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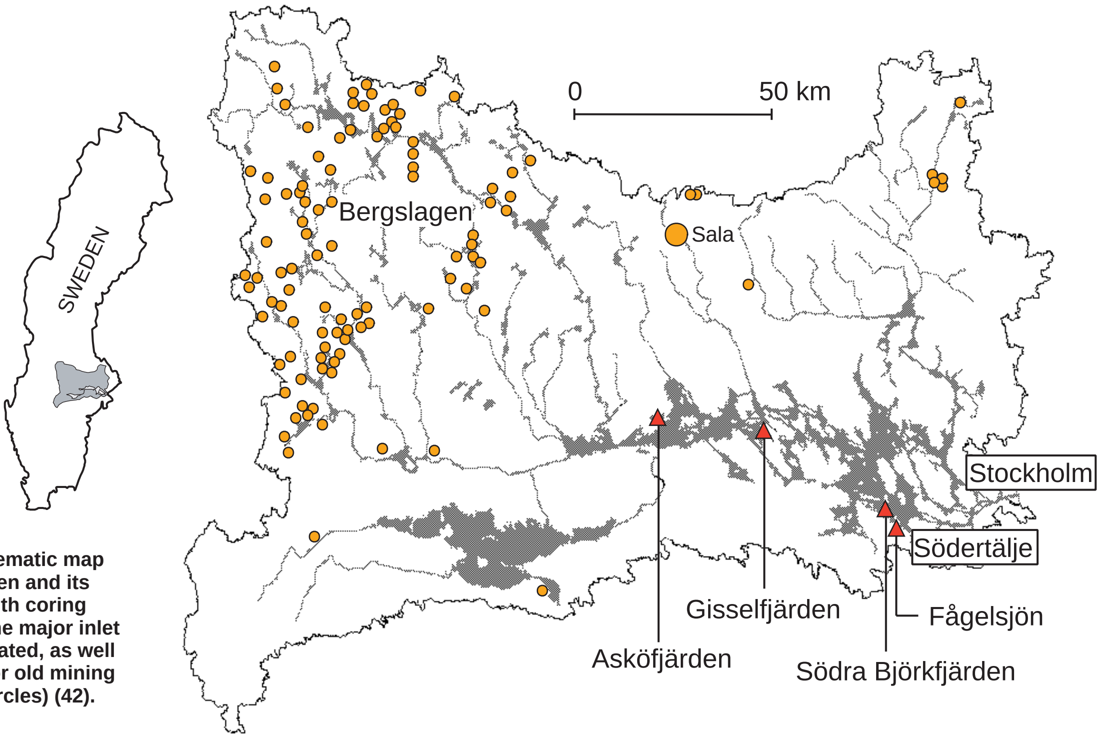


Figure 1. Schematic map of Lake Mälaren and its catchment, with coring sites and some major inlet streams indicated, as well as some major old mining sites (filled circles) (42).

Lake Mälaren is the water supply and recreation area for more than 1 million people in central Sweden and subject to considerable environmental concern. To establish background data for assessments of contemporary levels of trophy and heavy metal pollution, sediment cores from the lake were analyzed. Diatom–inferred lake–water phosphorus concentrations suggest that pre-20th century nutrient levels in Södra Björkfjärden, a basin in the eastern part of Mälaren, were higher (c. 10–20 μg TP L^{-1}) than previously assumed (c. 6 μg TP L^{-1}). Stable lead isotope and lead concentration analyses from 3 basins (S. Björkfjärden, Gisselfjärden and Asköfjärden) show that the lake was polluted in the 19th century and earlier from extensive metal production and processing in the catchment, particularly in the Bergslagen region. The lake has experienced a substantial improvement of the lead pollution situation in the 20th century following closure of the mining and metal industry. The lead pollution from the old mining industry was large compared to late-20th century pollution from car emissions, burning of fossil fuels and modern industries.

INTRODUCTION

Mälaren is the third largest lake in Sweden, and it is a water supply and recreation area for a large number of people living around the lake and in the city of Stockholm. Concerns for the water quality of the lake have led to the implementation of a variety of measures since the 1960s to reduce nutrient inputs, particularly through improvements in sewage treatment. These measures have contributed to a 60% decline in phosphorus inputs to the lake (1). The long–term goal for Mälaren's water quality is that nutrient concentrations shall not exceed background values by more than twofold, and for metals sediment concentrations shall not exceed six times the background value (2). This inherently introduces the questions: what was the natural state of the lake and how can we define background conditions?

The lake was isolated from the Baltic Sea by land upheaval in a gradual process between 900 and 1300 AD (3). At that time, the population of the area was in the order of 30 000 people (4) or higher (5) and human impact from agriculture was already extensive in the large catchment of the lake (6–8). Trading places and urban centers lay around the lake (9), of which the Viking town Birka on the island of Björkö near the mouth of the lake is most famous. Mining for iron, copper, silver, and many other metals developed and became very important in the following centuries. When these activities culminated in the 19th century, there were hundreds or even thousands of mines and numerous furnaces and forges, abandoned or still in use, particularly in the Bergslagen region to the northwest of the lake (10). In the 20th century, mine after mine closed and today, no mining activities remain in the catchment of Mälaren.

A paleolimnological investigation of sediment cores from the lake was initiated in order to study the impact of early land–use and pre–industrial mining and metallurgy on the water quality of Mälaren, with the goal to establish a background for assessments of the contemporary levels of trophy and heavy metal pollution. The main technique for analysis of past nutrient conditions was diatom analysis, and stable lead isotopes and lead concentrations were used as indicators for past loading of heavy metals.

MATERIAL AND METHODS

A 90-cm master core, used for studies of both past trophic conditions and for heavy metal pollution, was taken in September

The master core from Södra Björkfjärden was taken in summer, while the additional long cores from Gisselfjärden and Asköfjärden and from the small forest lake Fågelsjön were collected in winter using a freeze corer (left) and a Russian peat corer operated with rods (right). The freeze corer, shown here, just before lowering it into the lake sediment, allows the taking of undisturbed cores of unconsolidated surface sediments. The freeze corer is filled with dry ice and the sediment freezes *in situ* in the lake bottom. Photo: C. Savage.

