.

Lake and catchment areas were estimated from 1:100 000 topographic maps (1995 editions, Norsk Polarinstitut). Catchment geology was estimated using 1:500 000 geological maps (1982 edition for northern sheet (Hjelle and Lauritzen 1982), and 1993 edition for southern sheet, Norsk Polarinstitut). Catchment and lake data are presented in Tables 1 and 2.

The lakes and their catchments are described in detail elsewhere (Birks et al. 2004). They are 5 - 85% vegetated, mainly lichen- and moss-tundra. Bare rock and unvegetated soil make up the remainder, and are most significant at Daltjørna (T) (95%) and Vassauga (S) (50%). Direct human impacts are present at Tendammen (U), but undetectable or negligible at the other sites. At Tendammen a pump house supplied water to the settlement of Colesbukta (abandoned by 1988), which housed coal-mine workers. The mine, closed in 1967, did not drain into the lake.

### *Elemental analysis*

Water content and loss-on-ignition at 550°C (LOI) were measured at University College London. LOI provides a measure of organic matter concentration, which overestimates organic matter where values are lower than 100 mg g<sup>-1</sup> (e.g. Mackereth 1966). However, organic matter trends are faithfully reproduced, and for this analysis organic matter was assumed to be equal to LOI. All elemental analyses were performed at the Department of Geography, University of Liverpool. Sediment from Arresjøen (Arsj) was analysed earlier using a different procedure: total elemental concentrations were determined by atomic absorption spectrometry (AAS) on total digests using HF, HNO<sub>3</sub>, HClO<sub>3</sub>, and HCl using a Unicam 939 Atomic Absorption Spectrophotometer. For all other samples, two methods were used. Extractable Cu, Fe, Mn, Pb, and Zn were determined using AAS. Dried samples (0.25 g) were extracted with 8 ml 10% AnalR hydrochloric acid. Accuracy was checked using NIST SRM 2704 Buffalo River Sediment. Total Si, Ca, K, and Ti were determined by X-ray fluorescence spectrometry (XRF) using a Metorex Xmet920 X-ray fluorescence spectrometer, with <sup>55</sup>Fe and <sup>109</sup>Cd isotope sources. Deconvolution of the spectra, mass attenuation correction, and calibration were achieved using the program DECONV (Boyle 2000).

### *Spheroidal carbonaceous particles (SCPs)*

Sample preparation and analysis followed Rose (1994) and Rose et al. (2004). In brief, sequential attack using HNO<sub>3</sub>, HF, and HCl removed most of the sediment, leaving a suspension dominated by carbonaceous material. An aliquot was transferred to a cover slip and counted under a light microscope at x 400 magnification.

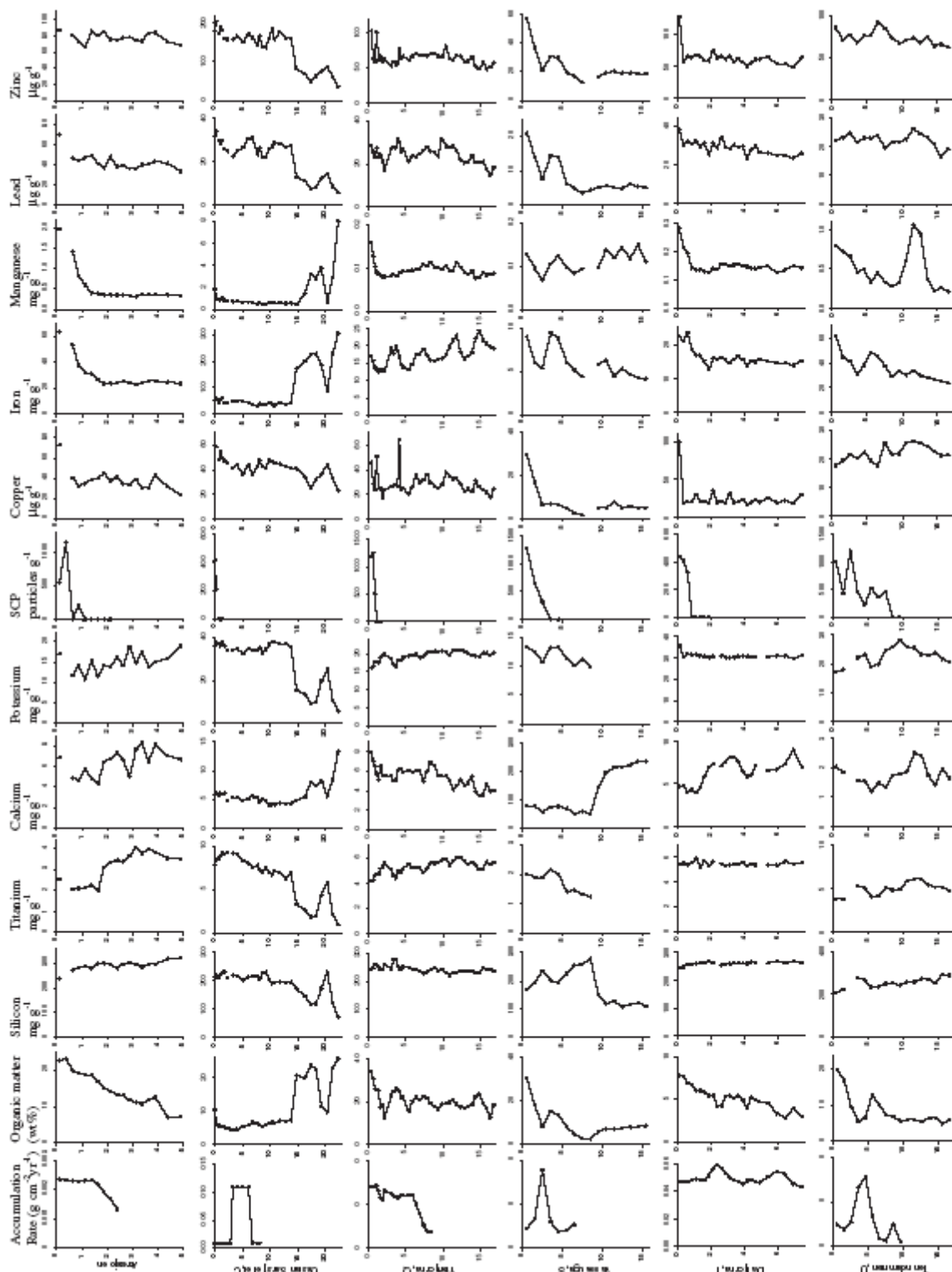


Figure 2. Depth profiles of elemental concentrations. The major elements are total concentrations measured by XRF. The trace elements are acid extractable concentrations, except for Arresjeen where they are total concentrations.

