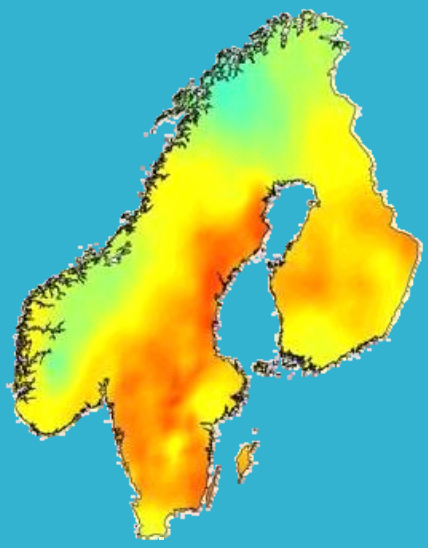
REPORT



Mercury in Nordic ecosystems

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Summary in Swedish

För första gången presenteras här en omfattande sammanställning av data angående atmosfärisk deposition av kvicksilver i Norden samt förekomst av kvicksilver i sediment och i insjöfisk. Undersökningen visar att, trots emissionsbegränsningar i Norden och Europa så påverkas nordiska ekosystem alltjämt av långväga transport av kvicksilver. Den geografiska utbredningen av kvicksilver i sediment och fisk beror på nedfallets genografiska utbredning men är även beroende av specifika egenskaper hos enskilda ekosystem. Utvärdering av kvicksilverförekomst i luft och nederbörd visar tydligt att långväga transport av kvicksilver från europeiska källor alltjämt bidrar till att förorena miljön i Norden, men att emissioner på en global skala också bidrar.

Med hjälp av en omfattande sammanställning och utvärdering av data av kvicksilver i insjöfisk har geografiska variationer i förekomsten i de nordiska länderna kartlagts. Vidare jämförs resultatet av karteringen med geografiska variationer i kvicksilverdeposition och förekomst av kvicksilver i sediment i syfte att undersöka kopplingen mellan långväga transport av kvicksilver och metylkvicksilver i fisk.

Utvärderingen av kvicksilverhalter i nederbörd visar att fortsatt finns en påverkan från antropogena källor i Europa. Årsmedeldepositionen av kvicksilver varierar kraftigt och är beroende av nederbördsmängder. En utvärdering av mätdata från stationer kring Nordsjön visar att kvicksilverhalter i nederbörd har minskat år från år sedan början av 1990 talet (Wängberg et al., 2007). Orsaken är minskande kvicksilverutsläpp från Europeiska källor. Emissioner av gasformigt kvicksilver (TGM) minskade också i början av 1990 talet vilket gav upphov till en signifikant minskning av TGM i bakgrundsluft. Dock syns ingen minskning på senare år och halterna av TGM i bakgrundsluft i Skandinavien har nu nått en nivå som närmar sig den hemisfäriska bakgrundshalten.

Kvicksilver i sediment från insjöar i Norge, Sverige och Finland har undersökts. Jämförelser mellan kvicksilverhalter i yt- och bottensediment, där de senare härrör från år 1600 till 1850, visar att kvicksilverhalten i sjösediment ökat med en faktor 2-5 sedan industrialiseringen. De mest förorenade sedimenten påträffades i kustområden i södra Norge, i sydvästra Finland och i Sverige i ett bälte från västkusten i Sydsverige via Mellansverige till Bottenviken i nordost. Dock indikerar en undersökning av ytsediment (från 0 - 0.5 cm djup) och sub-ytsediment (från 0.5 -1.0 cm djup) från sjöar i Norge att kvicksilver i ytsediment har minskat något på senare år, vilket stämmer väl överens med minskade kvicksilverhalter i deposition som nämns ovan.

Utredningen visar att förekomsten av förhöjda kvicksilverhalter i insjöfisk i stora drag överensstämmer med utbredningen av kvicksilverförorenade sediment. Fördelningen av sjöar med förorenade sediment kan i sin korrelera väl med nuvarande depositionsmönster av kvicksilver. I vissa fall kan dock även utsläpp av kvicksilver från tidigare industriella verksamheter påverka nivåerna. Emellertid är kopplingen mellan förekomst av kvicksilver i miljön, förorsakad av långväga transport eller lokala utsläpp, och metylkvicksilver i fisk mer komplicerad. En detaljerad riskbedömning kräver att även lokala faktorer beaktas, såsom avrinningsområdens storlek, dess innehåll av våtmarker samt halt och mobilitet av DOC i avrinning och ytvatten. Dessutom finns variationer mellan olika akvatiska ekosystems benägenhet att bilda metylkvicksilver.

Summary

This report provides a first comprehensive compilation and assessment of available data on mercury in air, precipitation, sediments and fish in the Nordic countries. The main conclusion is that mercury levels in Nordic ecosystems continue to be affected by long-range atmospheric transport. The geographical patterns of mercury concentrations in both sediments and fish are also strongly affected by ecosystem characteristics and in some regions possibly by historical pollution.

An evaluation of geographical variations in mercury concentrations in precipitation indicates that the influence from anthropogenic sources from Central European areas is still significant. The annual variability of deposition is large and dependant of precipitation amounts. An evaluation of data from stations around the North Sea has indicated a significant decrease in mercury concentrations in precipitation indicating a continuous decrease of emissions in Europe (Wängberg et al., 2007). For mercury in air (TGM), the geographical pattern is less pronounced indicating the influence of mercury emissions and distribution over a larger geographical area (i.e. hemispherical transport).

Comparison of recent (surficial) and historical lake sediments show significantly elevated concentrations of mercury most likely caused by anthropogenic atmospheric deposition over the past century. The highest pollution impact was observed in the coastal areas of southern Norway, in south western Finland and in Sweden from the coastal areas in the southwest across the central parts to the north-east. The general increase in recent versus old sediments was 2-5 fold.

Data on mercury in Nordic freshwater fish was assembled and evaluated with respect to geographical variations. The fish data were further compared with temporal and spatial trends in mercury deposition and mercury contamination of lake sediments in order to investigate the coupling between atmospheric transport and deposition of mercury and local mercury pollution effects (i.e. methylmercury in fish).

The general pattern of fish contamination follows to some extent a pattern similar to that of current and previous atmospheric pollution. Large areas have fish with mercury concentrations exceeding the health advisory guideline of 0.5 mg/kg or 1.0 mg/kg (for northern pike) in the EU and of around 0.3 mg kg^{-1} in the USA, thus restricting their use for human consumption.

A more comprehensive assessment of factors influencing levels of methylmercury in fish has to include a number of other parameters such as catchment characteristics (*e.g.* relative size, presence of wetlands), contents and fluxes of DOC in soil run-off and surface waters as well as methylation potential within ecosystems.

Content

Introduction

Mercury remains a problem in many parts of the world with contamination of terrestrial and aquatic ecosystems and resulting risks of human exposure mainly via consumption of contaminated fish. Measures to control emissions of mercury to the environment have led to decreasing emissions in many countries in Europe and North America. This decrease is to some extent compensated by increased emissions in other parts of the world. Given the specific properties of mercury, atmospheric long-range transport may occur over hemispheric or even global scales thus counteracting regional efforts to reduce this problem. A successful strategy to develop long-term solutions to the mercury problem should thus be based on activities on local, regional and global scales.

The Nordic countries Sweden, Norway and Finland have a common situation with atmospheric mercury originating from long-range transport being the main source to ecosystem contamination. Many lakes are contaminated to a level where the fish are no longer suitable for consumption. The contamination is a result of decades of elevated mercury input via the atmosphere and, in some specific locations, direct releases to water bodies. Uptake and accumulation of mercury in aquatic food chains is a complex process involving many steps of transport and transformations. The level of contamination of fish is thus not only directly linked to the mercury input but also to external factors which may influence these processes. Examples of this are land use changes, climate and hydrology.

This report was initiated with the intention to compile and summaries the level of contamination of Nordic ecosystems by mercury. The data compilation and analysis is focused on atmospheric concentrations and deposition, concentrations in fish and in sediments.

1 Atmospheric Mercury in Scandinavia

Due to its high vapour pressure in combination with its relative high thermodynamic stability, mercury is predominately emitted to the atmosphere as Gaseous Elemental Mercury (GEM). In contrast to other heavy metals, most of the mercury in the atmosphere is in the gaseous phase rather than bound to particles. The atmospheric residence time of GEM has been estimated to be about one year. This is long enough to allow distribution on a hemispherical scale. Hence, the atmosphere constitutes an efficient means for transport of mercury. GEM is only slowly removed from the atmosphere by washout or dry deposition, but may be adsorbed onto soot particles or be oxidised to divalent soluble mercury compounds that more easily are removed from the atmosphere. Thus, rain and snow contains oxidised mercury and wet deposition of mercury constitutes a constant input of mercury to land and aquatic environments. The deposited mercury is accumulated in the soil or transported to streams or lakes where it may be available for bioaccumulation. Here mercury measurements from five Scandinavian EMEP stations are evaluated. The measurements were made during 1996-2002 and the result is used to map the mercury deposition and distribution in Scandinavia.

1.1 Field Measurements

The locations of the EMEP stations where precipitation samples were collected are shown in Figure 1. Concentrations of TGM were obtained from three of these sites, Rörvik, Lista and Pallas. Rörvik is an EMEP station which has been in use since 1977. The station was moved to Råö, a site 2.6 km south west of the Rörvik station, 1st of January 2002. In the Figures below the denotation Rörvik/Råö is therefore used. Precipitation samples for Hg(tot) analysis were collected using bulk samplers (Berg et al., 2002). The collector is equipped with electrical heating and can therefore also be used during the cold season. The sampling was performed on monthly basis. All precipitation samples were analysed by acid digestion followed by reduction with Sn(II) using the purge and trap method and detection of gaseous elemental mercury with CVAFS (Brosset, 1987; Bloom and Fitzgerald, 1988). Annual mercury deposition values were calculated using equation I,

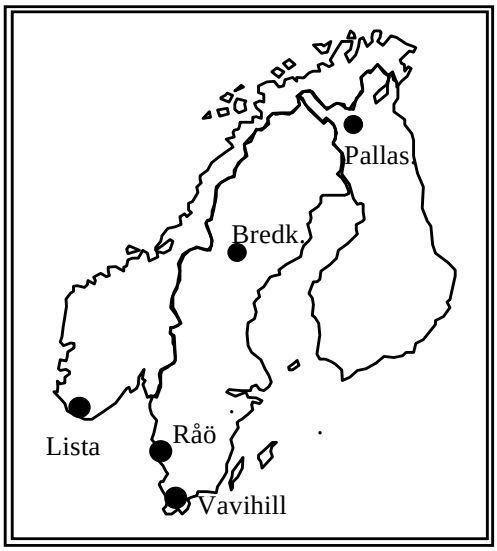

Figure 1. EMEP stations where mercury in air and precipitation were measured.

where $Hg(tot)_i$ (ng L^{-1}) denotes the mercury concentration in each precipitation sample. The precipitation amount associated with each sample is indicated by P_i (mm, 1 mm = 1 L m^{-2}).

$$\text{Hg deposition} = \frac{1}{1000}\sum_{i=1}^{12}(P_i \cdot Hg(tot)_i) \qquad [\mu g\ m^{-2}] \qquad (I)$$

The sum i = 1 to 12 corresponds to monthly samples collected during one year. Yearly averaged [Hg]tot concentrations in precipitation were calculated according to,

$$[Hg]tot = \frac{\sum_{i=1}^{12} P_i \cdot Hg(tot)}{\sum_{i=1}^{12} P_i} \qquad [\mu g\ m^{-3}] \qquad (II)$$

TGM is an operational defined gaseous mercury fraction present in ambient air, which consists of GEM plus a small fraction (often less than 3%) of oxidised gaseous mercury. TGM was measured using the manual gold-amalgamation technique. Air is pulled through a quartz glass tubes containing an Au adsorbent. The sampling time is 24 h and the total air volume is measured with a gas volume meter. The sample is analysed by thermal desorption and mercury detection with CVAFS (Brosset, 1987; Bloom and Fitzgerald, 1988). Yearly average TGM data used in this evaluation is based on 24 h measurements made 1 to 2 times per week.