1998 with a Mackereth corer in Södra Björkfjärden, a basin in the eastern part of Mälaren near the outlets at Stockholm and Södertälje. This core was complemented by a 65-cm long HON–Kajak gravity core from the same site. Lead and flyash analyses confirm that the 2 cores are very similar, and that the main study core is representative for this 44-m deep basin (11). For the metal pollution study, additional long cores (150–250 cm) were collected from Gisselfjärden (at 12-m water depth) and Asköfjärden (at 7-m water depth). A long core was also collected from a small forest lake, Fågelsjön, situated immediately south of Södra Björkfjärden, with the aim of assessing the atmospheric lead pollution history of the area (Fig. 1).

Preparation of diatom slides for microscopy and diatom counting followed standard methods and the diatom taxonomy of Krammer and Lange–Bertalot (12) and Wunsam et al. (13). Mean epilimnetic total phosphorus (TP) concentrations were estimated from the sedimentary diatom assemblages using a calibration dataset developed for Swedish lakes (14). The weighted–averaging (WA) model was applied to the diatom data using the programs CALIBRATE version 0.81 (15) and WACALIB version 3.4 (16). The program ANALOG 1.6 (H.J.B. Birks and J.M. Line, unpubl.) was used to evaluate the occurrence of analogs for fossil diatom assemblages in the modern dataset. Poor analogs were identified using a χ^2 distance dissimilarity coefficient, where the χ^2 distance for a fossil sample was greater than 50% of the χ^2 distance distributions within the modern dataset.

Pigment analyses, as well as carbon and nitrogen stable isotope analyses, were made to complement the diatom–based assessment of past trophic levels. Pigments were analyzed using high–performance liquid chromatography (HPLC) following extraction in a mixture of acetone, methanol, and water (80:15:5) (17). Total carbon and total nitrogen concentrations and carbon and nitrogen isotopes, calibrated and expressed in the conventional delta notation (δ^{13}C and δ^{15}N), were determined simultaneously using automated elemental analyzer isotope ratio mass spectrometry (IRMS) (18).

Lead stable isotopes and lead concentrations were analyzed using inductively coupled plasma mass spectrometry (ICP–MS) following a strong acid digestion of freeze–dried sediment samples in conc. HNO_3+$HClO_4$ (v/v 10:1) (19). Common lead has 4 stable isotopes (^{204}Pb, ^{206}Pb, ^{207}Pb and ^{208}Pb), the proportions of which vary in different regions of the earth for geologic reasons. Consequently, lead from different sources can have different isotope compositions. The ^{206}Pb/^{207}Pb ratio is used most often as a source signature in environmental studies. The bedrock in Sweden and thus also unpolluted soils and sediments have natural ^{206}Pb/^{207}Pb ratios higher than 1.3 (19), higher values that are typical of Precambrian shield regions (20). In contrast, lead in ores has a lower ratio: 1.17 in ores exploited in pre–industrial time in mainland Europe (19); 1.04 in Australian lead, which was extensively used as an anti–knock additive in petrol and which gave late 20th century aerosols in Sweden a ratio of about 1.14–1.16 (21); and about 1.03 in ores from the Bergslagen region (22). This difference in signatures makes lead isotope analysis a powerful complement to concentration analysis in tracing lead pollution in the environment and to disentangle sources (23).

By combining the lead isotope and concentration results we can roughly estimate the contributions of lead from different sources to the lead composition of the sediments of Mälaren and Fågelsjön, and how the proportions have changed over time (24). For this, we assume that there are 3 major sources of lead: *i*) Natural lead from the catchment, whose $^{206}Pb/^{207}Pb$ ratio and lead concentration are determined from lower, pre–pollution sections of the cores. *ii*) Long–range air–transported lead pollution that originated to a large extent from mainland Europe and the British Isles and had a ratio of about 1.17 prior to AD 1900. During the 20th century the isotope ratio of atmospheric pollution declined further to about 1.14 due to the addition of lead from petrol. *iii*) Local lead, produced from ores in the Mälaren region that had a $^{206}Pb/^{207}Pb$ ratio of about 1.03.

One level (87–88 cm depth) in the core from Södra Björkfjärden was radiocarbon dated. This was a bulk sample, as terrestrial macrofossils were not found, which gave a calibrated age of 420–660 AD (radiocarbon age 1505 ± 60 BP; Ua–14594). This date seems at least a few hundred years too old, since no brackish–water diatoms were present in the sediment. It is common that dates of bulk sediment samples overestimate age due to reservoir effects. In the sediments of Swedish lakes that only receive pollution inputs from the atmosphere some approximate sediment age dates can be inferred from the lead record. This indirect dating method is based on analyses of the sediment records from > 30 small, forested lakes in Sweden. Three characteristic features can be used as chronological markers: a lead concentration peak c. AD 0; a large concentration increase and $^{206}Pb/^{207}Pb$ ratio decline c. AD 1000; and a lead concentration maximum c. 1970 (25).

Spheroidal carbonaceous flyash particle (SCP) counting was used to infer dates in recent sediments. These particles, which are derived from fossil-fuel burning, appear in sediments from the mid-19th century, increase markedly in concentration from about 1950 and peak in the early 1970s. There is usually a good temporal correlation between lead and SCP records in Swedish lake sediments from the 20th century (26).

RESULTS AND DISCUSSION

Past Trophic Conditions

Results of analyses to assess past trophic conditions are summarized in Figure 2. The Figure includes key diatom species and pigments, C/N ratio, carbon and nitrogen isotopes, and the diatom–inferred total phosphorus concentration.

Lake sedimentary organic matter is derived from the settling of organic detritus that was produced in the lake or transported to it from the surrounding catchment. A stable atomic C/N ratio of about 9 in the sediment core and high $\delta^{15}N$ values (≥ 5‰) are both characteristic of in–lake organic matter (27), and indicate that the predominant source of sedimentary organic matter to the Södra Björkfjärden site has always been from in–lake production. While carbon and nitrogen concentrations are very stable from 20 down to 90 cm (1.6% and 0.2%, respectively), their concentrations increase twofold from 20 cm to the surface (3.3% C and 0.46% N) indicating an increased supply of organic matter to the sediment.