### Dating

The cores were analysed for  $^{210}\text{Pb}$ ,  $^{226}\text{Ra}$ , and  $^{137}\text{Cs}$  by direct gamma assay in the Liverpool University Environmental Radioactivity Laboratory, using Ortec HPGe GWL series well-type coaxial low background intrinsic germanium detectors (Appleby et al. 1986). The datings are described in detail by Appleby (2004).

*Numerical methods*

A principal component analysis (PCA) of the elemental concentration data was performed using CANOCO (ter Braak 1990). Association of the heavy metals with the mineral elements was tested using redundancy analysis (RDA) with forward selection of predictor variables (also using CANOCO).

The presence of anthropogenic heavy metal contamination was tested using a modified version of the method of Hilton et al. (1985), who demonstrated that the present-day natural supply of some heavy metals could be predicted from the Mg concentration using a simple regression on pre-industrial sediment. Whereas the principal objective of Hilton et al. (1985) was to determine the base-line, the procedure is equally suitable for testing whether an anthropogenic component is present. An anthropogenic component weakens the link between heavy metals and the remaining sediment. If it is present, then a model fit between the heavy metals and the other sediment parameters should strengthen as recent layers are progressively excluded from the model. If no such pattern is observed then it can be argued that no significant anthropogenic component is present. Our procedure also differs from that of Hilton et al. (1985) in that we modelled heavy metals on the major element concentrations using RDA and then tested for statistical significance by Monte Carlo permutation tests.

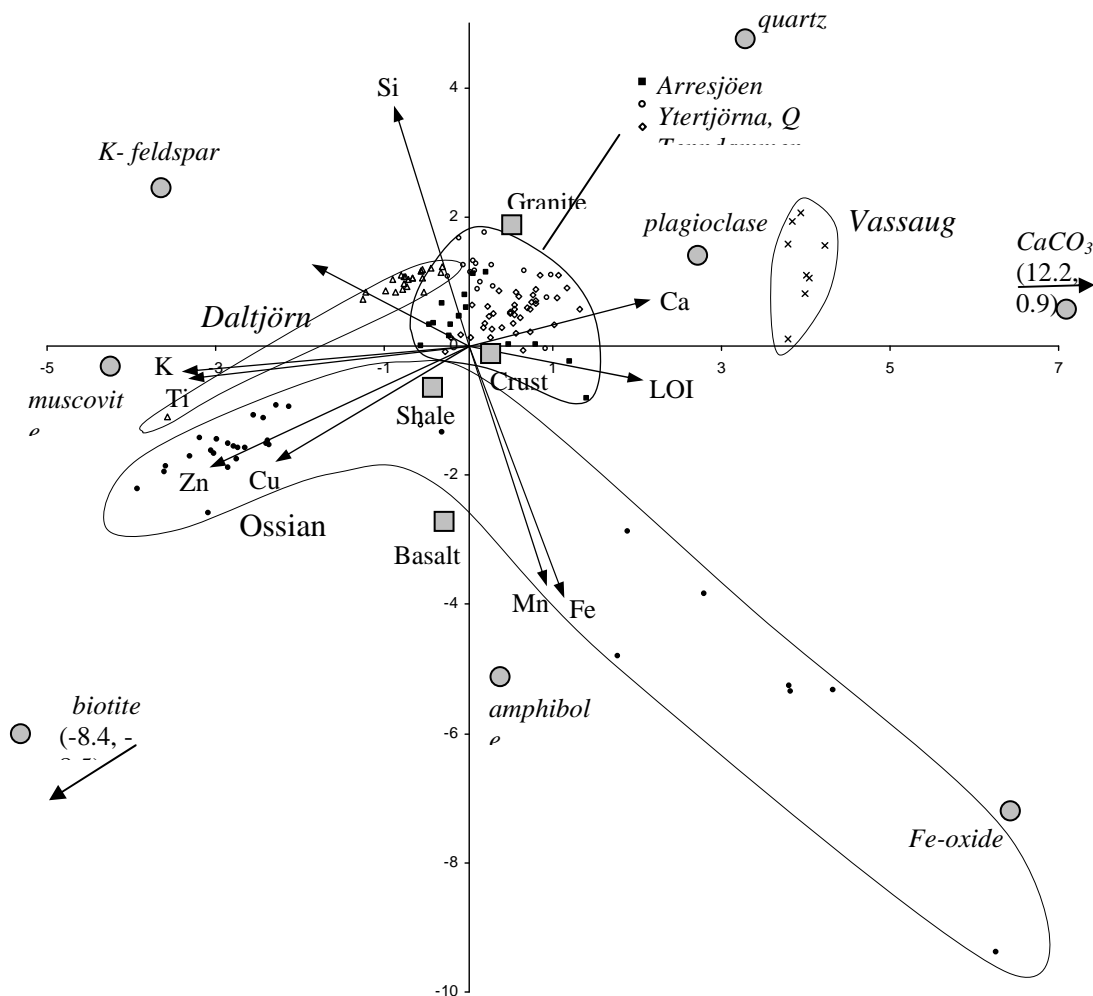


Figure 3. PCA biplot of the sediment composition data. Mineral composition data (Deer et al. 1966) and average rock types (Krauskopf 1982) were not included in the analysis and are passively projected onto the PCA axes.

## Results

Concentration-depth profiles (Figure 2) illustrate the major temporal trends in elemental variation. Between-site differences and compositional comparison with common rock types and minerals are illustrated using a PCA biplot (Figure 3). The accumulation-rate profiles for Ti and organic matter are shown in Figure 4. The results of the tests for atmospheric heavy metal contamination are shown in Figure 5. The most important patterns at each site are outlined in the following sections.