Total Particulate Mercury (TPM). Particles for mercury analysis were sampled using an open phase mini particle sampler. A quartz micro-fibre filter (Munktell MK 360) of 7 mm diameter is housed in a quartz glass tube of 140 mm length. The filter is supported by a pure Ni-screen grid. The sampling device serves as both particulate trap and pyrolyzer for airborne particulate mercury. Air is drawn through the quartz tube at a flow rate of 3-5 L min^{-1}. After sampling, the mercury content is analysed via pyrolysis where the trap is heated to 800-900 ^{o}C in a stream of argon. In this step, all mercury is decomposed to Hg^0 and subsequently transferred to the gas phase and detected by CVAFS.

1.2 Mercury Wet Deposition and Mercury Concentrations in the Atmosphere

Annual Hg(tot) deposition during 1999 – 2002 at the five Scandinavian sites is shown in Figure 2. The highest deposition was obtained at the Lista station. As is apparent from Figure 2 the mercury deposition tends to vary somewhat from year to year.

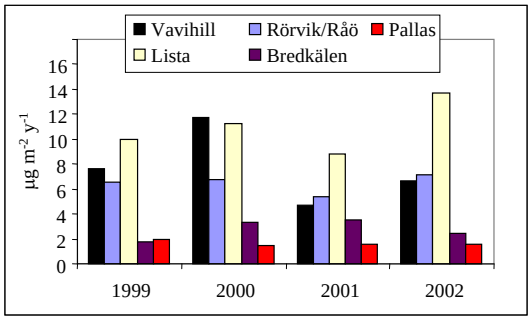

Figure 2 Hg(tot) wet deposition in Scandinavia 1999 to 2002

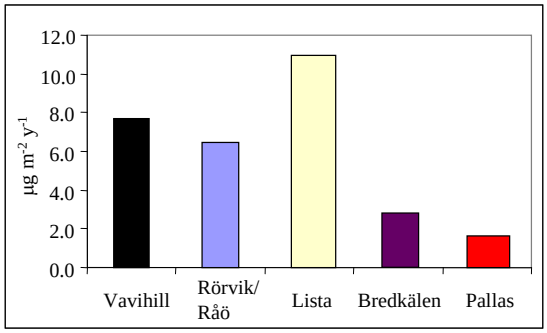

Figure 3 Average annual mercury wet deposition during 1999-2002.

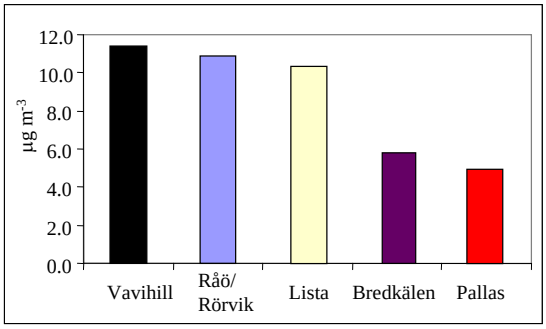

Figure 4 Yearly averaged [Hg](tot) concentrations in during precipitation during 1999-2002.

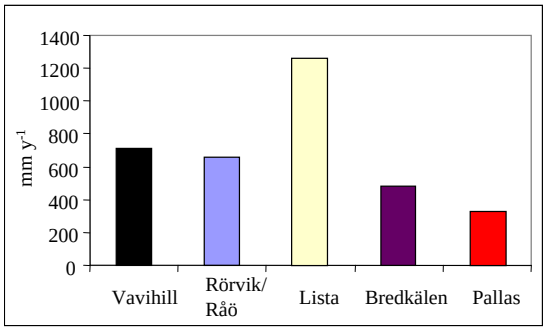

Figure 5 Average annual deposition amounts 1999-2002

In order to compare the mercury deposition at the five sites and look for geographical trends average deposition values were calculated. The result is shown in Figure 3. It appears to be a geographical trend with decreasing mercury deposition from south to north. The Lista station seems not to fit into this trend, however. If instead comparing mercury concentration, [Hg](tot), in precipitation as is made in Figure 4, then all measurement sites fits into a south to north decreasing trend. The total deposition flux of mercury at a certain site depends on the mercury concentration in precipitation times the precipitation amount. Average precipitation amounts are shown in Figure 5 and it is obvious that the large mercury deposition obtained at Lista is due to the rather large deposition amounts received there. Hence the actual mercury deposition at a site is dependant of both [Hg](tot) (which has a geographical component) and deposition amounts. This influence also explains the great difference in mercury deposition between Vavihill and Pallas. The average [Hg]tot during 1999-2002 was 11.5 µg m^{-3} at Vavihill. This is more than two times higher than at Pallas (5.0 µg m^{-3}). However, since the precipitation amounts are also a factor of two higher at Vavihill in comparison to Pallas, the mercury deposition in the south is almost 5 times higher than in the north. TGM is only measured at three EMEP stations in Scandinavia. Annual average values from 1996-2002 are shown in Figure 6. The variation between stations and the variation from year to year is quite small, all data are confined within the range 1.30-1.86 ng m^{-3}, but the difference between individual samples may be much greater. Average 24 h TGM values between 1.1 and 4.2 ng m^{-3} was measured at the Råö site during 2002, for example. However, 80% of all measurements yielded concentrations within ± 0.3 ng m^{-3} around the annual average concentration. Due to the relatively long atmospheric lifetime of mercury, annual TGM averages from Scandinavian background sites are likely to reflect the hemispherical background concentration. That is, the influence from regional sources is expected to be low. The average TGM at Lista during the whole period 1996-2002 is 1.81 ng m^{-3}. Slemr et al., 2003, estimated the background TGM concentration

in the Northern Hemisphere to be 1.7 ng m^{-3} during 1996 to 2000. The average TGM in the Northern Atlantic Ocean was estimated to be somewhat higher, 1.88 ng m^{-3}, during the same period. It should be mentioned that these estimates are based on a relatively limited number of data, including the measurements at Lista, but stem also from ship cruises and measurements at some additional sites. However, the values from Lista should represent that of the westerly border input of TGM to Northern Europe. On the other hand, the corresponding value from Rörvik/Råö (1.6 ng m^{-3}) is more difficult to interpret. Due to the proximity to mercury sources in Europe one should expect the concentrations to be somewhat higher there then at Lista, but it seems to be the opposite. We are not able to explain this observation at present. The TGM concentrations at Pallas are somewhat lower in comparison to those measured at Rörvik/Råö. The average TGM calculated for 1996-2002 is 1.5 ng m^{-3} at Pallas. This difference is probably on the limit to be significant considering the precision of the measurements, but may reflect the proximity of Rörvik/Råö to European source areas as is discussed below. It can be concluded that average TGM concentrations at the Scandinavian EMEP stations were within 1.5 - 1.8 ng m^{-3} during the investigated period.

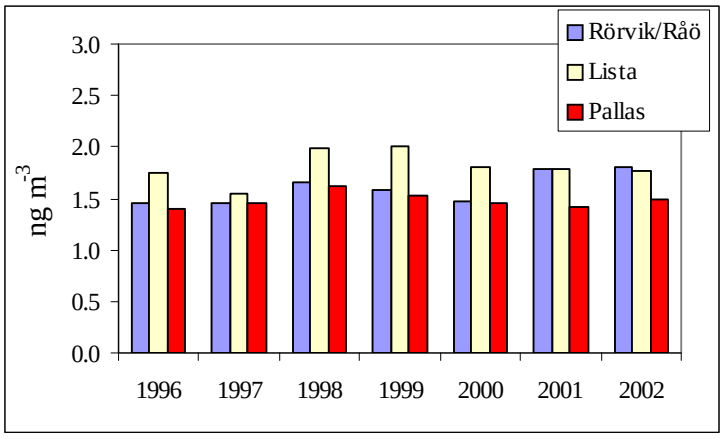

Figure 6 TGM at three EMEP stations during 1999-2002.

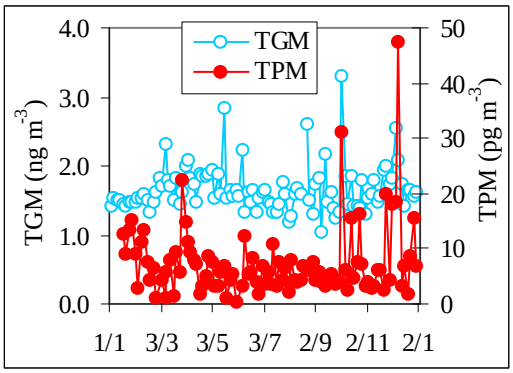

Figure 7. 24 h TGM and TPM samples at Rörvik 2001.

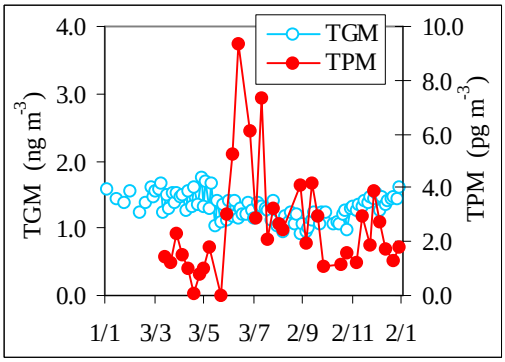

Figure 8. 24 h TGM and TPM samples at Pallas 2001

Figure 7 and Figure 8 show typical 24 h TGM concentrations from Rörvik and Pallas. TPM data from the two sites is also shown. As mentioned earlier TGM values from these sites are quite similar. On the other hand, TPM concentrations between the sites differ considerably. The two TPM data sets are not directly comparable because the Pallas values correspond to 7 days average measurements while the Rörvik data is 24 h averages. However, TPM at Rörvik/Råö is 3 to 6 times higher than that of Pallas on a yearly average basis. Characteristic for the Rörvik/Råö data is

occasions with elevated TPM concentrations. Some of these events has been investigated and according to back trajectory analysis peaks in TPM is often due to long range transport from Poland or the former East Germany (Wängberg et al., 2003). The TPM study showed a similar influence from these sources on the southern part of Sweden as is presented here concerning mercury deposition. Observations of TPM can be used to detect long range transport of mercury. In this respect TPM is a much more sensitive parameter than TGM, Wängberg et al., (2003). As shown in Figure 7 some days with elevated TPM coincide with elevated TGM, however the variation in TPM is more dynamic. A similar evaluation of the TPM data from Pallas is unfortunately not possible due to the lower temporal resolution of these measurements. However, one may guess that due to the large distance the influence from sources in Central Europe is likely to be small in Pallas.

To conclude, a geographical trend with higher deposition of mercury in southern Scandinavia than in the north is observed. The reason for this is likely to be due to mercury emissions from Europe and especially Central Europe. Here only TPM data from two measurement sites are presented. But the present data in combination with earlier measurements performed within the MOE project during 1998 - 1999 (Munthe et al., 2003; Wängberg et al., 2003) strongly indicate that the distribution of TPM also has a similar geographical trend as mercury deposition. Regarding TGM the difference between south and northern Scandinavia is small. Much higher TGM values were measured earlier. During the period 1985-1989 the average TGM concentration was reported to be 3.2 ng m^{-3} at Rörvik and 2.7 ng m^{-3} during 1990-1992 (Iverfeldt et al., 1995). Iverfeldt et al. (1995) concluded that the decrease in atmospheric burden of mercury was due to decrease in mercury emissions from European source areas. This dramatic decrease occurred around 1990 when the economy in the former East block collapsed and many coal fired power plants, chlor-alkali plants using mercury cells etc. were closed down. Wet deposition of mercury as well as of sulphur also decreased dramatically during these years. The mercury deposition at Rörvik during 1989-1990 was about 2.5 times higher than at the present, Iverfeldt et al. (1995). A slight decrease has also been observed in deposition data from stations around the North Sea (Wängberg et al., 2007) most likely due to emission reductions in Europe. The reason why we do not see a strong south-north decreasing gradient in TGM, nor a significant decrease over time is that the average TGM value in southern Scandinavia as well as in most of the Southern Baltic Sea area now is close to the hemispherical background concentration and thus influenced by emission trends over a larger geographical area (Wängberg et al., 2007).