There are variations in the carbon isotope composition ($\delta^{13}C$) that represent significant changes in the Södra Björkfjärden core, such as a small (–0.5‰) decline at 60-cm depth. For recent sediments we have not corrected for the Seuss effect, i.e. the –1.4‰ depletion of $\delta^{13}C$ in atmospheric CO_2 that has occurred since 1840 (28), since this requires a time dependent correction. Consequently, the –1‰ depletion of $\delta^{13}C$ observed towards the sediment surface in the Södra Björkfjärden core, if corrected for this historic depletion, would in fact represent a +0.5‰ increase, which is comparable to observed increases in recent sediments from the nutrient-enriched Baltic (29). Since a change in the relative contribution of aquatic and terrestrial organic matter can be excluded based on a stable C/N ratio in the whole core, the variations in $\delta^{13}C$ likely result from changes in the aquatic community, increased productivity, and/or changes in nutrient sources that lead to changes in isotope fractionation dynamics. However, specific interpretations of the complex mechanisms behind the carbon isotope changes cannot be established without more data.

For nitrogen, the isotope composition ($\delta^{15}N$) increases gradually by +1‰ upcore from 90-cm to 25-cm depth, after which a more rapid increase of +3‰ occurs up to 5 cm, where $\delta^{15}N$ reaches a maximum value of 9‰. The gradual increase in $\delta^{15}N$ may reflect a gradual increase in lake productivity, since the stability of the C/N ratio and carbon and nitrogen concentrations in this section of the core argue against diagenetic effects on organic matter composition (27). The more substantial near–surface increase in $\delta^{15}N$ is clearly indicative of a nutrient increase, and is comparable to the increases measured in recent sediments throughout the Baltic Sea basin, particularly in eutrophicated coastal waters receiving riverine inputs (29). These recent increases in $\delta^{15}N$ result from inputs of isotopically–heavier human–derived nitrogen (agricultural fertilizer and wastewater) to coastal waters and the resultant increases in aquatic productivity (29, 30).

When conditions are favorable, algal pigments (chlorophylls and carotenoids) preserve well in the sediment record. The taxon–specificity of some pigments means that they are useful biomarkers for characterizing past algal community structure, while changes in sedimentary pigment concentration can give a measure of past lake productivity (31). Unfortunately, the pigment record from Södra Björkfjärden is poorly preserved in the lower part of the core (below 40 cm), as indicated by the absence of all pigments except for pheophytin *a*, which is a degradation product of chlorophyll *a*. Pigment flux studies in lakes have shown that most pigment degradation occurs prior to burial in the sediment during sinking through the water column (32). Therefore, pigment degradation is more intense in deep lake basins such as Södra Björkfjärden. The pigment record suggests that the algal community has comprised cryptophytes (alloxanthin) and chrysophytes/diatoms (fucoxanthin). The cryptophytes increased from the early 19th century (35 cm), whereas the chrysophytes/diatoms have increased only in more recent time (7 cm). The annual lake monitoring program since 1966 supports the conclusion that cryptophytes and diatoms are the most abundant algal groups in Södra Björkfjärden (1). The absence of other diagnostic markers, e.g. chlorophyte and cyanophyte pigments, is most likely caused by the absence or very low production of these algal groups and not degradation, since their pigments are less labile than fucoxanthin (31).

An increase in sedimentary pigment concentration signifies increased algal production. However, when a lake becomes more productive, a number of positive feedbacks probably can also amplify the sedimentary pigment signal by discouraging pigment degradation. These feedbacks include increased turbidity of the water column that impedes light penetration, which in turn reduces the bleaching of pigments, as well as increased duration and intensity of hypolimnetic anoxia, which decreases pigment oxidation (31). Despite poor and possibly altered preservation conditions over time, there are some clear signals in the pigment profiles. The increase in alloxanthin (cryptophytes) and pheophytin *a* (all algae) concentrations in the early 1800s and c. 1950 suggests increased lake productivity; the latter is concurrent with changes in isotope–inferred as well as diatom–inferred increases in lake productivity.

The assemblage of sub-fossil diatom species preserved in sediments provides useful information about the ambient aquatic conditions at the time when the diatoms were growing, as there are clear associations between species assemblage and water-chemistry variables. For example, nutrient concentrations can