### *Ossian Sarsfjellet (C)*

An abrupt change in the elemental composition of the sediment in Ossian Sarsfjellet, not apparent from the basic lithological description, is seen at 14 cm (Figure 2). Both above and below this depth the sediment is relatively Fe-rich, and the heavy metal/K ratios are approximately the same. However, beyond this there is little similarity in the nature of the upper and lower sediment.

In the upper sediment there is a strong negative correlation between organic matter and Ti ( $r = -0.86$ ), suggesting separate controls for the mineral and organic matter supply. The PCA biplot (Figure 3) shows two mixing trends depending upon the Fe concentration, both trends meeting at an end-member similar to average shale. In the upper sediment, this shale component mixes with an end member similar in composition to biotite (K, Fe, and Ti enriched). In the lower Fe-enriched sediment, the shale component mixes with an oxide component highly enriched in Fe and Mn.

Sediment in the interval 3.5 to 7.0 cm experienced extremely rapid accumulation, but differs from the more slowly accumulated sediment mainly in its distinctly lower  $^{210}\text{Pb}$  activity. Redistribution of older marginal lake sediment, via slumping or storm activity, for example, provides a plausible explanation.

The heavy metals have very similar concentration profiles, which are also similar to those of Fe, Mn, and K in the upper 14 cm of sediment (Figure 2). However, there is also some similarity with the organic matter concentration profile, such as the minima at 3 cm. These associations are confirmed by RDA, which shows a significant relationship of Pb with K, organic matter, Si, and Ca, accounting for between 55 and 67% of the Pb variance. The variance explained by the model does not increase as the upper samples are excluded (Figure 5), suggesting that there is no significant atmospheric Pb signal. RDA shows that much of the Cu and Zn variance can be explained by K (approximately 40% of variance explained for both metals). However, organic matter also influences Cu (20% of variance), while Ca influences Zn (10% of variance).

### *Ytertjørna (Q)*

There is a strong negative correlation between organic matter and Ti ( $r = -0.82$ ), pointing to separate supply controls for mineral and organic matter. The Fe concentration pattern varies with scale. Fine-scale variation is antithetic to K and Ti, but over the whole core they correlate positively, supporting a role for a mineral such as biotite. The local anticorrelation of Fe with K and Ti suggests that an oxide component is present that is independent of the clastic mineral matter. Locally, there is a positive correlation of organic matter with Fe, suggesting an association of organic matter with the oxide component. However, the long-term trend for organic matter is for an up-core increase, contrasting with the decrease in clastic Fe and Ti.



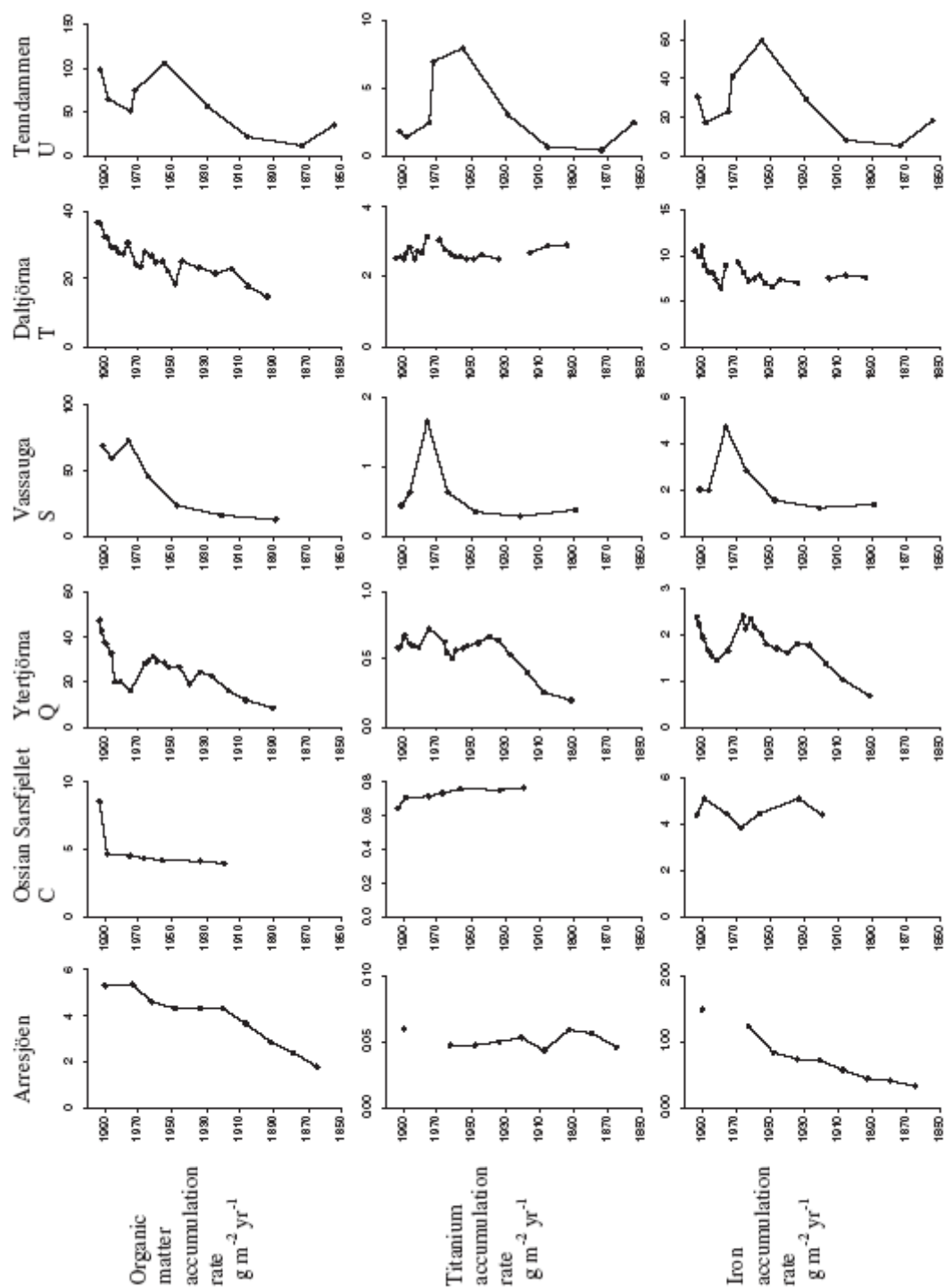


Figure 4. Calculated sediment fluxes profiles for Ti, Fe, and organic matter. Organic matter shows an increasing trend at all sites. Ti and Fe are more variable, but 4 of the 6 cores shows flux maxima around 1970.