1.3 Deposition of mercury in forests

Mercury is deposited to forested ecosystems via wet and dry processes. The presence of the forest canopy enhances dry deposition since the large surface area allows interactions between gaseous mercury species and needles, leaves etc. Dry deposition is believed to occur mainly via mercury in the form of RGM (Reactive Gaseous Mercury), an operationally defined fraction most likely in the form of $HgCl_2$, and via deposition of particulate mercury. A part of the dry deposited mercury in the forest canopy is washed off by precipitation and deposited via throughfall. The remainder is deposited to the forest floor via litterfall (i.e. needles, leaves, branches). Most of the mercury deposited to the forest floor is bound strongly to organic matter and accumulated in the soil. A small fraction of the mercury will leave the forest floor via re-emission of elemental mercury and run-off. The main fluxes of mercury in forested ecosystems are depicted in Figure 9.

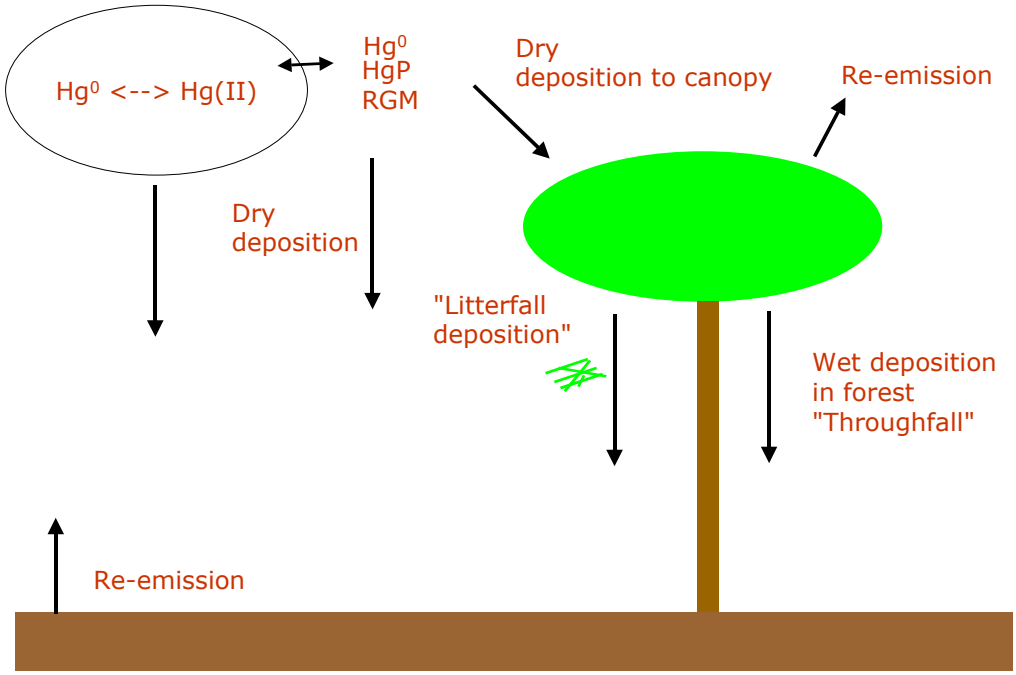

Figure 9 Atmospheric fluxes of mercury in forested ecosystems

1.4 Measurement data

A limited number of studies have been performed on mercury deposition in forested ecosystems in Europe and North America e.g. in Sweden (Iverfeldt, 1991; Hultberg *et al.,* 1994; Munthe *et al.,* 1995a,b; Lee *et al.,* 1994, 2000), North America (St Louis *et al.,* 1996; Driscoll *et al.,* 1998), and Germany (Schwesig *et al.,* 1999; Schwesig and Matzner, 2000). All results show that deposition of mercury is greatly enhanced by the air-canopy interactions i.e. by dry deposition.

In Table 1, some examples of measured deposition fluxes of mercury in Europe are given.

Table 1. Deposition fluxes of TotHg and MeHg in forested ecosystems.

Site	Wet deposition (open field), µg m^{-2} yr^{-1} TotHg/MeHg	Total deposition (forest) µg m^{-2} yr^{-1} TotHg/MeHg	Ratio Total/wet TotHg/MeHg	Reference
Uraani, FI	5.1/0.09	37/0.27	7.3/3.0	Porvari and Verta, 2003
Svartberget, SE	7/0.08	33/0.47	4.7	Lee *et al.*, 1994, 2000
Gårdsjön, SE	10/0.12	46/0.75	4.6	Munthe et al., 1995a, b

Although the fluxes vary between the different locations, the data shows that total deposition is enhanced by a factor of 4.6 to 7.3 in comparison to wet deposition.

The measured wet deposition levels in Scandinavia (Figure 3, Table 1) are almost identical with wet deposition in 13 monitoring stations in North-East America with annual variation from 3.1 to 10.6 µg/m^2 at different stations during 1996-2002 (Vanarsdale et al. 2005).

1.5 Concluding remarks - atmosphere

The concentration of mercury in precipitation is 10-11 μg m^{-3} in Southern Scandinavia but only 5-6 μg m^{-3} at inland sites in Northern Sweden and Finland. These observations can be interpreted in terms of a north-south gradient caused by emissions of mercury in Central Europe. The mercury wet deposition at various sites is also strongly determined by local precipitation amounts. Hence, coastal areas with frequent rain events receive more mercury than more dry regions. A similar source receptor relation is also found for TPM, while the distribution of TGM is more uniform. The reason for this is obviously connected to the much longer residence time of elemental mercury making the average concentration of this compound some 100 times higher than for example TPM. The mercury emissions from European sources decreased dramatically during the 1990-1995. This change reduced the mercury deposition and also TGM and TPM concentrations in Southern Scandinavia. In recent years, observations suggest that a slight decrease of wet deposition has occurred while TGM levels are more or less constant.

2 Mercury contamination of Nordic lake sediments

2.1 Introduction

Mercury contamination of fresh water and their biota has become a widespread and serious problem in many parts of the world, including the Nordic countries. In the Northern Hemisphere, anthropogenic emissions have increased the background concentrations of mercury in air by a factor of 2-3 since before industrialisation (EU 2001, Lindqvist et al. 1991, Landers et al. 1998). Although the emission sources of mercury deposited in the Nordic countries are mainly located in other parts of Europe and the world, a large amount of mercury has previously been emitted to the atmosphere from local point sources, mainly chloralkali plants, in Sweden (Lindqvist et al. 1991), Finland (Lodenius 1985) and Norway (Rognerud and Fjeld 1990). The total emissions (1935-1980) to the atmosphere from these point sources within the region can be estimated at approximately 1000 tonnes (Figure 10).

Since the 1980's, emissions of mercury to the atmosphere from the Nordic countries have declined significantly (Munthe et al. 2001). In the absence of a comprehensive network of atmospheric monitoring stations for mercury deposition, analyses of lake sediments can be used as a proxy for spatial and temporal trends in deposition (Landers et al. 1998, Johansson et al. 2001, Rognerud and Fjeld 2001). Mercury deposited on the lake surface, and supplied from the catchment, is likely to be scavenged by particles and deposited to sediments. Most of the mercury retained in lakes eventually becomes associated with bottom sediments. In the deepest part of a lake, sediment-associated mercury can be buried chronologically and be isolated from active biogeochemical cycling.

However, mercury in surface sediments serves as a source of potentially available mercury for bacterially mediated production of methyl mercury, which accumulates in biota and biomagnifies in food chains. Generally, the highest concentrations of mercury in piscivorous fish in Fennoscandia have been observed in regions where high concentrations of mercury in surface sediments are frequently observed (Håkanson et al. 1988, Rognerud et al. 1996, 2002). In Finland, south-central regions have revealed higher concentrations than northern regions, both in sediment and in fish (Verta 1990).

Lakes are an important ecological habitat in the Nordic countries, and lake sediments have been used to uncover temporal and spatial trends in deposition of mercury and other heavy metals in Sweden (Johansson 1985, Bindler et al. 2001, Johansson et al. 2001), Finland (Verta et al. 1989, Mannio 2001) and Norway (Rognerud and Fjeld 2001). Here we report the spatial distribution of mercury concentrations in lake sediments from almost 400 lakes distributed throughout Fennoscandia. Also, we show temporal trends indicated by differences in LOI-adjusted mercury concentrations in surface and subsurface sediments from 210 lakes in Norway as a proxy for historical changes in atmospheric mercury deposition between the late 1980s and first part of the 1990s.

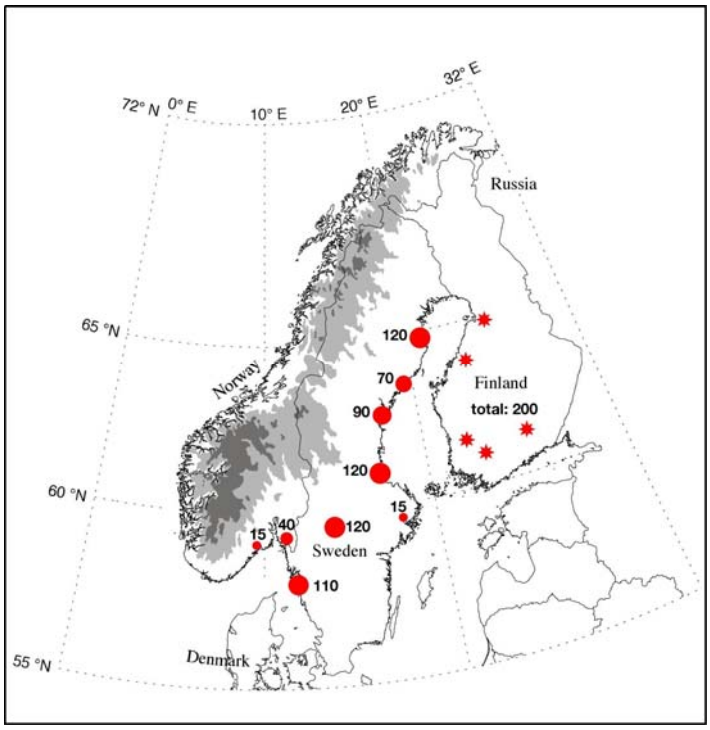

Figure 10 Accumulated mercury emissions to the atmosphere (tonnes) from point sources in Fennoscandia (1940 – 1980). Data collected from: Lindqvist et al. 1991 (Sweden), SFT 1988 (Norway), and calculated from Lodenius 1985 and Mukherjee 1989 (Finland). The map also shows the mountain areas, with light shadings representing 500–1000 m a.s.l., and dark shadings representing areas >1000 m a.s.l.

2.2 Materials and Methods

2.2.1 Sampling programs and lake selection

Here we present mercury concentrations in lake sediments from 389 Nordic lakes (231 located in Norway, 99 in Sweden and 59 in Finland). The Norwegian lakes were sampled during a countrywide survey in 1996–1997 that is part of a national monitoring program conducted by the Norwegian Pollution Control Authority (Rognerud and Fjeld 2001). The lakes were selected based on the following criteria: coverage throughout the country, but with more lakes in areas where we expected greater influence of atmospheric mercury deposition; a wide range in lake size and water

quality; and no significant local pollution. Sub-surface sediment samples (0.5–1 cm layer) from 210 of the Norwegian lakes were also analyzed.