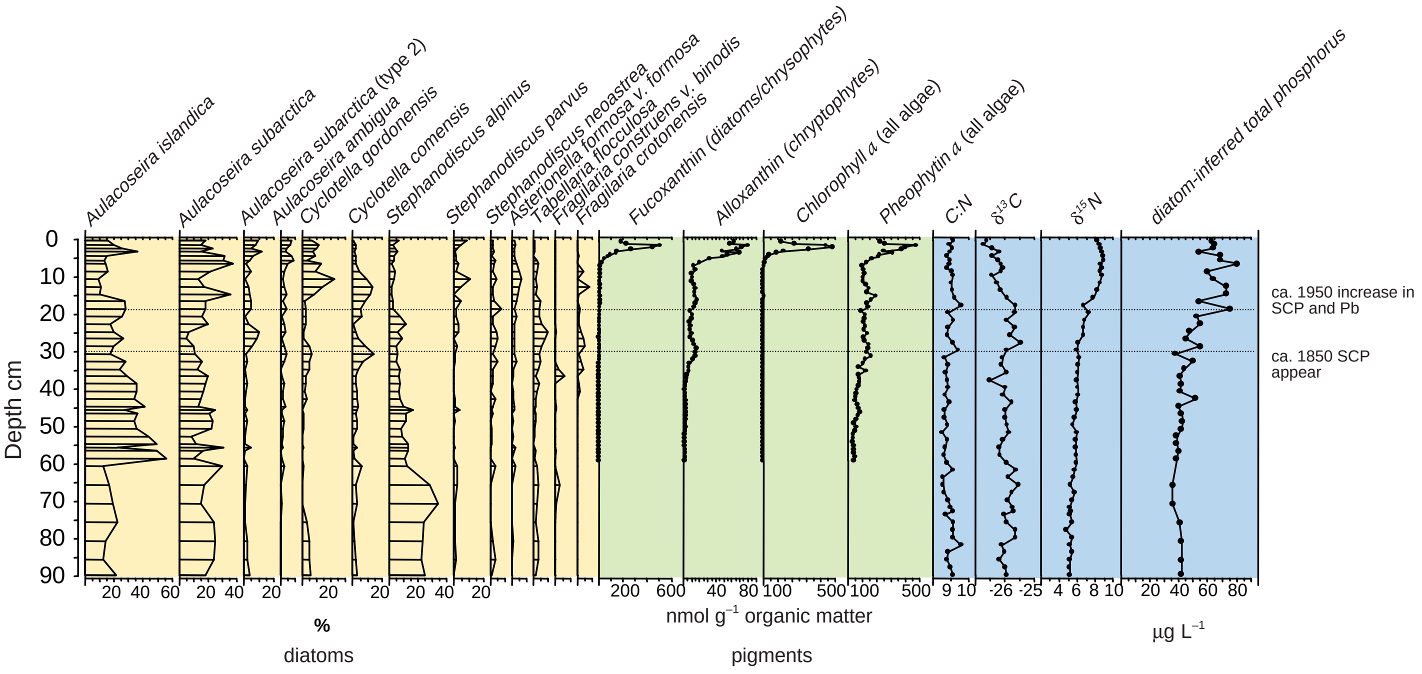


Figure 2. Selected diatoms and algal pigments, carbon and nitrogen, and diatom–inferred total phosphorus in a sediment core from Södra Björkfjärden. Dating of the sediment core is based on the flyash stratigraphy (SCP).

influence which species are best able to grow (33) in addition to determining the total abundance of diatoms. Since diatoms tend to preserve well in lake sediments they offer the possibility of inferring past lake–water conditions, such as is done in paleolimnological studies that infer variables such as pH, salinity, and TP (34).

The diatom flora in the core from Södra Björkfjärden is composed mainly of planktonic species. There are 2 main periods of change in the diatom record. The first occurs between c. 65-cm and 55-cm sediment depth, with a decline in the relative abundance of *Stephanodicus alpinus* Hustedt and a sharp increase in *Aulacoseira islandica* (O. Müller) Simonsen, possibly reflecting increased silica concentrations. The valves of *Aulacoseira islandica* appeared to be highly–silicified in the sediment samples. The second major change is between 20-cm and 10-cm depth where the percentage abundance of *Cyclotella* species and of *Stephanodiscus parvus* Stoermer & Håkansson increases. This change suggests increased nutrient (phosphorus) availability. The reconstructed TP values suggest a rather stable level around 40 μg L^{-1} below 30 cm, above which it increases to c. 70 μg L^{-1} at 20 cm (c. 1950) with a decline to 50–60 μg L^{-1} in the top few centimeters. These diatom–inferred TP concentrations are higher than the actual concentrations measured in the water of Södra Björkfjärden over the past few decades (35), indicating that the model overestimates TP concentrations. Measurements of the surface water (0.5 m depth) gave annual mean concentrations around 30–40 μg TP L^{-1} in the late 1960s and values of 20–30 μg TP L^{-1} in the period since 1970, which suggests that the diatom–inferred TP concentrations are c. 20–30 μg TP L^{-1} higher than true values.

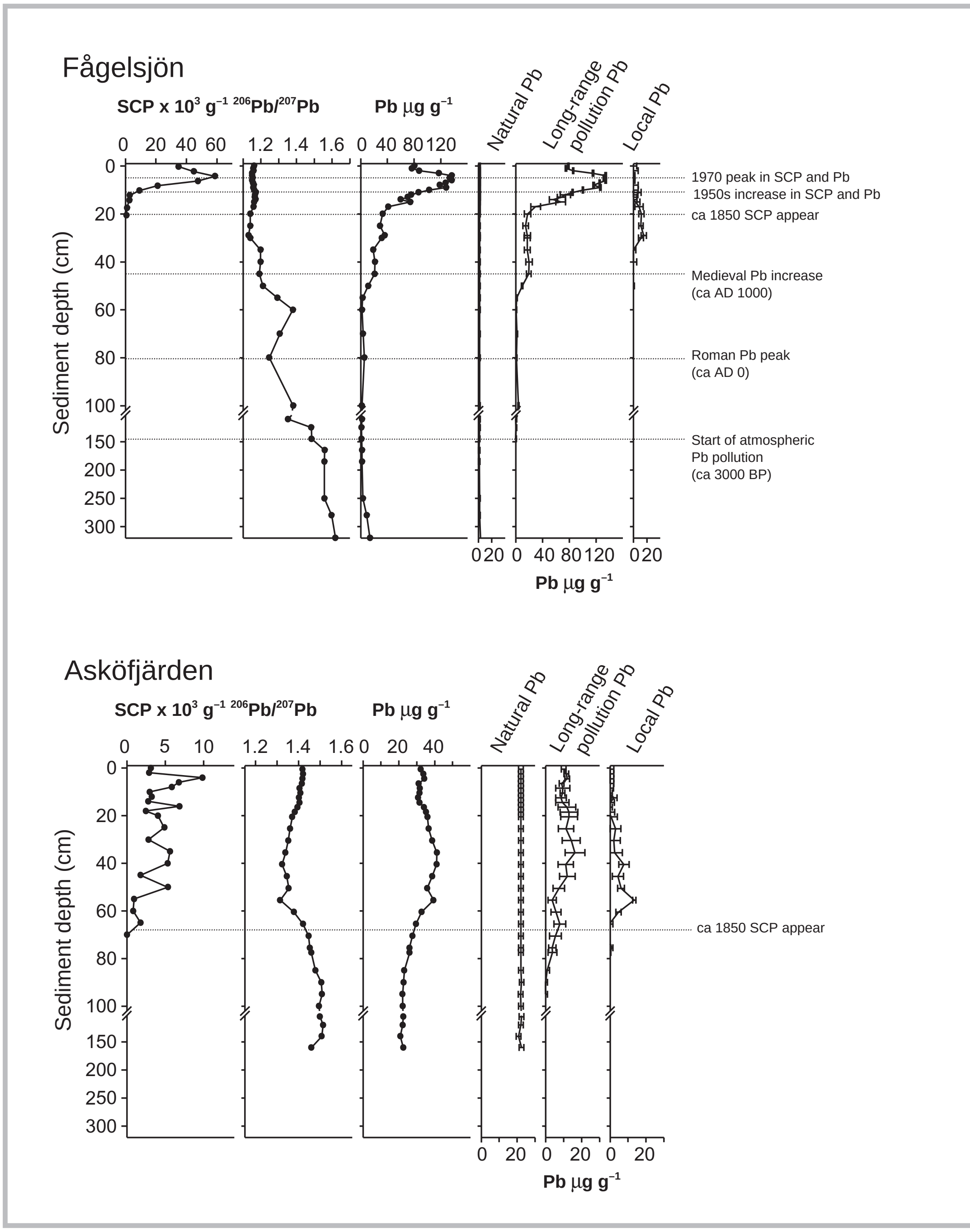
The dataset used to estimate past TP concentrations was developed from relatively small lakes and was validated against monitored TP data for the smaller, but deep Ekoln basin of Mälaren (14). This validation suggests that the model underestimates TP at high concentrations (approx. > 80 μg L^{-1}) and overestimates TP at lower levels. Bradshaw and Anderson's (14) reconstruction of lake–water TP in Ekoln, using the same model as for Södra Björkfjärden, indicated that the diatom–inferred TP values were generally 10–20 μg TP L^{-1} above observed values for the late 20th century. While nearly all the diatom species preserved in the Södra Björkfjärden sediments are represented in the calibration dataset and there are good analogs for the species assemblages of all fossil samples (χ^2 distance dissimilarity < 50%), the dataset does not include good analogs for lake basins of the size of Södra Björkfjärden. Consequently, the current TP model may not be ideal for application to sites of this type.