The temporal variation in mass accumulation rate is weakly related to the elemental composition of the sediment (Figure 2), with a slight tendency for rapid sediment accumulation to be associated with an elevated organic matter concentration. This contrasts with results from the other sites.

The heavy metals in Ytertjørna show a complex relationship with the sediment. In general the trends in the concentration profiles resemble those of Mn, Ca, and K, but they share troughs and peaks with organic matter and Fe. RDA confirms these associations. 40% of the Pb variance is explained by organic matter, with another 20-30% by Ca and Ti. Except for the uppermost samples, where there is enrichment in both Ca and Mn, the proportion of variance explained varies little as the upper samples are excluded (Figure 5), suggesting that any atmospheric Pb signal is insignificant. RDA shows an influence of both K and organic matter on both Cu and Zn (30% and 15%, respectively, of variance explained for Cu, and 15% and 6% for Zn). Zn is, however, far more strongly influenced by Ca (37%).

#### *Vassauga (S)*

This is superficially different from the other sites in that carbonate enrichment is seen and other metal concentrations are generally low. As a consequence Ti and organic matter are positively correlated due to common dilution. The sediment divides into distinct upper (black) and lower (grey) units. The lower sediment unit comprises approximately 60% CaCO<sub>3</sub> (calculated from the Ca concentration), with the rest of the material dominated by aluminosilicates and organic matter. At 9 cm, the carbonate concentration falls sharply, as does the organic matter; Si, on the other hand, increases strongly.

In the upper 8 cm of sediment, Ti and other aluminosilicate elements increase in concentration up-core, while Si decreases. The independently varying silica is explained by quartz sand, which is abundant in the strandflat catchment.

The importance of the beach-sand flux in controlling the composition of the uppermost sediment is demonstrated by the <sup>210</sup>Pb data. Such certainty is not possible for the cause of the change at 9 cm, due to the absence of <sup>210</sup>Pb data below 6 cm. However, the similarity of the Ca and organic matter concentration profiles through the period of rapid change is more easily explained by common dilution, than by simultaneous supply failure of otherwise independently varying Ca and organic matter. Thus, a step-up in the supply rate of beach sand is the most likely explanation for the sharp fall in Ca concentration at 9 cm. There is no evidence from which to determine why this supply increase occurred.

The high and variable dilution by Si leads to a high correlation between most other elements. However, it is clear that the heavy metals and Fe are best correlated with organic matter (most markedly for Zn ( $r = 0.99$ ) and Pb ( $r = 0.97$ )). All the metals show a greater similarity to the organic matter curve than the Ti curve, and indeed heavy metal/K and heavy metal/Ti ratios increase strongly up-core. Under these circumstances detection of anthropogenic contamination is not possible.

#### *Daltjørna (T)*

Daltjørna shows the least down-core variation of all the cores. Consequently, inter-element correlations are weak. Organic matter and Ca show the greatest variations. The patterns shown in Figure 3 can largely be explained by mixing a K-rich mineral such as muscovite, with a Ca mineral such as plagioclase. The near-surface enrichment in Fe and Mn, over the top 1 cm, shows that an oxide component is also present.

Daltjørna has the highest catchment:lake area ratio, which probably explains its high sediment accumulation rate (Figure 2). The accumulation-rate profile shows no long-term trend, but has two peaks (at 2 - 3 and 5 - 6 cm). There is some sign of decreased organic

matter concentration associated with both of these peaks, compatible with mineral enrichment of the rapidly supplied sediment. The absence of Ti enhancement at the accumulation rate peaks can be explained by the high mineral matter content of the sediment (80+ % mineral matter). A small increase in mineral supply rate will dilute the organic matter proportionately, but will have little impact on the mineral element concentrations. Accumulation-rate impacts on Fe and heavy metals are ambiguous. The first peak (5 - 6 cm) may coincide with the depressed Pb and Zn at the base of the core. There is, however, no such effect for the shallower peak. This suggests that mineral supply and heavy metal supply rates are linked.

Strong enrichment of the surface sample is seen for Cu, Pb, and Zn. If this is disregarded, Daltjørna shows up-core increases in the concentrations of Cu and Pb only. Such an increase is also seen for organic matter. RDA confirms the role of organic matter; 40 % of the Pb variance is explained by organic matter, with another 20-30% by Ca and K. Except for the uppermost samples, the proportion of variance explained varies little as the upper samples are excluded from the analysis (Figure 5), suggesting that any atmospheric Pb signal is insignificant. RDA shows that 20% of the Cu variance is explained by Ti. 53% of the Zn variance is explained by organic matter, Ca, and K (26%, 17%, and 10%, respectively).

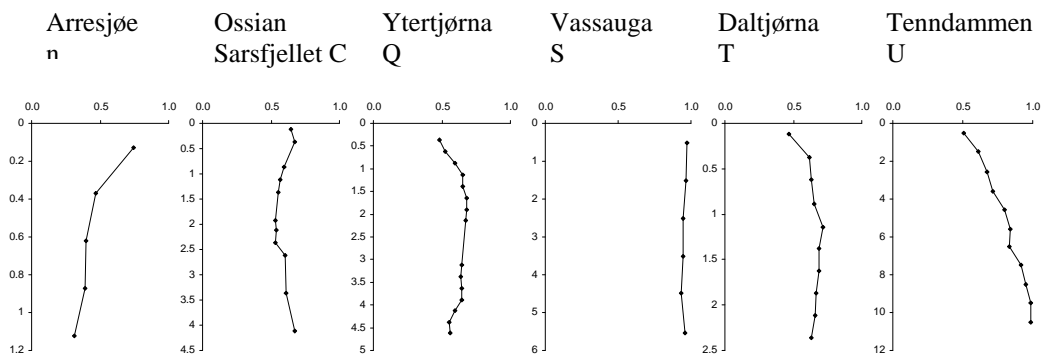


Figure 5. Change of  $R^2$  values as upper samples are progressively excluded from the redundancy analysis model. If a significant anthropogenic signal is present the values should fall up-core.