The Finnish sediment data include results from 34 lakes sampled during summer 2002 and 2003 as a part of the NMR Sediment mercury Mapping Project, 16 lakes sampled in the HAPRO-project in 1986 (Verta et al. 1990), 5 lakes sampled in an AMAP-project in 1993 (SYKE database) and four revisited HAPRO lakes in 1999 (Mannio 2001). The NMR mercury Mapping lakes were selected from SYKE mercury Fish Monitoring lakes, but a few lakes were chosen to achieve a better spatial distribution. The HAPRO lakes were originally chosen from studies of acid sensitive lakes with known chemistry and absence of direct human impact (Verta et al. 1990) and the AMAP lakes are located north of the Arctic Circle in Finnish Lapland.

The Swedish lake sediments were sampled in the period autumn 1998 to spring 2002, and the lakes are all a part of a national environmental monitoring program for reference lakes. The lakes have not been limed, and there are no point sources of metals in the catchments (Johansson et al. 2001).

2.2.2 Description of lakes

The lakes and the catchments covered a wide range of conditions regarding bedrock geology, thickness of overburden, amount of precipitation and types of vegetation. The lakes were located at altitudes of 6 to 1243 m, and they were generally small to medium sized, typically 10 to 30 m deep, with a surface area of 0.5 to 1 km² and catchment areas of 4 to 15 km² (Table 2). In Norway, 11 large lakes deeper than 100 m were included, whereas no lakes deeper than 28 m were sampled in Sweden and Finland.

Table 2. Summary statistics of limnological characteristics of the study lakes (n = 389).

country	lakes	max. depth, m			lake area, km²			catchment area, km²		
	n	median	min.	max.	median	min.	max.	median	min.	max.
Finland	59	9.0	1.5	28	0.74	0.03	18,9	11,5	0,2	2877
Norway	231	29	5	270	0.98	0.02	137	14.4	0.12	4518
Sweden	99	13	2.4	28	0.51	0.033	41.2	4.76	0.3	275

2.2.3 Sampling procedures

The Norwegian sediment samples were collected with a gravity corer (75 cm long acrylic tube, 65 mm inner diameter, 2 mm wall thickness). The cores were extruded and sectioned as soon as possible after their retrieval to minimize disturbance of the flocculent surface sediment. The Swedish sediment samples were collected with a gravity corer fitted with plexiglas tubes and a slicing unit for immediate vertical sub-sampling (Johansson et al. 2001). The Finnish sediment samples were collected with a piston-less light-weight gravity corer (inner diameter 95 mm, slightly modified from Axelsson and Håkanson 1978) or a Limnos corer (inner diameter 94 mm, Kansanen et al. 1991). In the Finnish NMR Mapping Campaign, sediment cores with a length of 18-52 cm were evaluated.

In all national surveys the samples were taken from the deepest part of the lake. The sediment cores were usually sectioned in the field in following way: Norwegian samples: 0-0.5 cm surface section, 0.5 – 1 cm subsurface section and a 1 cm section from the deepest part of the core (mean length ± SD: 35.5 ± 9.2 cm). Finnish samples: 0-1 cm surface section and 1 cm reference section (33.7 ± 7.7 cm). Swedish samples: 0-2 cm surface section and a reference section from 30-32 cm. The section

from the deepest part of the core is called reference sediment and has an age of about 150 to 400 years, depending on the local sedimentation rate. Annual sediment accumulation rates of 0.5-2 mm are typically found in similar cores from similar lakes in Fennoscandia (El-Daoushy 1986, Appleby 2000, Bindler et al. 2001, Kaste et al. 2001, Munthe et al. 2004). The reference sediments are probably/typically oldest in the mountain areas and youngest in the lowland plains.

2.2.4 Analytical methods

The Finnish sediment samples were freeze-dried before analyses. The 1966-1999 samples were digested in hot (HNO_3 with concentrated H_2SO_4) and analyzed by cold vapour atomic absorption spectrophotometry (CVAAS). In 2002-2003 a different method was used, based on on-line analysis of combustion products. Sub-samples of known amount were subjected to dry combustion with oxygen, where the decomposition products were carried through a catalyst to a gold trap for selective trapping of mercury by amalgamation. After release by a short heat-up, mercury was quantified using CVAAS.

The Swedish samples were freeze dried before analyses. Sub-samples of known amount were digested in concentrated HNO_3 under pressure at 120°C and then diluted with distilled water to a final volume of 100 ml. Concentrations of mercury were analyzed using CVAAS.

The Norwegian samples were dried at 60 °C, digested in concentrated HNO_3 under pressure at 120 °C and then diluted with distilled water to a final volume of 100 ml. Concentrations of mercury were analyzed using cold vapour atomic fluorescence spectrophotometry (CVAFS). Standard reference sediment (BRC-320) was submitted randomly among samples.

Sediment samples were analyzed for loss on ignition (LOI, ignition loss at 550 °C) in all national surveys. A more detailed description of sampling procedures and other methods is given in Johansson 2001, Verta et al. 1990 and Rognerud and Fjeld, 1999.

Mercury concentrations were adjusted for different organic content by simply dividing the mercury concentrations by LOI (loss on ignition). This will give a better indication of spatial patterns in mercury deposition because deposited mercury is strongly associated to the organic fraction of sediments (Figure 11).

2.3 Results

2.3.1 Organic content

In general, the sediments can be characterized as rich in organic matter. Typically, the organic fractions (measured as loss on ignition, LOI) were in the range of 20–50 % for both reference and surface sediments (Figure 11). There was no systematic difference in organic content in the different national surveys between countries, although the 9 lakes with the highest organic content in the sediments (LOI > 70%) were all located in southern Finland and Sweden. In general, the LOI values in surface and reference sediments within the lakes were not statistically different (pairwise t-test, $p > 0.05$).

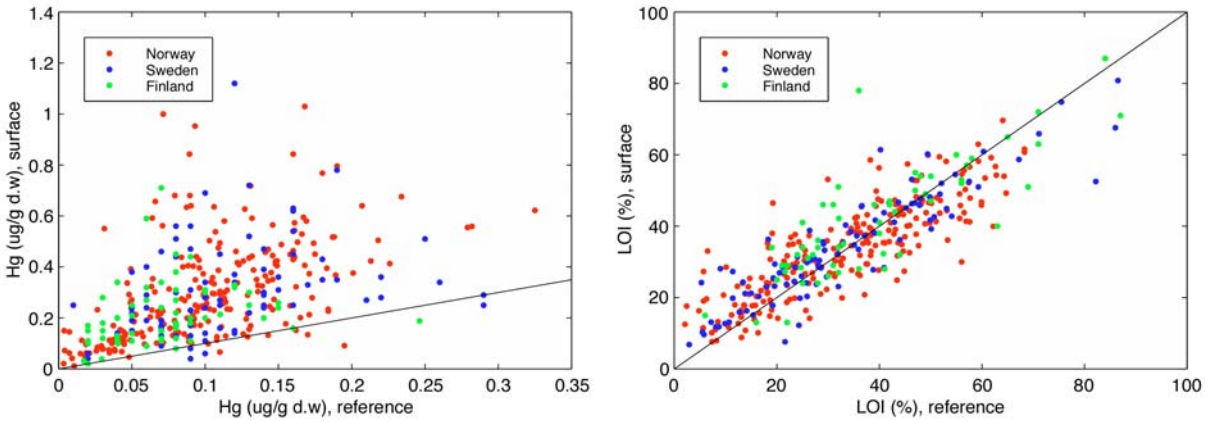

Figure 11. Concentration of mercury in surface versus reference sediments (left panel) and content of organic matter (loss on ignition, LOI) in surface versus reference sediment (right panel). The line shows the 1:1 relationship.

2.3.2 Mercury concentrations

Mercury concentrations were significantly higher in the surface sediments compared to reference sediments (Figure 11). The median ratio between surface and reference sediments (contamination factor) was around 2.8, whereas the 10th and 90th percentiles were 1.3 and 6.1, respectively.

Both surface and reference sediments showed a general increase in mercury concentrations with LOI values increasing from 5 to 45 % (Figure 12). Smoothed curves fitted to the relationship between mercury concentrations and LOI indicate that there was an approximately positive linear relationship between the two variables up to LOI values of 45% (roughly at the 75th percentile for LOI), but the increase in mercury levelled off at LOI values above this. Downward extrapolations of the curves indicate very low mercury concentrations in the inorganic material, consistent with Swedish bedrock survey data (Henriques 1974).

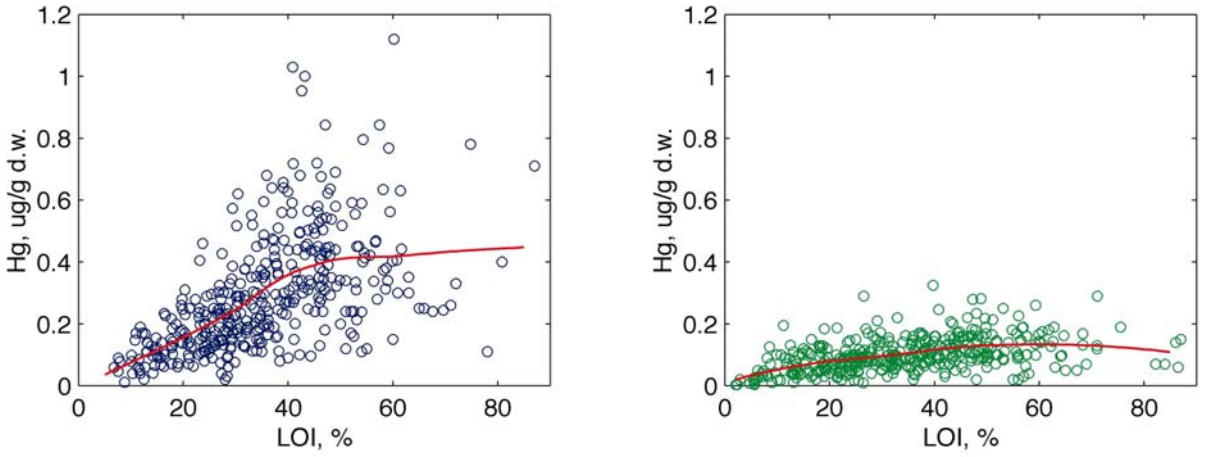

Figure 12 Concentrations of mercury versus LOI (loss on ignition, i.e. organic matter) in surface (left panel) and reference sediments (right panel). The general trends have been described by a locally weighted regression (LOESS, a statistical technique for robust, locally weighted scatter plot smoothing)

2.3.3 Differences between surface and sub-surface sediments

The differences in mercury levels between surface (0–0.5 cm) and sub-surface (0.5–1 cm) sediments in the Norwegian dataset were minor, and a paired t-test showed that the mean difference did not differ significantly from zero (Fig. 13). The differences were commonly in the range of -0.18 to +0.17 μg Hg·(g LOI)$^{-1}$ (10th to 90th percentiles). The map of the differences between the concentrations of these two layers showed a noticeable increase in a confined area in southwest of Norway by 0.05–0.15 μg Hg·(g LOI)$^{-1}$, whereas there was a decrease in the northern and central parts of the country.