Despite overestimation in the TP model, there is useful information both in the diatom record at Södra Björkfjärden and the diatom–inferred TP reconstruction. The timing of the changes and the trends in TP concentration are reflected in the sediment record. The increased diatom–inferred TP suggests that more nutrient–rich conditions developed in the lake basin after c. 1850, with a greater increase occurring c. 1950, trends also reflected in the δ^{15}N and pigment records. The more recent decline in phosphorus concentration in the lake is also reflected by the diatom assemblage and diatom–inferred TP. Although the diatom–inferred TP concentrations is obviously overestimated, the diatom assemblage of the pre-19th century period suggests moderately nutrient–rich conditions, perhaps 10–20 μg TP L^{-1}, and not the low concentrations that have been predicted from catchment–lake modeling (~6 μg L^{-1}) (35). Extensive agriculture in the catchment ever since lake formation 1000-yrs ago may have contributed to a nutrient transfer to the lake and elevated lake production. These results are in line with other long-term studies that have shown that the cultural eutrophication of lakes has a history longer than just centuries (36, 37), particularly in Europe where population growth and agricultural development have impacted lakes for thousands of years.

Lead Pollution History

The pollution histories of lead and SCP in the sediments of Fågelsjön generally conform to the established patterns for Swedish lakes receiving their pollution from the atmosphere (26, 38). Besides slightly higher lead concentrations in sediments deposited just after formation of the lake, background lead concentrations are stable and quite low, < 2 μg g^{-1} (Fig. 3). As is typical for old sediments (> 3000-yrs old), the ^{206}Pb/^{207}Pb ratio of the deeper sediments (150-cm to 320-cm depth) is high, 1.6. There is an initial decline in the ^{206}Pb/^{207}Pb ratio at 145 cm depth (inferred age of 3000 BP), a small peak in lead concentration and a corresponding trough in ^{206}Pb/^{207}Pb at 80 cm (Roman lead

Figure 3. Spheroidal carbonaceous flyash particle, lead isotope and lead concentration stratigraphies from the 3 coring sites in Mälaren and in Fågelsjön. In the 3 right panels, the total lead concentration is divided into natural lead (the contribution from the catchment soils), long–distance pollution lead (the fraction derived by atmospheric deposition from distant pollution sources), and the local pollution lead (lead from ores in Bergslagen, transported to the study sites by water and air).



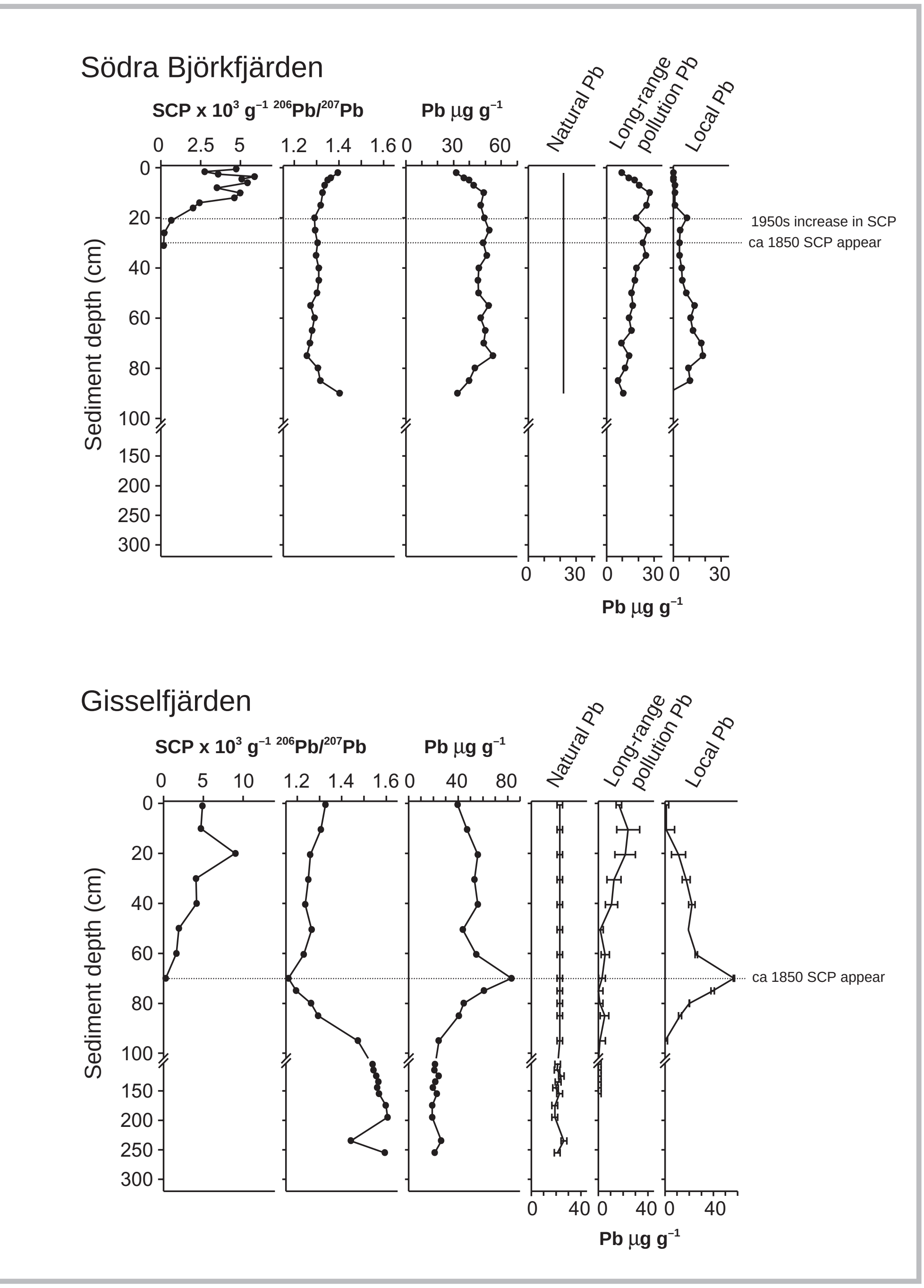
peak, c. AD 0), and increasing lead concentrations and declining isotope ratios above 50 cm (Medieval lead increase, c. AD 1000). The post-war (post-1950) increase and the 1970s peak in SCP and lead from atmospheric deposition are also clearly visible in Fågelsjön.