### Tenndammen (U)

There is a strong negative correlation between organic matter and Ti ( $r = -0.81$ ) showing that an independently varying organic matter component is present. The elemental composition of the sediment is strongly related to mass accumulation rate (Figure 2). Rapid sediment accumulation is associated with low concentrations of organic matter and Fe, and high concentrations of Si, Ti, and K. The concentrations of Ca, Cu, Mn, Pb, and Zn are unaffected by the accumulation rate peak suggesting that the heavy metals were supplied with the sediment.

Sediment below 8 cm shows a positive correlation of Cu, Pb, and Zn with Ti, K, and Fe. A role for mineral matter in governing the heavy metals is clearly indicated. Above this depth the situation is very different. Ti and K fall up-core, while heavy metals and Fe continue to rise. RDA shows that the Pb concentration in the lower part of the core is explained dominantly by Ti (68% of variance) but also by organic matter (26% of variance). Above 7 cm the role for Ti weakens (Figure 5). In the upper sediment, the Pb and Zn concentration profiles are similar to that for Fe. If the Pb profile is corrected for the mineral contribution based on the Pb:Ti relationship of the basal sediment (Figure 6), it closely resembles the SCP record (Figure 2), which may point to atmospheric contamination. However, if both Ti and organic matter are used to predict the Pb for the upper 7 cm, then the pattern is far less clear,

and the upper two samples actually have less Pb than expected. RDA shows that 78% of the Cu variance and 45% of the Zn variance are explained by Ti.

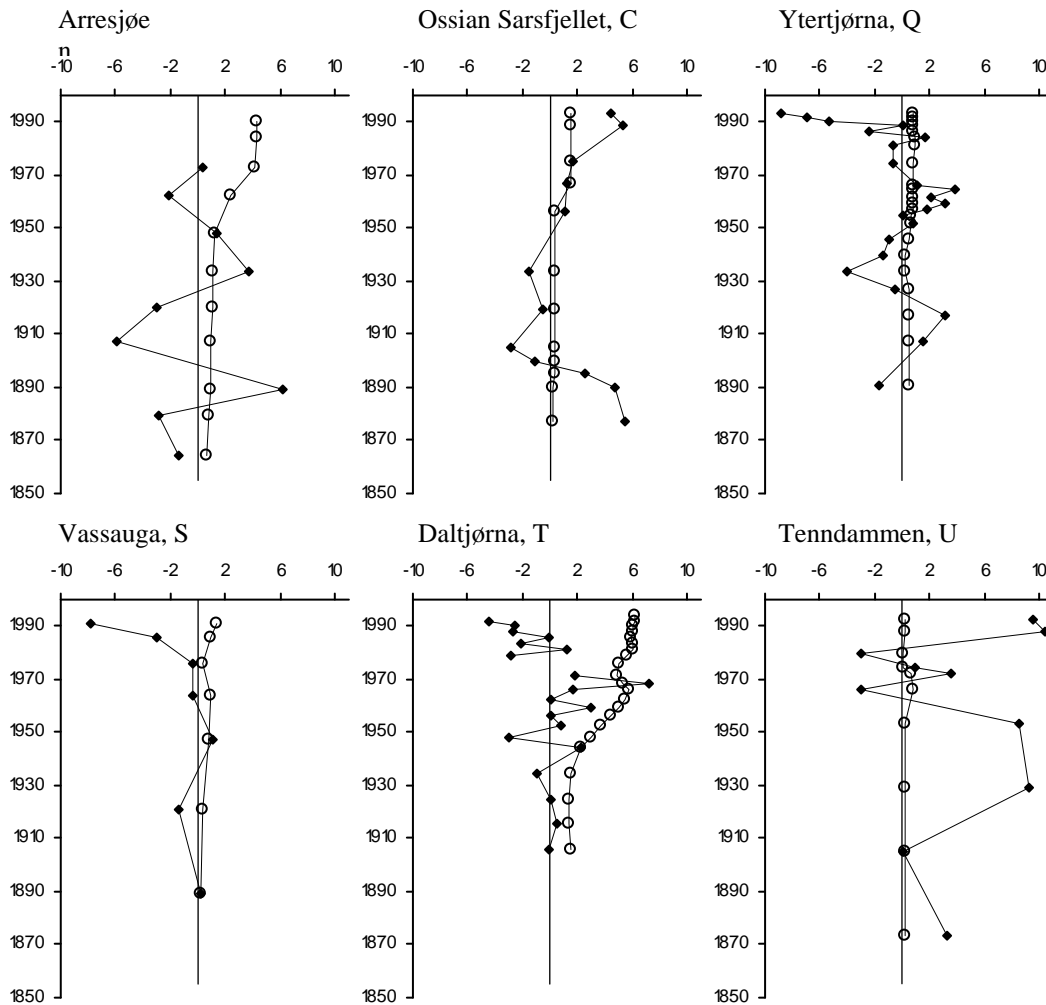


Figure 6. Corrected heavy metal concentration profiles (closed circles, by the method of Hilton et al. (1985)) and expected curves calculated from Greenland ice-core data (open circles, using the lake model of Boyle and Birks (1999)).

### Arresjøen (Arsj)

Down-core variation in major element concentrations reveals three main groupings (Figure 2). First, organic matter varies independently, increasing steadily up-core. Second, lithophile elements decrease up-core, showing a strong negative correlation with organic matter ( $r$  for Ti and organic matter = -0.82), suggesting separate controls over mineral and organic matter supply. Third, Fe and Mn show enrichment in the top 1 cm of the sediment, suggesting that an oxide component is present.

The similarity between the sediment accumulation rate and organic matter concentration profiles (Figure 2) suggests a trend of increasing organic matter supply. In contrast, mineral matter supply rates are approximately constant.

The heavy metals all show enrichment in the uppermost sample. If this is disregarded, a slight up-core concentration increase remains for Pb. Organic matter also increases up-core, and an association of Pb with organic matter is confirmed by the RDA; about 40% of the Pb variance can be explained by organic matter variation. RDA also shows that 20% of the Cu variance and 6% of the Zn variance can be explained by organic matter. Cu is also partially

explained by Ca variation (14% of variance), while Zn is partially explained by both Ca and Ti (8% and 16%, respectively). The amount of Pb variance explained does not increase with the exclusion of the upper sediment sample (Figure 5) suggesting that any anthropogenic contamination in the signal is insignificant.