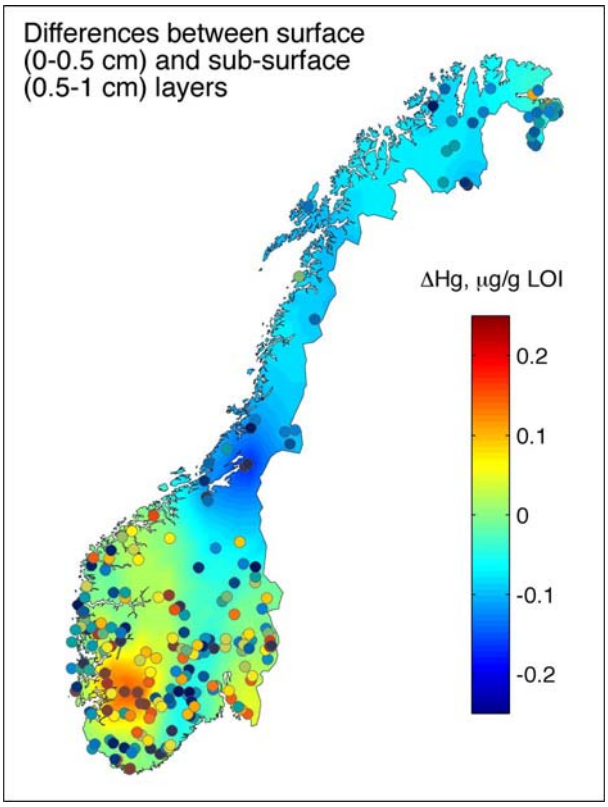

Figure 13 Generalized maps (kriged) showing the differences between mercury concentrations in surface sediments (0–0.5 cm) and sub-surface sediments (0.5–1 cm). Superimposed are the results from the individual lakes. The differences in concentrations (ΔHg) are given per gram organic matter (LOI, loss on ignition).

2.4 Spatial distribution

2.4.1 Organic content

The organic content (indicated by LOI) in lake sediments was highest in south-eastern Finland, and south Sweden. In these regions, LOI values above 50% were frequently observed. In the coastal areas of southern Norway, eastward in the central parts of Sweden and the western part of Finland typical LOI-values were between 35% and 50%. In the mountain areas of Scandinavia and agricultural areas in Sweden and Finland, LOI values between 10% and 35% were frequently observed (Figure 14).

2.4.2 Mercury concentrations

Concentrations of mercury in surface sediments were highest in the coastal areas of southern Norway and southwest Sweden. Elevations were also found in most of central Sweden and the eastern part of northern Sweden, as well as in southern and eastern Finland. In these areas, the concentrations generally were above 0.3 µg Hg g^{-1} dw (dry weight) (Figure 15). There were low concentrations of mercury (<0.15 µg g^{-1} d.w) in mountain areas in southern Norway and central Scandinavia and in the northern part of the Nordic countries. However, the scatter-maps show that concentrations of mercury differed significantly between lakes located close to each other. This indicates that local lake/catchment characteristics can modify the influence of atmospheric deposited mercury on sediment concentrations. Concentrations of mercury in reference sediments are significantly lower than the surface concentrations, but the spatial pattern showed some similarity (Figure 16).

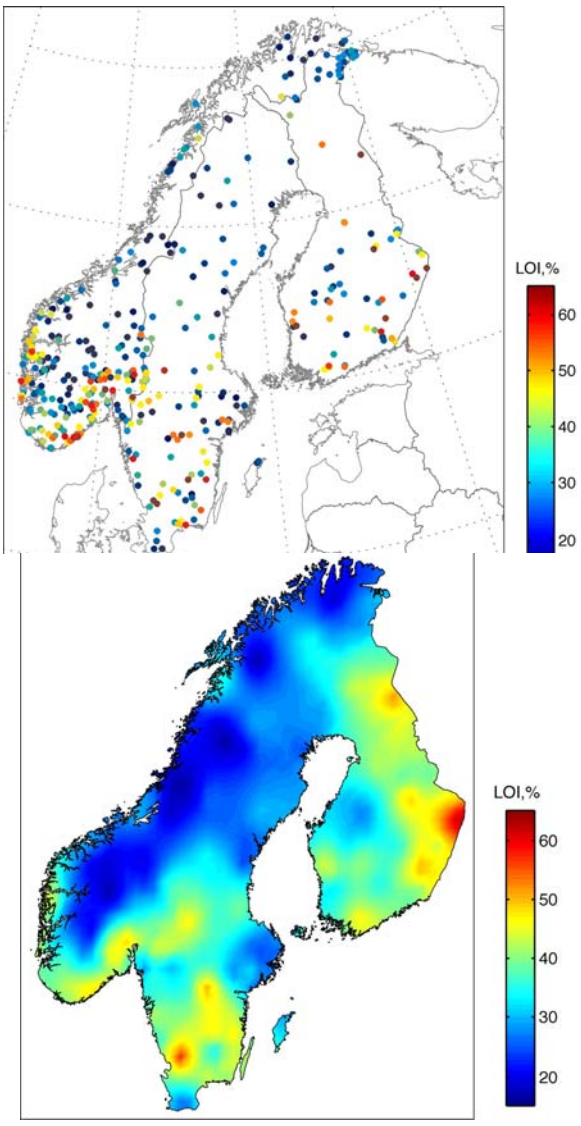

Figure 14 Organic content in surface lake sediments measured as loss on ignition (LOI, % dw). Data are presented as a scatter-map and a kriged map (spatially interpolated).

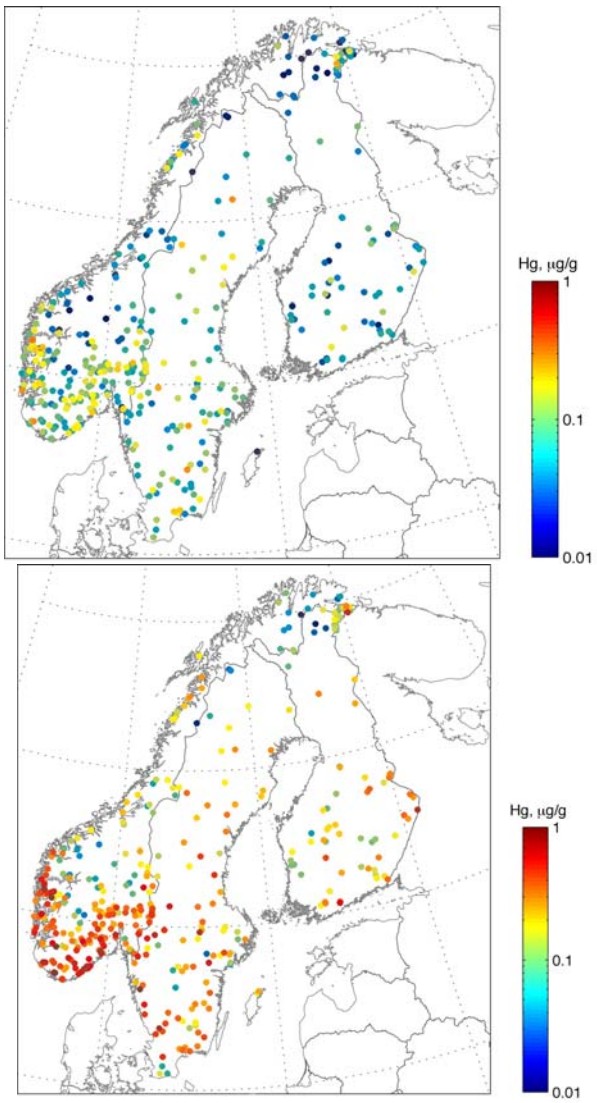

Figure 15 Mercury concentrations (mg kg^{-1} dw) in reference sediments (left panel) and surface lake sediments (right panel) presented as scatter-maps.

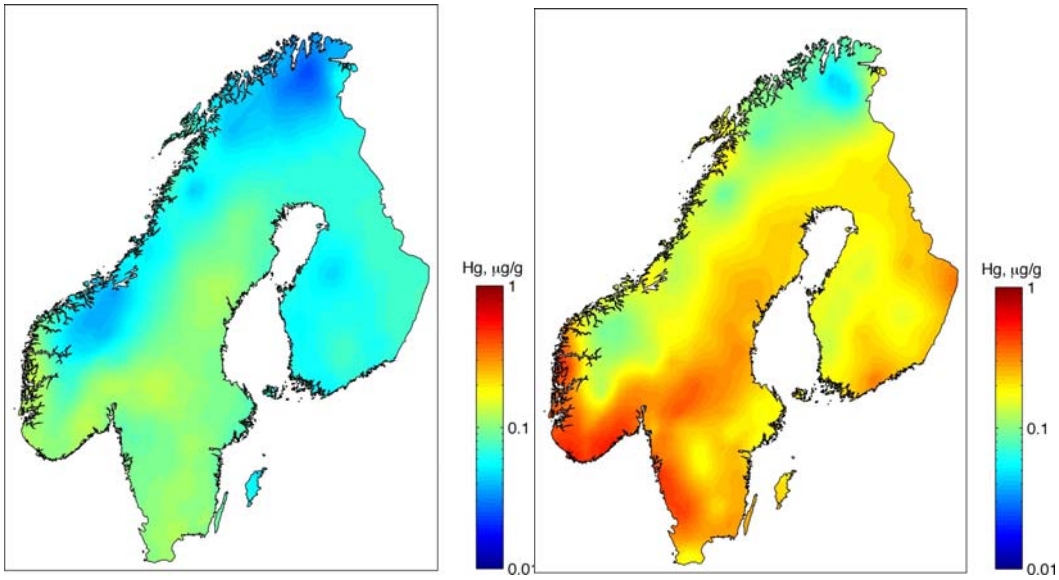

Figure 16 Mercury concentrations (mg kg^{-1} dw) in reference lake sediments (left panel) and surface sediments (right panel) presented as kriged maps (spatially interpolated).

Mercury was associated to the organic sediment fraction (LOI) in surface sediments and to a lower extent also in reference sediments (Figure 12). Since the mercury concentration in the inorganic sediment fraction seems to be negligible compared to the concentration in the organic sediment fraction (Figure 12), additional maps were produced for LOI-adjusted mercury concentrations (Figure 17 and Figure 18). Interestingly, the spatial distributions of LOI-based mercury concentrations in both surface and reference sediments were slightly different from those presented in the maps of the unadjusted concentrations (Figure 15 and Figure 16).

In surface sediments, the LOI-adjusted mercury concentrations were higher in central Scandinavia and lower in Finland relative to the unadjusted (dry-weight based) mercury concentrations. Elevated concentrations in east-central Sweden and the eastern part of northern Sweden are evident in both reference and surface sediments, both with and without LOI-adjustment. Areas with elevated adjusted values in the surface sediments were more extended towards the central parts of Norway. The lowest LOI-adjusted surface values were observed in mountain areas of southern Norway and northern Fennoscandia.

The adjusted values in reference sediments show a slightly different regional distribution than the unadjusted map. The LOI-adjustment revealed elevated levels along the west coast of southern Norway and in central areas of Scandinavia. Generally, in the boreal forested areas the reference concentrations of mercury were slightly lower in Finland than in Sweden and Norway.