As in the sediments of Fågelsjön, the post–war increase and 1970s peak in SCP are clearly visible in Södra Björkfjärden. The first SCP appear at 30 cm and their concentration increases rapidly above 20 cm with a peak at 5 cm. In Gisselfjärden and Asköfjärden this pattern is present for SCP, but extended in depth relative to Södra Björkfjärden due to much higher sediment accumulation rates at those sites. In contrast to Fågelsjön and other Swedish lakes, however, there is no post–war increase and 1970s peak in lead concentration in the core from Södra Björkfjärden or in the cores from the two other Mälaren sites. On the contrary, the lead concentration peak precedes the flyash record, and the lead concentration steadily declines while the ^{206}Pb/^{207}Pb ratio increases towards more natural high values during the course of the 20th century. This is an unambiguous sign of a decreasing pollution influx to the sediment.

Why are there no clear signs of the post–war increase and 1970s peak of airborne lead pollution in Mälaren? We suggest that Mälaren was already significantly polluted by the 19th century and earlier as a result of inputs from regional mining and metal production, and that the lake experienced a substantial improvement in the 20th century following closure of the mining and metal industry in Bergslagen and other parts of the catchment. The pre-20th century lead pollution, transported to the lake mainly by water, was so large compared to the modern pollution from car emissions and the burning of fossil fuels, that the latter was overshadowed by the improvement.

The bedrock in the Bergslagen region is rich in minerals. Low technology iron production dates to the Iron Age, and from the Early Medieval period large–scale mining for iron and copper developed (39). Silver and lead production also occurred at a number of sites, of which Sala was the most important. The mining industry in the Sala area dates to the 11th century, and it produced silver and lead from sulfide ores that had a lead content of 50–70% and a silver content of 2–4% (40). Mining ceased at the beginning of the 20th century, but metal production using other raw materials continued until the post–war period. Like at numerous other sites in the catchment of Mälaren, today only mining pits, remains of smelting–houses and other buildings, and heaps of waste rock and slag, remain from this great industrial period, which played a significant role in the economic history of Sweden. Most old metal industries were located at watercourses, which effectively spread the lead pollution and certainly also many other metals to Mälaren. At present, waste and slag, which release metals, pose a threat to the aquatic environment, but these emissions seem low compared to the emissions from the mining and the metal industries when these were still in operation. For lead, at least, the pollution levels continuously improved in the 20th century.

Even though most lead was transported to Mälaren with inlet streams, there was also local air pollution deposition from the many industries in the vicinity of the lake that produced and smelted metals. Evidence of such local pollution is provided by



the sedimentary lead record of Fågelsjön. In Fågelsjön, the ^{206}Pb/^{207}Pb ratio fell below 1.17 in sediments deposited from 30 cm to 20 cm below the sediment surface. Based on the SCP record, we estimate that this corresponds to a period of a few hundred years prior to 1900. Since long–range atmospherically transported lead pollution from Europe had ^{206}Pb/^{207}Pb ratios of about 1.17, deposition of which was substantial in Sweden already in the Medieval period (38, 41), then ratios below 1.17 indicate an important component of lead pollution with a much lower isotope ratio, like that produced in Bergslagen (1.03). It seems likely that Fågelsjön received airborne emissions from some industry in the Södertälje/Stockholm area that used such lead, or it represents a regional signal of air pollution around Mälaren.

Further analyses and radiometric dating of the cores will be required to fully assess the pollution history of Mälaren and its catchment. However, comparison of lead profiles among sites suggests that even roughly–dated sediment cores can be used to reveal variations in the extent, timing and the sources of metal pollution (Fig. 3). For example, Fågelsjön, with a very low natural lead concentration (< 2 µg g^{-1} dry sediment), was sensitive to an influx of early airborne lead pollution from distant European (continental and British Isle) sources. This site discloses a temporal pattern that is typical for Swedish lakes (19), except for a period prior to 1900, when local air pollution had a large influence (Fig. 3). The sediments of Mälaren, on the other hand, have a high natural lead concentration (20–25 µg g^{-1}), and here the influence of pollution was evidenced later. It remains to be exactly dated when local sources around Mälaren started to pollute the lake, but it seems that Södra Björkfjärden was polluted earlier (Medieval time) than Gisselfjärden and Asköfjärden. This is plausible given that Södra Björkfjärden receives water from Sagaån, the river where Sala is situated, while the 2 western sites are affected by mining that commenced later and where silver/lead mining played a minor role early in history.