## **Discussion**

### *Between-site variation in major sediment components*

The major element concentration profiles show very great differences between sites (Figure 2). Ossian Sarsfjellet (C) and Vassauga (S) both show an abrupt change in composition at depth, though with different causes. Ytertjørna (Q) shows cyclical variations in composition, while Daltjørna (T) shows very little stratigraphic variation. There are, however, some features in common. All of the sites except Vassauga (S) have extremely low Ca concentrations, and thus very little Ca-rich material. The Ca concentrations, far lower than those of even acid igneous rocks, show a high degree of compositional maturity. As extreme weathering of the catchment soils is unlikely, compositionally mature source rocks are implied. This is reasonable for Daltjørna (T), and more so for Tenndammen (U), with catchment rock types of fluvial origin. However, it seems rather unlikely for Ossian Sarsfjellet (C), Ytertjørna (Q), and Arresjøen (Arsj), where the bedrock is dominated by crystalline basement (Table 2). An alternative explanation is that mechanical sorting has left behind the coarse-grained Ca-rich minerals such as plagioclase and amphibole, separating them from Ca-poor mineral such as micas. Vassauga (S) is an exception to this, being rich in calcium carbonate. There is no clear source for this in the bedrock. However, this site is located on the coastal strand-flat, with documented beach deposits of quartz sand. It is not improbable that shell sands are also present providing a source of calcium carbonate.

### *Temporal changes in major sediment components*

At Ossian Sarsfjellet (C) the early oxide-rich sediment lies well below the limit of the dating (extrapolation of the dates points to the middle of the sixteenth century (Appleby 2004)). The high elevation catchment has minimal vegetation and has a high proportion of glacial ice (Birks et al. 2004). The absence of any plausible source for an enhanced supply of dissolved Fe and Mn in the past suggests that the boundary represents a dramatic increase in the supply of clastic mineral matter. There is, however, no sign of such a change in sediment supply rate in the biological record, although chironomid concentrations do decline at 14 cm (Brooks and Birks 2004).

Ossian Sarsfjellet (C) shows another feature not seen in the other lakes. At some time during the twentieth century a period of extremely rapid sediment supply occurred. The sediment deposited during this event is very similar to the rest of the core, though it is slightly Ti-rich and organic matter-poor. Whether this event is due to rapid catchment erosion, or to redistribution of older marginal lake sediment cannot be determined, as similar sediment would result from both processes. Diatom evidence, which might have answered this question, is lacking due to poor preservation at this site (Jones and Birks 2004).

The other four lakes show no such dramatic changes. However, similar but more subtle changes over the 20th century are observed in all cores. This can be seen clearly in the accumulation rates of organic matter and Ti (Figure 4). The Ti accumulation rate profiles all lack significant long-term trends, but show a number of sharp peaks. It is striking that four of the six cores (not Ossian Sarsfjellet (C) or Arresjøen (Arsj)) show a Ti supply peak dating to

approximately 1970. A regional event is implied, leading to enhanced supply of coarser clastic sediment either from the catchment or from littoral sediments. In either case, the widespread occurrence of the phenomenon requires a regional cause such as a change in climate.

The organic-matter accumulation-rate record differs from the clastic sediment record, though several sites also show the 1970 spike. The organic-matter accumulation rate is characterised by an underlying up-core increase. This can be explained by:

- Eutrophication, leading to enhanced within-lake production of organic matter.
- Steady state progressive diagenetic consumption of a constant natural supply of organic matter. This should lead to an exponential decay with depth. The sediment surfaces were observed to have distinct algal growth, and it is likely that the relatively high organic matter concentrations at the sediment surface are a consequence of this.
- Enhanced top-soil erosion, bringing in oxidation-resistant soil organic matter.

No analysis of the organic matter has been undertaken, yet its origin can be evaluated through correlations with other elements in the sediment record. This kind of evidence suggests that the first and second explanations are unlikely. Autochthonous organic matter should cause dilution of heavy metals, assuming that the metal capture efficiency for the lakes is high. The whole-lake metal capture efficiency can be estimated using the simple model described in Boyle and Birks (1999). This shows that Arresjøen has the lowest capture efficiency because of its exceptionally low sediment accumulation rate. However, even at Arresjøen, the capture efficiency for Pb is between 80% (based on the distribution coefficient estimates of Boyle and Birks (1999) and 90% (based on values found by Appleby (1997)). For the other lakes at least 90% capture efficiency for Pb is found with both estimates for the distribution coefficient. If the organic matter was autochthonous it would have to result in a dilution of the heavy metal concentration. At Ossian Sarsfjellet (C) and Arresjøen the effect would be small, but at the other sites the increases in organic-matter accumulation rate should result in at least a halving of the Pb concentration in the near-surface sediment. The absence of this effect confirms that the enhanced organic-matter supply is linked to an enhanced supply of Pb.

The major trends in organic matter are therefore best explained in terms of an allochthonous supply of soil organic matter. However, there is an additional effect in the uppermost samples at some sites. The uppermost samples at Ytertjørna (Q), Vassauga (S), and Daltjørna (T) all show an increase in organic matter concentration which, while not diluting the Pb, does not correlate positively with it. This suggests that at three of the sites the organic matter in the surface sediment is different from that which underlies it. An algal origin is plausible, though not verified by other means.

#### *Iron and manganese supply: particle transport, authigenesis, and diagenesis*

Near-surface Mn and Fe enrichment of lake sediments is common, due to post-burial remobilisation and upward diffusion (Engstrom and Wright 1984; Davison 1993). All of the lakes studied show clear surface-enrichment in Mn, but there appears to be more than one explanation for this. At Vassauga (S) the near-surface Mn enrichment is no greater than for organic matter, both of which can be entirely attributed to an up-core decrease in dilution by quartz sand. At Ossian Sarsfjellet (C) the near-surface Mn increase is similar to that of organic matter, Cu, Pb, and Zn. As these materials have widely varying solution phase mobility, a

diffusive explanation is inadequate. Instead, a supply factor linking all of the substances is more plausible. At Arresjøen (Arsj), Daltjørna (T), and the surface sediment at Ytertjørna (Q), the sediment Mn profiles are all distinctively different from the other elements, and have the classic exponential shape found where diagenetic redistribution of Mn is occurring. At depth in Ytertjørna the Mn profile is smooth across peaks in both accumulation rate and Ti. Such a pattern is compatible with diagenetic remobilisation. At Tenndammen (U) the sharp peak in sediment accumulation rate has no impact on the Mn profile, which may indicate post-burial diffusive enrichment. In addition, a strong subsurface Mn peak is present at this site. At Lake Baikal, such peaks are attributed largely to the preservation of a surface enrichment zone by rapid burial (Granina et al. 1993).