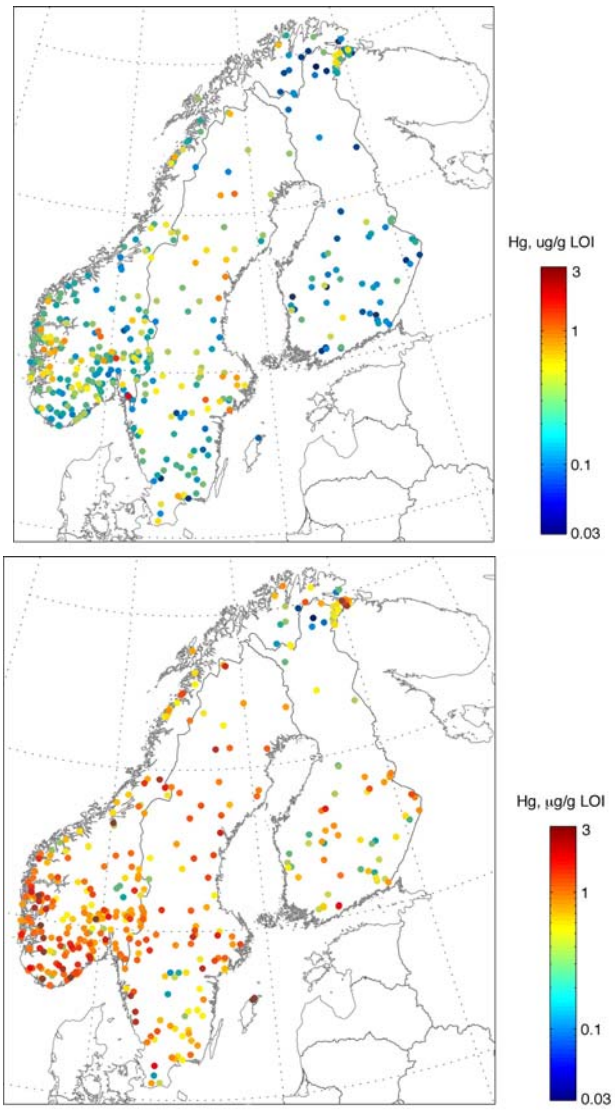

Figure 17. Scatter-maps showing concentrations of mercury normalized with respect to organic matter content (LOI: loss on ignition) in reference sediment (left panel) and surface sediments (right panel).

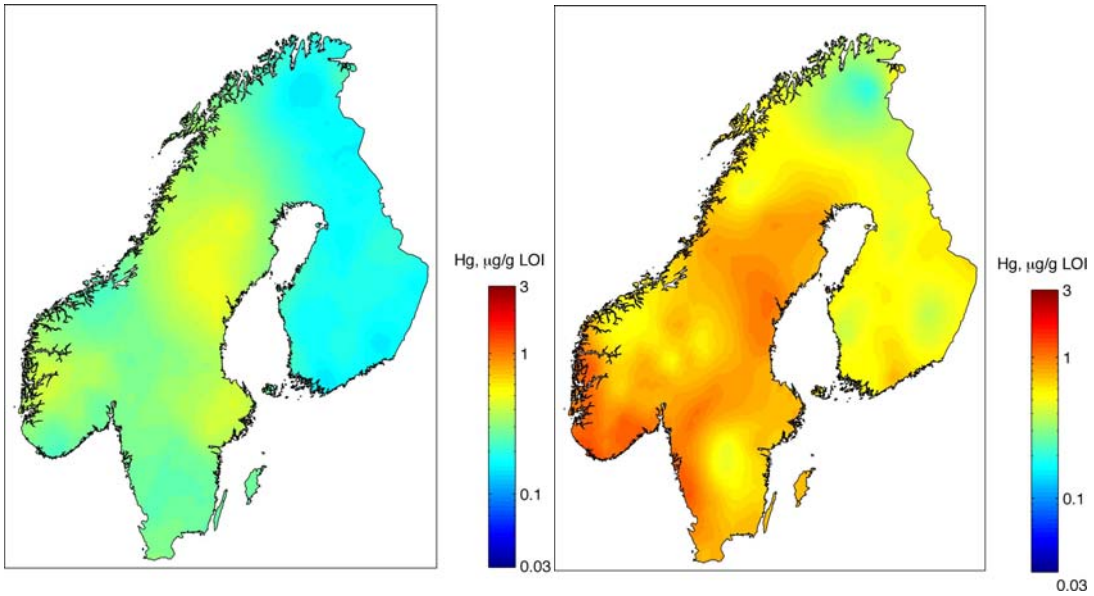

Figure 18. Generalized maps (kriged) showing concentrations of mercury normalized with respect to organic matter content (LOI: loss on ignition) in reference sediment (left panel) and surface sediments (right panel).

2.5 Discussion

Our compilation shows significant increases in the concentrations of mercury in recently deposited lake sediments compared to concentrations in pre-industrial sediments, particularly in the southern forested regions of Fennoscandia and in the eastern part of central and northern Sweden. When compared to concentrations deposited in pre-industrial times (reference sediments), the concentrations in surface sediments, likely to be deposited during the last 10–20 years, were elevated by a factor of typically 5 in southern Fennoscandia and less than 2 in northern areas. This general pattern is attributed mainly to atmospheric deposition of long range transported mercury, mainly emitted in other parts of Europe, but also from other parts or the world. This pattern may also have been influenced by local mercury emissions in Fennoscandia, which were significant especially during the period of 1935–1965 (data from: Lindqvist et al. 1991, Verta 1990, Rognerud and Fjeld 2001). As there are no major known non-atmospheric sources of mercury pollution in the lake catchments, the temporal increase in mercury concentrations in the organic fraction from the "old" reference sediments to "recently" deposited surface sediments is most likely caused by an increased deposition of anthropogenic mercury in the atmosphere. This is consistent with results from similar studies in Canada and North America (summarized by Jackson 1997, Fitzgerald et al. 1998, and Landers et al. 1998). Some of the lakes in eastern Finland have catchments with black shale formations in their bedrock. Black shale is rich in organic carbon and sulphur and occasionally bears elevated mercury concentrations (Loukola-Ruskeeniemi 1990). In a detailed study, no increase of mercury in organic lake sediments (both at the surface and at a reference depth of about 23 cm) was found in these lakes compared to others in the same region but with granite or quartzite in their bedrock (Loukola-Ruskeeniemi et al. 2003).

2.5.1 Sediment characteristics and mercury content

The sediments consisted of a mixture of inorganic material derived from till in the catchments and organic matter produced in the lake or supplied from the catchments. The samples were taken from deep bottom areas where sediment deposition has been relatively undisturbed and therefore in general have a high organic content (Håkanson and Jansson 1983). The highest organic fraction (LOI) in surface sediments was observed in the forested areas of southern Fennoscandia. The nearly identical LOI values of surface and reference sediments indicate stable conditions as well as a very low decomposition rate. The latter is consistent with the finding that most of the organic matter in the water of forest and sub-alpine lakes originates from terrestrial humic substances that are already highly decomposed or refractory (e.g. Meili 1992), and this applies even more to the organic matter incorporated into lake sediments.

The positive relationship between mercury and LOI in surface and reference sediments indicates that mercury is strongly associated with organic matter. Organic complexing agents are known to play an important role in the transport of mercury to lake waters and sediments. Since the terrestrial input of organic matter is high in most boreal surface waters and dominates the input even to clear mountain lakes, most of the total mercury load to Nordic lakes originates from Hg-organic complexes brought into the lakes by soil run-off (Meili 1991b). This is consistent with other studies emphasizing the importance of organic matter as a carrier of mercury from the catchments to lakes and further to the sediments (Meili 1991a, Johansson 1985, Fjeld et al. 1994, Grigal 2001, Rekolainen et al. 1986). The comparatively low mercury levels in the most organic sediments in Finland can be due to substantial input of very old organic carbon (supplied to lakes) from extended peatlands in the catchments contributing to both a natural dilution and a less pronounced contamination, an influence that may have been enhanced by recent dredging.

The relationship between mercury concentrations and LOI indicates that geogenic mercury concentrations (found in the inorganic sediment fraction) are generally very low in the Nordic countries. The inorganic fraction of the lake and riverbank sediments consists of fine-grained particles derived from till, a material which commonly has moved rather short distances (some hundred meters) away from the parent bedrock (Ottesen et al. 1989). In Fennoscandia, Precambrian gneiss, granites and sandstones rich in quartz and feldspars are the dominating types of bedrock (Sigmond 2002). Generally, such rock types have very low concentration of mercury, typically below 0.01 $\mu g\ g^{-1}$ dw (Henriques 1974). This lends further support to the conclusion that mercury in most Nordic lake sediments is almost entirely associated with the organic fraction.

2.6 Temporal trends

Mercury concentrations in dated cores from Fennoscandian lakes consistently show a gradual increase in contamination from the late 1800's to the 1960's, followed by a short period of small changes and a recent decrease in the 1990's (e.g. El-Daoushy and Johansson 1983, Bindler et al. 2001, Kaste et al. 2001, unpubl. new Swedish data). Of particular interest at present is the most recent decline as a potential response to pollution abatement, which here was tested by comparing surface and subsurface sediments in a large Norwegian data set. The difference in mercury concentration between these two layers was in general minor, indicating that a rather stable level of mercury contamination in the lakes during the period 1975-1995. An increase in concentration was found in an area where the annual precipitation had increased by about 10% during the 1990's (Førland et al. 2000). However, several cores from southern Sweden (Munthe et al. 1995a, Bindler et al. 2001, unpubl. new data), Norway (Fjeld and Rognerud 2001, Kaste et al. 2001) and Finland

(Mannio 2001), collected in the late 1990's and early 2000's, show in general a decreasing trend in mercury concentration and mercury flux to sediments during the 1990's. It may be noted here that even though sediment cores from accumulation sediments can be used to reconstruct trends in atmospheric deposition of mercury, their use for quantifying absolute atmospheric deposition rates or even the magnitude of relative changes is not straightforward. Some reasons are a spatially variable influence of sediment focusing of mercury associated to fine grained particles in the deep waters (Renberg 1986) and a variable ratio between mercury supplied from the catchments and mercury deposited on the lake surface (Meili 1991b, 1995, Meili et al. 2003). Further, temporal variations in mercury flux to the sediments do not reflect the atmospheric deposition trends if variations in the flux of organic matter (the main carrier particles for mercury) to the sediments are induced by natural or anthropogenic variations in factors such as storm-induced sediment resuspension, hydraulic conditions and export of soil organic matter from catchments, which all will modify the mercury fluxes to lake sediments, also at constant atmospheric deposition. Thus, mercury concentrations in organic matter (the LOI-adjusted concentrations) may be a more robust indicator of changes in mercury contamination for drainage lakes than either dry-weight based concentrations or net accumulation fluxes in sediments.

2.7 Spatial trends

2.7.1 General

The scatter-maps of mercury in sediments show highly variable concentrations, even in lakes located close to each other. A statistical model based on 132 lakes from southern Norway showed that surface sediment concentrations of mercury were positively correlated to concentration in terrestrial mosses (surrogate variable for atmospheric deposition), organic content in sediments, lake depth and negatively to pH of lake water (Fjeld et al. 1994). Similar results with respect to the empirical relationships between mercury and organic content in sediments and water pH have also been found earlier in Sweden (Håkanson et al. 1988) and Finland (Rekolainen et al. 1986, Verta et al. 1990). The statistically strong influence of organic content and lake depth on sediment mercury concentrations found by Fjeld et al. (1994) was explained by a strong complexing of mercury to organic ligands, by more fine-grained organic material enriched with mercury to accumulate in deeper lakes (with also longer water residence time), and by mercury apparently more strongly sorbing to humic matter in acidic lakes. Thus, the scatter-maps of mercury concentrations in sediments show a spatial distribution where atmospheric deposition has been modified by properties connected to carbon cycle, water quality and morphometry of the different lakes.

The kriged maps show a generalized picture emphasizing large-scale trends in the data set and downplaying local variations. It is important to bear in mind that the organic content in sediments is lower in high altitude lakes than in the boreal lowlands (Rognerud and Fjeld 1990) and that acidic lakes are more frequent in southern parts of Fennoscandia (Skjelkvåle et al. 2001), and this will influence the spatial pattern of mercury concentrations. After adjusting for differences in organic matter content in sediments, the concentrations show a slightly different spatial distribution, where some longitudinal gradients that can be attributed to regional differences in watershed characteristic appear less pronounced while latitudinal gradients persist, in accordance with expected deposition patterns (Ryaboshapko et al. 1999, Ilyin et al. 2006). Further, also temporal trends within a lake may be better reflected by adjusted concentrations, which are likely more independent of changes in sediment characteristics induced by climatic or land-use changes (see also above). We therefore assume that the adjusted concentrations give the best indication of relative differences in spatial

distribution of mercury deposition in pre-industrial times (reference sediments) and in modern times (surface sediments).