CONCLUSIONS

Mälaren was culturally eutrophicated already from the formation of the lake in Medieval time. Consequently, it is not possible to define a true natural background (pre–agricultural) total phosphorus (TP) lake–water concentration. Our paleolimnological analyses reveal that TP concentrations from Medieval time until the 19th century were not as low as has been predicted from theoretical modeling (about 6 µg L^{-1}). On the other hand, it was neither as high as our diatom–inferred values suggest (40 µg L^{-1}). Based on an apparent 20–30 µg L^{-1} bias in the modeled TP *versus* actual concentrations, we would infer that background values for Södra Björkfjärden were about 10–20 µg L^{-1}; other basins in Mälaren may have had other background values. We believe a more extensive paleolimnological study could contribute to determining realistic operational TP values, which takes into consideration that the catchment of the lake has always been, and will likely be, influenced by agriculture and other human activities. Such a study should include development of a transfer function more appropriate than the existing one for translating diatom assemblages in lake sediments of large lakes into lake–water TP values, and it should include sediment core studies in other basins of the lake. Despite limitations in the current methods, the reconstructed TP history of Södra Björkfjärden clearly shows a 20th century increase in lake–water phosphorus concentrations as well as declining values in recent time.

Besides the impacts of nutrients, the lead isotopes and lead concentrations of the sediment cores from Mälaren clearly show that the lake was also severely impacted by waterborne emissions for several hundred years from the mining and metal industry in the catchment, perhaps even from Medieval time. Dating of the sediment records using radiometric methods must still be done, but the flyash record indicates that the lead pollution was high throughout the 19th century. The improvement of the pollution situation, following the cessation of the mining and metal production in the 20th century, has been so extensive that the post–war atmospheric lead pollution peak (c. 1970), which is so pronounced in most Swedish lake sediments, is hidden by the improvement. The mining around Mälaren was primarily for iron, copper and silver, which were often produced from complex ores; consequently, other metals should also be analyzed to fully understand the timing and impact of the ancient mining and metal industry.

This study of sediment cores from Mälaren draws attention to early (pre-industrial) impacts on the quality of the lake caused by human activities in the catchment. It contradicts the perception that the pre-industrial (pre-1850) environment represented clean and undisturbed conditions. A lack of a proper understanding of background conditions may lead to unrealistic goals for reductions of current emissions.