At Vassauga (S), Daltjørna (T), Tenndammen (U), and Arresjøen (Arsj) there is also Fe surface-enrichment. At Vassauga this is best explained by the up-core decrease in dilution by quartz sand. However, at Daltjørna, Vassauga, and Arresjøen surface-sediment Fe enrichment is most simply explained by diagenesis. In contrast, at Tenndammen the strong accumulation rate peak depresses the concentration of Fe, indicating that any diffusive fluxes are far lower than for Mn. At Ossian Sarsfjellet (C) and Ytertjørna (Q) the well-defined pattern of variation in Fe concentration is not compatible with significant remobilisation.

The coincidence of peaks in both organic matter and Fe concentrations is striking, and almost certainly arises from more than one cause. At Ossian Sarsfjellet (C) and Daltjørna (T) the near-surface increases in both organic matter and Fe can plausibly be attributed to the accumulation of organic-rich diagenetic/authigenic Fe phases (Tipping 1981; Fortin et al. 1993). A large part of the organic matter surface-enrichment at Tenndammen (U) and Arresjøen (Arsj) can also be explained this way. However, at Ytertjørna (Q) the organic matter concentrations are far too great for this explanation. If the highly correlated organic matter and Fe are to be explained by supply factors, then an external source is required. In these high pH (see Table 1), clear-water lakes a dissolved supply of Fe is unlikely to be significant. A plausible alternative is that both Fe and organic matter are both derived from soil organic matter. Such material is present in the thin catchment soils, though no chemical analysis of it was undertaken.

#### *Heavy metals - Natural impacts: diagenesis, sources*

Two aspects of the heavy metal profiles are immediately apparent (Figure 2). First, the concentrations vary greatly with depth, with little clear sign of trends. Second, at most of the sites there is a marked enrichment of heavy metals in certain samples, particularly close to the surface of the sediment column. From this it follows that very careful treatment of the natural controls over heavy metal concentration variations is needed if any sense is to be made of recent changes.

#### *Spatial trends in heavy metals*

Basic rocks are richer in Cu and Zn than acidic ones, while granites are richer in Pb than other igneous rock types. Pelites tend to be enriched in all trace elements, due to the high adsorption properties of the parent clays. However, these statements are only true on average, and great variation exists. Differences in the heavy metal concentration of rock types can be explained by specific associations of trace metals with some rock-forming minerals. In such minerals Pb correlates highly with K, and is found enriched in both micas and K-feldspars. Zn and Cu are more variable, but are enriched in pyroxenes and amphiboles (hence an association with basic

rock types) and with biotite (particularly Zn). The correlations of Pb, Cu, and Zn with K, Ti, and Ca shown by the RDA can all be partially explained by variations in these minerals.

Table 3 shows that there are between-site differences in the average heavy metal concentrations. All the heavy metals are relatively enriched at Ossian Sarsfjellet (C) and Arresjøen (Arsj), both of which are sited on the same geological unit (Table 2). Pb is also relatively enriched in Daltjørna (T), which is also enriched in K (Figure 2). The high heavy metal concentrations at Arresjøen are partly explained by the different analytical procedure. They were determined by total digestion, which typically dissolves significantly more Zn and Cu. The higher Cu and Zn at Ossian Sarsfjellet can plausibly be explained by geology, but this appears to be the exception not the rule. This is not to say that geological control is unimportant. RDA clearly shows the importance of mineral elements in explaining the stratigraphic patterns. However, the role of minerals in explaining the between-site differences in heavy metal concentrations is weakened by the similarity of the sites.

Table 3. Heavy metal concentrations. Standard deviations are in parentheses.

µg/g	Ossian S.	Ytertjørna	Vassauga	Daltjørna	Tenndammen pre-1910	Arresjøen
Cu	45.6 (6.0)	26.9 (4.7)	29.7	22.4 (4.9)	23.5 (2.0)	38.2 (10.5)
Pb	30.4 (10.1)	24.4 (3.9)	20.9	28.6 (3.1)	21.4 (2.8)	43.2 (8.3)
Zn	115.8 (32)	62.3 (7.1)	56.8	60.8 (6.2)	70.2 (6.6)	77.9 (6.2)

#### *Temporal trends in heavy metals*

RDA, which shows the importance of minerals in influencing heavy metal concentrations, also points to a key role for organic matter. This organic-matter component commonly covaries with Fe, most strikingly at Ytertjørna (Q), Vassauga (S), Daltjørna (T), and Arresjøen (Arsj). A possible explanation for this is common dilution. If the total-sediment supply rate is governed by the mineral-matter supply rate, as argued above, then the concentrations of all other components will rise and fall in response to variations in the accumulation rate. They will therefore correlate, even if they have separate and unrelated sources. However, this argument cannot be used to explain the up-core increase in organic matter, because there is no evidence for an up-core decrease in mineral supply rates that would be needed to drive it (Figure 4). Instead, an increase in supply of both Pb and organic matter is required. There are two possible sources: catchment, or underlying sediment via post-burial remobilisation. Two arguments can be used to reject post-burial remobilisation. First, the sediment accumulation rates are rather high to permit a significant diagenetic component (Boyle 2001b). Second, Mn, Fe, Pb, Zn, and Cu show little fractionation. Only supply in particulate form can explain this.

The arguments presented above suggest that there are two major natural supply pathways for metals into the lakes on Svalbard. First, there are very-fine particles or colloids that are rich in Fe, Mn, organic matter, and heavy metals. While we have no direct evidence, soil organic matter is the likely source for this material. Second, there is clastic mineral matter, probably dominated by micas (mainly biotite) and amphiboles. Variations in the proportions of these components are influenced by sediment supply rate, suggesting that erosion intensity is a factor in their separation. Further fractionation of these components may take place in the lake, as the finer, metal-enriched size fraction can more easily be transported to the centre of the lake by focusing mechanisms. These two allochthonous controls over the sediment heavy metal concentrations are modified by strong independent metal enrichment of the surface sediment at some sites. This is probably a stationary peak associated with



biogeochemical cycling (e.g. Boyle 2001a), and is particularly striking at Ytertjørna (Q) and Daltjørna (T).