2.7.2 Mercury in reference sediments

Based on an average annual net sediment accumulation rate of 1 mm, the reference sediments can be estimated to be deposited about 150-500 years before present. At that time, deposited mercury was most likely dominated by natural sources emitting mercury to the atmosphere (Lindqvist et al. 1991, Jackson 1997, Fitzgerald et al. 1998). The coastal areas in southern Norway showing elevated concentrations of mercury in reference sediments coincide with areas having high annual precipitation. Thus, elevated concentrations of mercury in reference sediments along this coast may be caused by an elevated natural deposition, potentially including also a more efficient washout of mercury from an atmosphere enriched in marine halogens (cf. Chapter 2). For southern Scandinavia, a minor influence of early anthropogenic sources in Europe can not be ruled out, given the prevailing south-westerly winds. However, the adjusted mercury concentrations in reference sediments were highest in central and northern Sweden. This can not be explained by an efficient washout from the atmosphere, because the annual precipitation is lower than in many other areas in Fennoscandia. Possibly, this might be related to a higher background (geogenic or anthropogenic) in these areas of early metallurgy, where smelting and associated activities were extensive at the time the reference sediments probably were deposited (e.g. Ek and Renberg 2001).

In Finland, the mercury concentrations in reference sediments were in general lower than those in Sweden and Norway, both for LOI-adjusted and unadjusted concentrations. This may reflect a smaller natural atmospheric deposition, due to less precipitation and washout of mercury from the atmosphere. The annual precipitation is typically 0.6-0.7 m in Finland and eastern Sweden, but more than 2 m in south-western Norway. Another explanation may be that atmospheric mercury deposition is diluted into a larger store of organic matter in forest soils and peatlands, the latter covering about 30 % of Finland. Peatlands may also contribute to retaining mercury more efficiently than in Norway, with less peatlands and steeper slopes, lower water retention in soils, and more particulate mercury in runoff.

2.7.3 Mercury in surface sediments

The highest concentrations in surface sediments were observed in southern/south-western parts of Fennoscandia, with a pronounced decrease with latitude and longitude. Emission budgets and model simulations of mercury show that most atmospheric deposition originates from long-range transport, especially from other parts of Europe (Lindqvist et al. 1991, Amundsen et al. 1992, Ryaboshapko et al. 1999). The modelled deposition patterns (see e.g. http://www.msceast.org/hms/results_map.html#hg) as well as the measured (Chapter 2) resemble the sediment concentration pattern in Norway and the southern parts of Sweden and Finland. However, in eastern Finland and central Sweden, sediment concentrations are relatively high, whereas the modelled long-range deposition for these areas is rather low. Some of the discrepancies may be explained by remains of mercury from local emissions at historical point sources (Figure 10) still causing elevated mercury concentrations in surface sediments. Fennoscandian surface soils are usually rich in organic matter, and atmospherically deposited mercury will be efficiently sorbed and retained in the catchments (Johansson et al. 1991, Meili 1991a, Meili et al. 2003). Since the lakes in the region typically have a large, and often forested catchment area, most of the mercury load to most of our lakes is supplied with the runoff from forest soils, associated to leached and eroded humic substances (Meili 1991b). Since the turnover of refractory organic matter in soils is very

slow, mercury in the surface soils will reflect the total atmospheric deposition accumulated over many decades or centuries (Johansson et al. 1991, Aastrup et al. 1991). Consequently, the historical emissions from point sources will probably influence mercury concentrations in drainage lake sediments many decades ahead (cf. Meili et al. 2003). On the other hand, Swedish soil surveys (e.g. Lindqvist et al. 1991) suggest that local pollution sources account for little to less than half of the top soil mercury and probably even less in current soil runoff (Meili 1991a, Meili et al. 2003). Accordingly, it is likely that the contribution from historical local point sources to the spatial distribution of elevated mercury concentrations in surface sediments is small and superimposed on a much larger deposition from long-range transport over all Fennoscandia.

2.8 Concluding remarks - sediments

Our compilation shows that in Fennoscandia, surface lake sediments have significantly elevated concentrations of mercury most likely caused by anthropogenic atmospheric deposition over the past century. The highest degree of pollution impact was observed in the coastal areas of southern Norway, in south western Finland, and in Sweden from the coastal areas in the southwest across the central parts to the north-east. Also the preindustrial mercury input is most likely atmospheric, since the geogenic concentrations of mercury in Nordic sediments are typically very low and thus cause minimal interference. This facilitates the detection of small changes in atmospheric fluxes of mercury from sediment archives, even though the response time may be long in many lakes. The contribution from historical local point sources to the spatial distribution of elevated mercury concentrations in surface sediments is small and superimposed on a much larger deposition from long-range transport over all Fennoscandia. There are indications that the rather stable level of mercury contamination in the lakes sediments during the period 1975-1995 has been followed by a decreasing degree of impact during the 1990's. Thus, sediment surveys carried out in the Nordic countries the next few years can uncover the response time of mercury contamination in lakes to the observed decrease in atmospheric deposition of mercury since 1990.

3 Mercury in Nordic freshwater fish

3.1 Introduction

Contamination of freshwater fish by mercury remains an environmental problem of concern in the Nordic countries. Previous studies have demonstrated a link between atmospheric deposition of mercury and levels of methylmercury in Swedish freshwater fish (e.g. Lindqvist *et al.* 1991, Meili et al. 2003). Here we examine a large dataset of mercury in fish from more than 2700 lakes in Sweden, Finland and Norway.

Mercury concentrations in Nordic freshwater fish have been reported regularly since the late 1960's (e.g. Johnels et al. 1967, NHT 1969). Since then, several mapping studies have been conducted in different countries or regions (Björklund et al. 1984, Håkanson et al. 1988, 1990, Verta 1984, Verta et al. 1986, Rognerud and Fjeld 1990, Witick et al. 1995, Rognerud et al. 1996, Johansson et al. 2001, Huuskonen 2001). These studies revealed a widespread occurrence of elevated mercury concentrations in several predatory fish species, also in lakes and rivers without any direct mercury input from local effluents (which are numerous in the Nordic region and mainly from paper industry). In addition to fish mercury mapping, several integrated assessments of mercury pollution in the environment have been conducted (Lindqvist et al. 1984, Vesihallitus 1986, Lindqvist et al. 1991).

Analytical results from fish mercury monitoring have to some extent been registered and compiled into national databases in Sweden and in Finland. However, numerous survey studies and scattered data are found outside any systematic data collections, particularly in Finland and in Norway. This is the first attempt to assemble some of these data and also to assess the mercury concentrations in different fish species all across Fennoscandia.

3.2 Methods

3.2.1 Data compilation

Available data of fish mercury concentrations from Sweden, Finland and Norway from national databases and from case studies were assembled and merged. The initial dataset contained some 46 600 individual or pooled fish data from Sweden (25 000), Finland (17 000) and Norway (4 600) covering 16 different fish species, sampled during the period 1965–2004. The data were then checked by each country experts, with the aim of compiling natural lakes or rivers without known point source of mercury. Data known not to meet these criteria were rejected from further analysis. Accordingly, we excluded lakes and rivers with known point sources of mercury (usually pulp and paper industry or chlor-alkali industry) as well as man-made reservoirs (where mercury levels are known to be elevated also without additional pollution, e.g. Verta et al. 1986, 1990, Porvari and Verta, 2003). Marine areas were not considered.

We also accepted only analyses of fillet (muscle tissue) or whole fish, which is commonly used to assess the human and wildlife exposure to mercury. Furthermore we made a number of checks to validate the data quality, such as examination of basic statistics and individual outliers from weight-length relationships or mercury-size relationships. Obvious errors (such as different units) were corrected if possible, and some observations were deleted. Small perch (< 25 g) were excluded, as they were overrepresented in the Swedish samples due to selectively sampling in some Swedish mercury monitoring programs. We only included total mercury concentrations related to wet weight; data on dry weight basis were converted to wet weight if possible. Also some measurements of methyl mercury (MeHg) were converted to total mercury based on the median MeHg/TotHg percentage of 98% derived from reported parallel analyses of total Hg and MeHg in pike.

Generally, a single site coordinate was given to all fish from a given lake. In some large lakes and rivers, data from several sampling locations were merged to represent a larger area. This was most common for some large Finnish lakes, where hydrologically separated open water regions could be clearly identified.

The oldest mercury data from 1960's and early 1970's were generated by neutron activation analysis (e.g. Steinnes and Johansen 1969), but the majority of the fish samples were analysed with cold vapour atomic absorption spectrometry (CVAAS) following somewhat different digestion/-combustion procedures (see sediment chapter above). Major laboratories used commercial certified reference samples in order to ensure good analytical quality. In addition, several intercalibration exercises with a reference fish muscle material were conducted already during 1960's (Häsänen 1969) and on a regular basis during the 1980's and 1990's. In some cases the intercalibration results based on different methods have been published (e.g. Surma-Aho et al. 1996). No systematic differences between different methods have been reported. Accordingly we did not reject any data solely because of analytical method or collection year.

Finally all lakes with less than 3 individuals (compare next paragraph) per species/taxon were rejected. The final database consisted of 28 938 mercury analyses from six fish species (97.6% individually analyzed) from 2 758 locations (97.4% of the sampled lakes). The first data was from 1965. Generally, most data from 1960's to early 1980's were from contaminated sites in Sweden and Finland and were thus rejected and only some 0.2 % of the accepted data was from before 1980.

3.2.2 Data standardization

For statistical comparisons and regional mapping of fish mercury levels across lakes and rivers with different fish communities, fish mercury data need to be harmonized with respect fish type. mercury concentrations vary widely among organisms within an ecosystem, but follow fairly consistent patterns related to species and size (e.g. Meili 1997). Accordingly, the mercury concentration in an organism is predictable from its typical deviation from the concentration in a standard organism within the same food web, and vice versa. Among commonly available variables, length and/or body weight is the most powerful single predictor of this deviation in the case of fish, also across species. As a standard organism, the 1-kg pike has been widely used since the earliest survey studies (Johnels et al. 1967). This fish has typically similar mercury concentrations as a 0.3-kg perch, which has a length of 30 cm (Meili et al. 2004). Another standard organism proposed to represent salmoniform fish is the 25-cm trout, which has a weight of 0.16 kg. The concentration in this fish is typically similar as in a 20-cm char, which has a weight of 0.07 kg, but four times lower than that in a 1-kg pike (Meili et al. 2004). We took advantage of such typically observed relationships among different species and size classes within lakes to harmonize the fish mercury data across the Fennoscsandia. Since only fish with size data were included in the standardized dataset, the number of fish was reduced correspondingly by about 1%. For standardized maps, data were further reduced, which explains some differences between maps with respect to the number and size of dots. Each site was given a value based on a single species, even if normalized data from several species were available. Priority was given to pike as long as at least 4 individuals were available from a site. If this was not the case, the site-specific value was based on at least ten individuals of another species, in the order of perch > trout > char.

3.3 Results

Compilation and harmonization of raw data from Sweden, Finland and Norway generated a data set containing mercury concentrations in fillets (muscle tissue) from 33 116 individuals (of which 4 883 were pooled to 709 samples for the subsequent data analysis). Data originate from five major taxa (Table 3). These are highly unevenly distributed among countries, which partly is due to the postglacial immigration history and abundance of habitats, but also reflects the fishing/-consumption patterns and monitoring preferences/traditions (see also maps below).

The five major taxa were used for further evaluation and mapping: Northern pike (*Esox lucius*), Eurasian perch (*Perca fluviatilis*), brown trout (*Salmo trutta*), Arctic char (*Salvelinus alpinus*), and whitefish species containing mainly of powan (*Coregonus lavaretus*) and vendace (*Coregonus albula*). For each of these fish taxa, the arithmetic mean concentrations of mercury at all sites were calculated to produce Nordic dot maps (Figure 19 and Figure 20) and statistics (Tables 3-5).