References and Notes

1. Wallin, M. (ed.). 2000. Lake Mälaren. Environmental quality and development 1965–98. *Mälaren's Water Protection Association, Report*. County administrative board, Västerås, Sweden, 94 pp. (In Swedish).
2. Lake Mälaren's water: goals and measures. 1993. *Mälaren's Water Protection Association, Report*, County administrative board, Västerås, Sweden, 20 pp. (In Swedish).
3. Risberg, J. Karlsson, S., Hansson, A.-M., Hedenström, A., Heimdahl, J., Miller, U. and Tingvall, C. in review. Environmental changes and human impact as recorded in a sediment sequence off–shore the Viking Age town Birka, south–eastern Sweden. *Holocene*.
4. Ambrosiani, B. 1980. The background of boat burials in the Mälaren valley. In: *Vendeltid*. Historia i Fickformat. Sandwall, A. (ed.). Statens Historiska Museum, Stockholm. (In Swedish).
5. Hansson, P. 1989. *Society and Iron in Sweden during the Iron Age and the Early Middle Ages. The Exemple of Närke*. PhD Dis., Societas Archaeologica Uppsaliensis: Dept. of Archeology, Uppsala University, Sweden, 174 pp. (In Swedish).
6. Welinder, S. 1974. *The Cultural Landscape in the Mälaren Region*. Department of Quaternary Geology, University of Lund, Sweden, Report 5–6. 2 vols., 367 pp. (In Swedish).
7. Borgegård, S.–O. 1990. Agriculture during the Viking Age in the Mälaren area. *Svensk Bot. Tidskr. 84*, 369–392. (In Swedish).
8. Karlsson, S. and Robertsson, A.–M. 1997. Human impact on the Lake Mälaren region during the Viking Age (AD 750–1050): a survey of biostratigraphical evidence. In: *Environment and Vikings.* Miller, U. and Clarke, H. (eds). *Birka Studies 4*, 47–72.
9. Callmer, J. 1994. Urbanization in Scandinavia and the Baltic Region *c* AD 700–1100: Trading places, centres and early urban sites. In: *Developments around the Baltic and the North Sea in the Viking Age: The Twelfth Viking Congress.* Ambrosiani, B. and Clarke, H. (eds). Birka Studies 3, 50–90.
10. Eriksson, G.A. 1955. *The Decline of the Small Blast–furnaces and Forges in Bergslagen after 1850*. Jernkontorets Bergshistoriska Skriftserie 15. PhD Dis., Dept. of Geography, Uppsala University, Sweden, 347 pp.
11. Renberg, I. 1999. *Nutrient Conditions in Södra Björkfjärden, Lake Mälaren, in the Last Thousand Years: A Paleolimnological Investigation*. Dept. of Ecology and Environmental Science, Umeå University, Sweden, 17 pp. (In Swedish).
12. Krammer, K. and Lange–Bertalot, H. 1986–1990. *Süsswasserflora von Mitteleuropa. Bacillariophyceae.* Vol. 1–4. Gustav Fischer Verlag, Stuttgart.
13. Wunsam, S., Schmidt, R. and Klee, R. 1995. *Cyclotella*–taxa (Bacillariophyceae) in lakes of the Alpine region and their relationship to environmental variables. *Aquatic Sci. 57*, 1615–1621.
14. Bradshaw, E.G. and Anderson, N.J. 2001. Validation of a diatom–phosphorus calibration set for Sweden. *Freshwater Biol. 46*, 1035–1048.
15. Juggins, S. and ter Braak, C.J.F. 1997. *CALIBRATE—A Computer Program for Species—Environmental Calibration by (Weighted–Averaging) Partial Least Squares Regression (0.81)*. University of Newcastle, UK.
16. Line, J.M., ter Braak, C.J.F. and Birks, H.J.B. 1994. WACALIB version 3.3—a computer program to reconstruct environmental variables from fossil assemblages by weighted averaging and to derive sample–specific errors of prediction. *J. Paleolimnol. 10*, 147–152.
17. Wright, S.W, Jeffrey, S.W., Mantoura, R.F.C., Lewellyn, C.A., Bjornland, T., Repeta, D. and Welschmeyer, N. 1991. Improved HPLC method for the analysis of chlorophylls and carotenoids from marine phytoplankton. *Mar. Ecol. Prog. Ser. 77*, 183–196.
18. Ohlsson, K.E.A. and Wallmark, P.H. 1999. Novel calibration with correction for drift and non–linear response for continuous flow isotope ratio mass spectrometry applied to the determination of δ^{15}N, total nitrogen, δ^{13}C and total carbon in biological material. *Analyst 124*, 571–577.
19. Brännvall, M.–L., Bindler, R., Emteryd, O. and Renberg, I. 2001. Four thousand years of atmospheric lead pollution in northern Europe: a summary from Swedish lake sediments. *J. Paleolimnol. 25*, 421–435.
20. Chow, T.J. 1965. Radiogenic leads of the Canadian and Baltic Shield regions. *Symp. Mar. Geochem. 3*, 169–184.
21. Hopper, J.F., Ross, H.B., Sturges, W.T. and Barrie, L.A. 1991. Regional source discrimination of atmospheric aerosols in Europe using the isotopic composition of lead. *Tellus 43B*, 45–60.
22. Johansson, Å. and Rickard, D. 1984. Isotopic composition of Phanerozoic ore leads from the Swedish segment of the Fennoscandian shield. *Mineral. Depos. 19*, 249–255.
23. Renberg, I., Brännvall, M.–L., Bindler, R. and Emteryd, O. Stable lead isotopes and lake sediments—a useful combination for the study of atmospheric lead pollution history. *Sci. Tot. Environ.* (In print).
24. Farmer, J.G., Eades, L.J., MacKenzie, A.B., Kirika, A. and Bailey–Watts, T.E. 1996. Stable lead isotope record of lead pollution in Loch Lomond sediments since 1630 A.D. *Environ. Sci. Technol. 30*, 3080–3083.
25. Renberg, I., Bindler, R. and Brännvall, M.–L. 2001. Using the historical atmospheric lead deposition record as a chronological marker in sediment deposits in Europe. *Holocene 11*, 511–516.
26. Wik, M. and Renberg, I. 1996. Environmental records of carbonaceous fly–ash particles from fossil–fuel combustion. *J. Paleolimnol. 15*, 193–206.
27. Meyers, P.A. and Ishiwatari, R. 1993. Review paper: Lacustrine organic geochemistry – an overview of indicators of organic matter source and diagenesis in lake sediments. *Organic Geochem. 20*, 867–900.
28. Schelske, C.L. and Hodell, D.A. 1995. Using carbon isotopes of bulk sedimentary organic matter to reconstruct the history of nutrient loading and eutrophication in Lake Erie. *Limnol. Oceanogr. 40*, 919–929.
29. Struck, U., Emeis, K.–C., Voss, M., Christiansen, C. and Kunzendorf, H. 2000. Records of southern and central Baltic Sea eutrophication in δ^{13}C and δ^{15}N of sedimentary organic matter. *Mar. Geol. 164*, 157–171.
30. Hodell, D.A. and Schelske, C.L. 1998. Production, sedimentation, and isotopic composition of organic matter in Lake Ontario. *Limnol. Oceanogr. 43*, 200–214.
31. Leavitt, P.R. 1993. A review of factors that regulate carotenoid and chlorophyll deposition and fossil pigment abundance. *J. Paleolimnol. 9*, 109–127.
32. Cuddington, K. and Leavitt, P.R. 1999. An individual–based model of pigment flux in lakes: implications for organic biogeochemistry and paleoecology. *Can. J. Fish. Aquat. Sci. 56*, 1964–1977.
33. Willén, E. 1991. Planktonic diatoms—an ecological review. *Algol. Stud. 62*, 69–106.
34. Stoermer, E.F. and Smol, J.P. 1999. *The Diatoms: Applications for the Environmental and Earth Sciences.* Cambridge University Press, Cambridge. 484 pp.
35. Persson, G., Olsson, H. and Willén, E. 1990. Lake Mälaren's water quality during 20 years. 1. Nutrients, transport, concentrations and plankton biomass. *Swedish Environmental Protection Agency, Report 3759*, 47 pp. (In Swedish).
36. Fritz, S.C. 1989. Lake development and limnological response to prehistoric and historic land–use in Diss, Norfolk, UK. *J. Ecol. 77*, 182–202.
37. Bradshaw, E.G. 2001. *Linking Land and Lake. The Response of Lake Nutrient Regimes and Diatoms to Long-term Land-use Change in Denmark*. PhD thesis, University of Copenhagen, Denmark. 118 pp.
38. Renberg, I., Brännvall, M.–L., Bindler, R. and Emteryd, O. 2000. Atmospheric lead pollution history during four millennia (2000 BC to 2000 AD) in Sweden. *Ambio 29*, 150–156.
39. Backlund, A.–C. (ed.). 1988. *The Book on Bergslagen: a Journey Through a Living History.* Rubicon, Stockholm, 224 pp. (In Swedish).
40. Meurman, R. 2000. *Silver Mountains in Iron Ore Country. The Beginning of Mining as Refelcted in Schmidt's Testhammer Datings.* PhD Diss., Dept. of Archeology and Ancient History, Uppsala University, Sweden, 184 pp. (In Swedish).
41. Brännvall, M.–L., Bindler, R., Renberg, I., Emteryd, O., Bartnicki, J. and Billström, K. 1999. The Medieval metal industry was the cradle of modern large–scale atmospheric lead pollution in northern Europe. *Environ. Sci. Technol. 33*, 4391–4395.
42. Fredén, C. (ed.). 1994. *National Atlas of Sweden: Bedrock and Soils*. Sveriges Nationalatlas, 208 pp. (In Swedish).

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