#### *Evidence for anthropogenic heavy metal contamination*

The Hilton et al. (1985) procedure has been used to test for an atmospheric heavy metal contribution. This is done by identifying a depth at which heavy metal concentrations vary independently. This approach reveals evidence for an atmospheric contamination signal only at Tenndammen (U), and even there the evidence is open to interpretation. It is clear that the onset of SCP contamination at Tenndammen (U) coincides with a change in the character of the sediment record (Figure 2). This suggests that human disturbance of the catchment coincided in time with the construction of the power stations at Longyearbyen, Barentsburg, and Pyramiden (Rose et al. 2004). This fact, and the similarity between the SCP and Pb concentration profiles, is compatible with an anthropogenic origin for the Pb, either directly from the power stations or indirectly from other human activities. However, it remains a possibility that human disturbance altered the natural supply of Pb from the catchment, decoupling it from the supply of Ti and K.

The absence of a clear atmospheric pollution record in the lake sediment at the six study sites has to be interpreted in the light of the weakness of the pollution signal and the natural variability of the sediment record. The RDA shows that no more than 70% of the variance in the heavy metals is accounted for by the observed natural factors. This suggests that the presence of atmospheric contamination at the sites cannot be rejected, but that any signal is hidden by the natural variability. To test this idea the RDA results have been used to predict a natural baseline for the Pb. In Tenndammen (U), the Hilton method provides a cut-off depth for the baseline model. In the other sites no such depth was identified, and the RDA model for pre-1950 sediment is used instead. After correction of the concentration data for this baseline estimate, profiles of predicted anthropogenic metal fluxes can be derived (Figure 6).

To interpret the baseline-corrected Pb profiles it is necessary to estimate what an expected profile should look like. This can be done by assuming that the Greenland ice-core data (Boutron et al. 1995) give representative values for historic metal concentrations in precipitation over Svalbard. By applying these data to a lake-sediment model that allows for water fluxes, expected sediment concentration profiles can be calculated (using the method of Boyle and Birks 1999). The water fluxes can be estimated from catchment/lake area ratios and effective precipitation. The former can be derived from map data. Effective precipitation is assumed to be 250 mm yr<sup>-1</sup> at all sites. Values calculated from these data are expressed in areal form (per m<sup>2</sup> of lake area) in Table 4.

Sediment fluxes have been measured at each lake, but only at a single coring site. There are two obstacles to estimating lake-wide sediment fluxes from these data; discrete sediment redistribution events and more continuous focusing. High accumulation-rate events, such as that in Ossian Sarsfjellet (C), may not reflect lake-wide processes. In addition, even where discrete events are absent, there may be considerable focusing, leading to an enhanced sediment accumulation-rate at the coring site. Information about both kinds of sediment redistribution is present in the <sup>210</sup>Pb record. Both redistribution processes lead to higher than expected <sup>210</sup>Pb inventories. Normalisation to a local expected <sup>210</sup>Pb inventory provides a means of minimising these effects. As a simple correction for this data set the Pb supply fluxes predicted from the Greenland ice-core data were scaled by the factors shown in Table 4 to allow for focusing.

The comparisons between the baseline-corrected heavy metal profiles and the expected patterns based on the Greenland ice-core data are shown in Figure 6. These data suggest that we should not expect to see a long-transported anthropogenic signal in Svalbard except at very slowly accumulating lake sites. The study lakes are simply too insensitive.

Table 4. Annual water and sediment fluxes for the study lakes

Lake	Annual water flux $\text{m}^3 \text{m}^{-2} \text{y}^{-1}$	Current annual sediment flux $\text{g m}^2 \text{yr}^{-1}$	$^{210}\text{Pb}$ inventory $\text{Bq m}^{-2}$	Scaling factor applied to predicted Pb flux
Ossian S. (C)	3.5	88	1914	1
Ytertjørna (Q)	2.7	140	7904	5.4
Vassauga (S)	12	250	2582	1.8
Daltjørna (T)	23	460	1497	1
Tenndammen (U)	3.5	540	1455	1
Arresjøen (Arsj)	2.3	23	1560	1

## Conclusions

The six lakes studied in Svalbard show striking temporal and between-site variations in the elemental composition of their sediment. The between-lake differences in major element concentrations can be attributed largely to differences in catchment composition. Geology also partially explains between-lake differences in heavy metal concentrations.

Temporal variation in sediment elemental concentrations can be attributed to changes in the supply of catchment-derived minerals. Of particular significance is the variable supply of two contrasting sediment sources. First, a clastic sedimentary material, supplied by soil erosion or resuspension of littoral sediments, enriches the sediment in elements associated with rock-forming minerals. Second, a fine particulate or colloidal sedimentary component enriches the sediment in organic matter, iron, manganese, and heavy metals. This component is probably soil organic matter.

Temporal variation in the supply of these two sediment components includes some local events affecting single sites, but also includes two features that are present in most, if not all, of these lakes, suggesting a regional cause. First, all of the lakes show an up-core increase in the concentration of organic matter, which can only partially be accounted for by diagenetic effects. This requires a regionally enhanced allochthonous supply of organic matter. Second, the four central and southern sites all show an event at about 1970 characterised by an enhanced accumulation of Ti-rich clastic sediment. This requires either enhanced allochthonous supply, or enhanced mobilisation of littoral sediments. In either case a regional driving force, such as a period of increased precipitation is required. Some of the highest annual precipitation in the last 90 years on Svalbard was between 1972 and 1975 (Førland et al. 1997).

At most of the sites, the heavy metal concentration profiles can be explained entirely by natural factors, largely controlled by the supply of soil organic matter and clastic minerals such as biotite and amphibole. However, at Tenndammen (U), situated close to the Svalbard's largest settlement at Longyearbyen, there is a possibility of detectable anthropogenic Pb enrichment.

The predicted level of long-transported anthropogenic Pb supply, based on data from the Greenland ice, suggests that the sites are not sufficiently sensitive to show any pollution impact.

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