Table 3. Number of selected individuals (of which 15% were analyzed as pooled samples).

Count	FI	NO	SE	sum
Char	42	435	352	829
Coregonus	329	217	17	563
Perch	1571	1547	1664	4782
Pike	3627	542	20351	24520
Trout	62	2360	0	2422
Sum	5631	5101	22384	33116

Table 4. Summary statistics of the fish mercury data from the Nordic countries: Fish characteristics and mercury concentrations (all data from 1965 - 2004).

Fish:	*Lake Counts n*	Fish Counts n	Mean length Cm	Median weight kg	10% weight kg	90% weight kg	Min weight kg	Max weight kg
Pike	*2517*	24520	48.1	1.009	0.525	1.530	0.025	12.64
Perch	*157*	4782	21.8	0.175	0.043	0.387	0.025	2.670
Trout	*136*	2422	28.8	0.386	0.720	0.690	0.008	12.70
Char	*40*	829	26.1	0.223	0.056	0.549	0.009	1.350
Whitefish sp.	*46*	563	26.4	0.213	0.043	0.440	0.006	1.155

Hg:	*Counts n*	*Counts n*	Mean mg/kg ww	Median mg/kg ww	10% mg/kg ww	90% mg/kg ww	Min mg/kg ww	Max mg/kg ww
Pike	*2517*	24520	0.73	0.65	0.260	1.30	0.01	6.02
Perch	*157*	4778	0.40	0.30	0.114	0.760	0.01	4.16
Trout	*136*	2422	0.13	0.08	0.023	0.257	0.01	3.14
Char	*40*	829	0.11	0.075	0.025	0.250	0.01	1.04
Whitefish sp.	*46*	563	0.12	0.10	0.050	0.190	0.01	0.57

Table 5. Summary of fish Hg concentrations collected 1965-2004 in different regions of the Nordic countries, and fraction of lakes with levels exceeding different guidelines in a standard fish (1-kg pike or 0.3-kg perch or 3.2-kg trout or 1.4-kg char).

Country	Region [1)]	Number of lakes	Median HgStd, mg/kg ww	Fraction of lakes with HgStd >1.0 mg/kg ww [2)]	Fraction of lakes with HgStd >0.5 mg/kg ww [3)]	Fraction of lakes with HgStd >0.3 mg/kg ww [4)]
All	all	2758	0.69	20%	61%	81%
Finland	all	285	0.51	10%	50%	80%
Sweden	all	2273	0.71	24%	69%	87%
Norway	all	200	0.34	4%	30%	59%
Finland	N	156	0.31	0.5%	19%	55%
Finland	S	255	0.53	28%	54%	80%
Sweden	N	102	0.31	7%	27%	50%
Sweden	C	592	0.82	34%	80%	94%
Sweden	S	1525	0.67	22%	67%	89%
Norway	N	33	0.23	3%	5%	17%
Norway	C <800m	31	0.31	3%	20%	49%
Norway	S <800m	107	0.45	7%	42%	75%
Norway	C >800m	12	0.15	0%	2%	15%
Norway	S >800m	17	0.14	0%	2%	10%

[1)] S= south, C= central, N= north; altitudes <800 m except where indicated.

N≥65, 65<C≤61, S<61

[2)] EU maximum concentration for marketing of specified predatory fish, e.g. pike.

[3)] EU general maximum concentration for marketing of fish

[4)] USEPA criterion for methyl mercury in fish

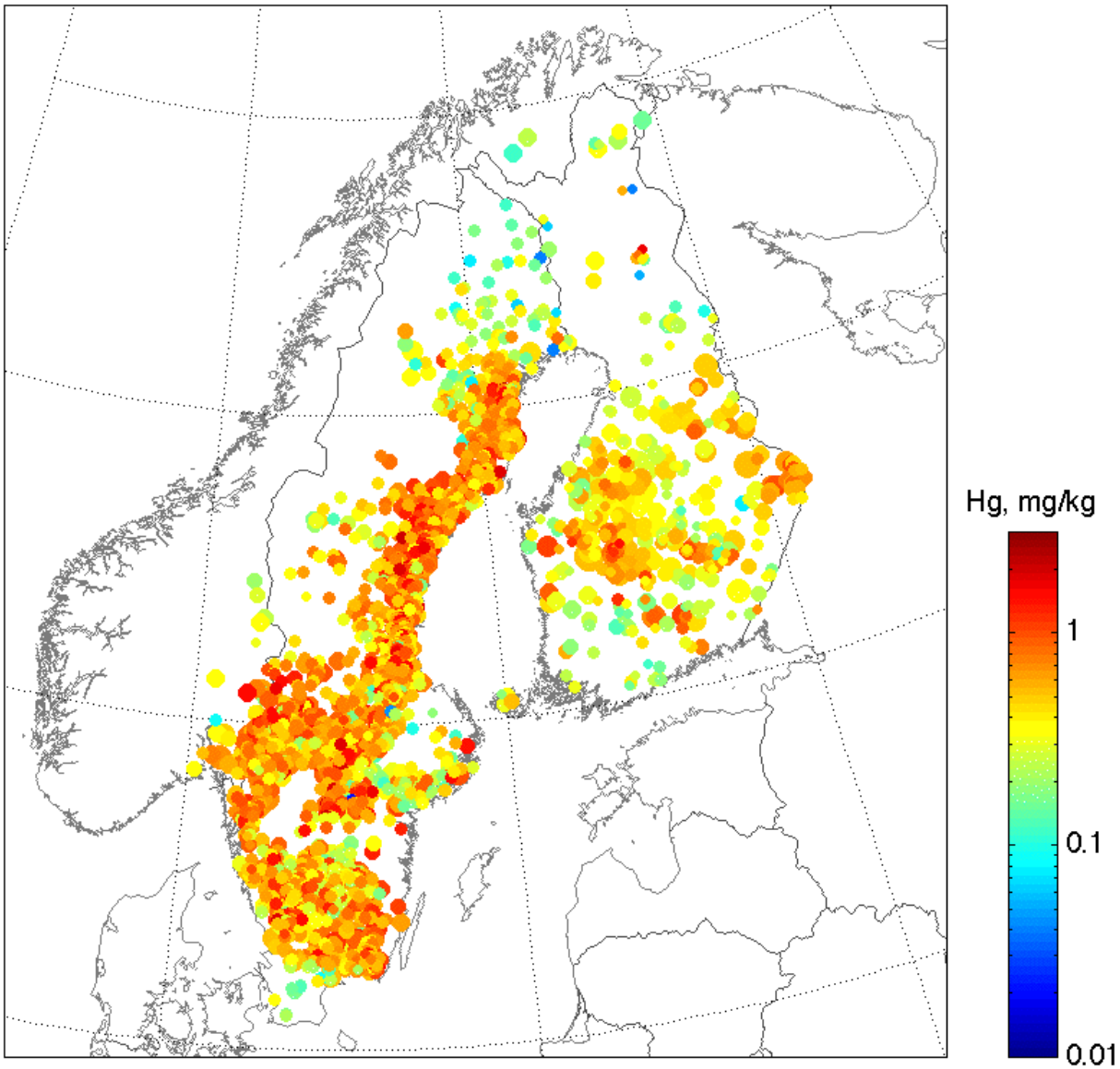

Figure 19. Nordic map showing the site-specific arithmetic means of observed mercury concentrations (mg kg^{-1} ww, without any adjustments) in pike (*Esox lucius*) collected in 1965-2004 in 2517 lakes and rivers. The dot size indicates the number of individuals analysed.

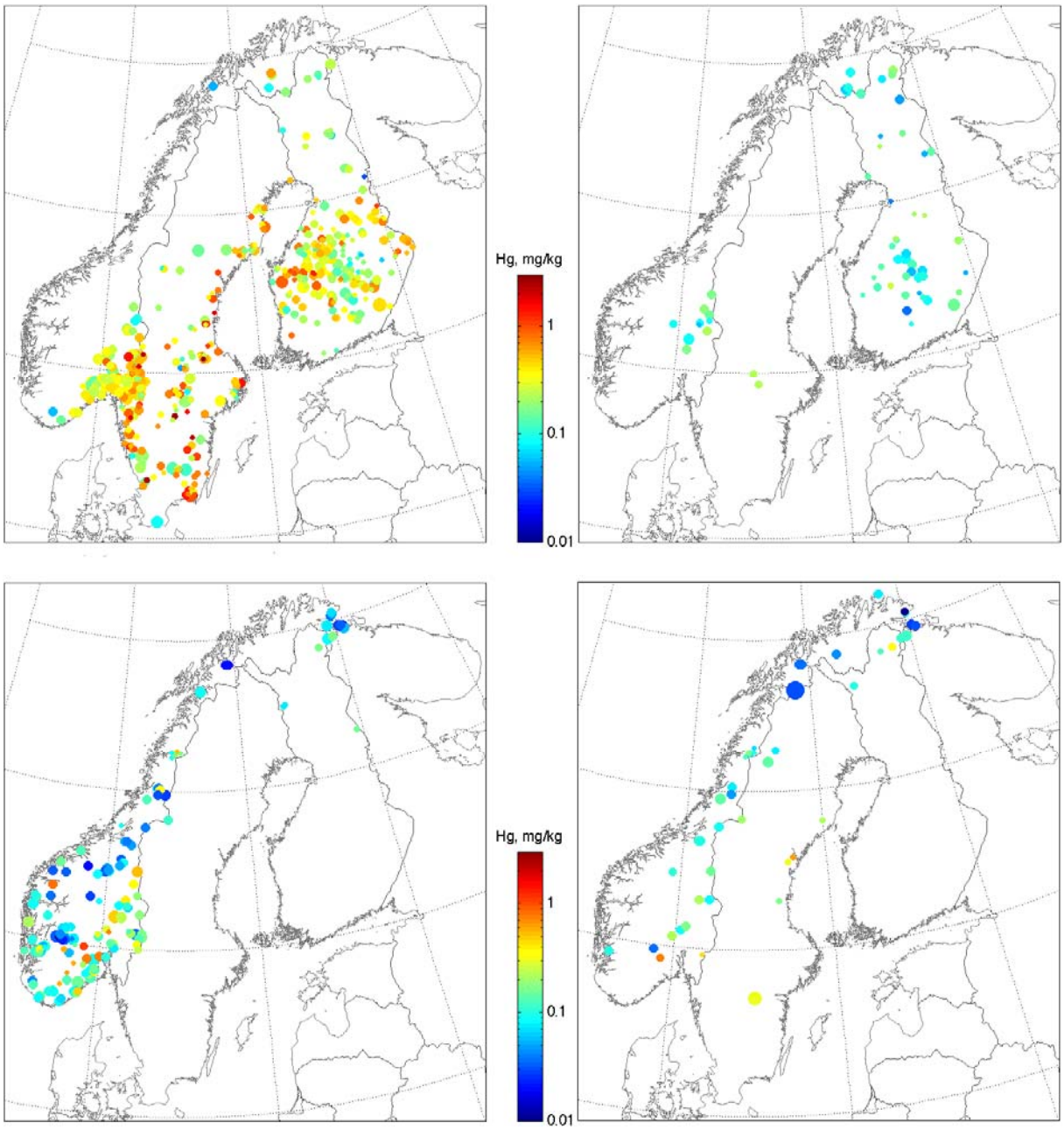

Figure 20. Nordic maps showing the site-specific arithmetic means of observed mercury concentrations (mg kg^{-1} ww, without any adjustments) in four important fish taxa collected in 1985-2002. Top left: perch (*Perca fluviatilis*), top right: whitefish sp. (*Coregonus spp.*), bottom left: trout (*Salmo trutta*), bottom right: char (*Salvelinus alpinus*). The dot size indicates the number of individuals analysed.

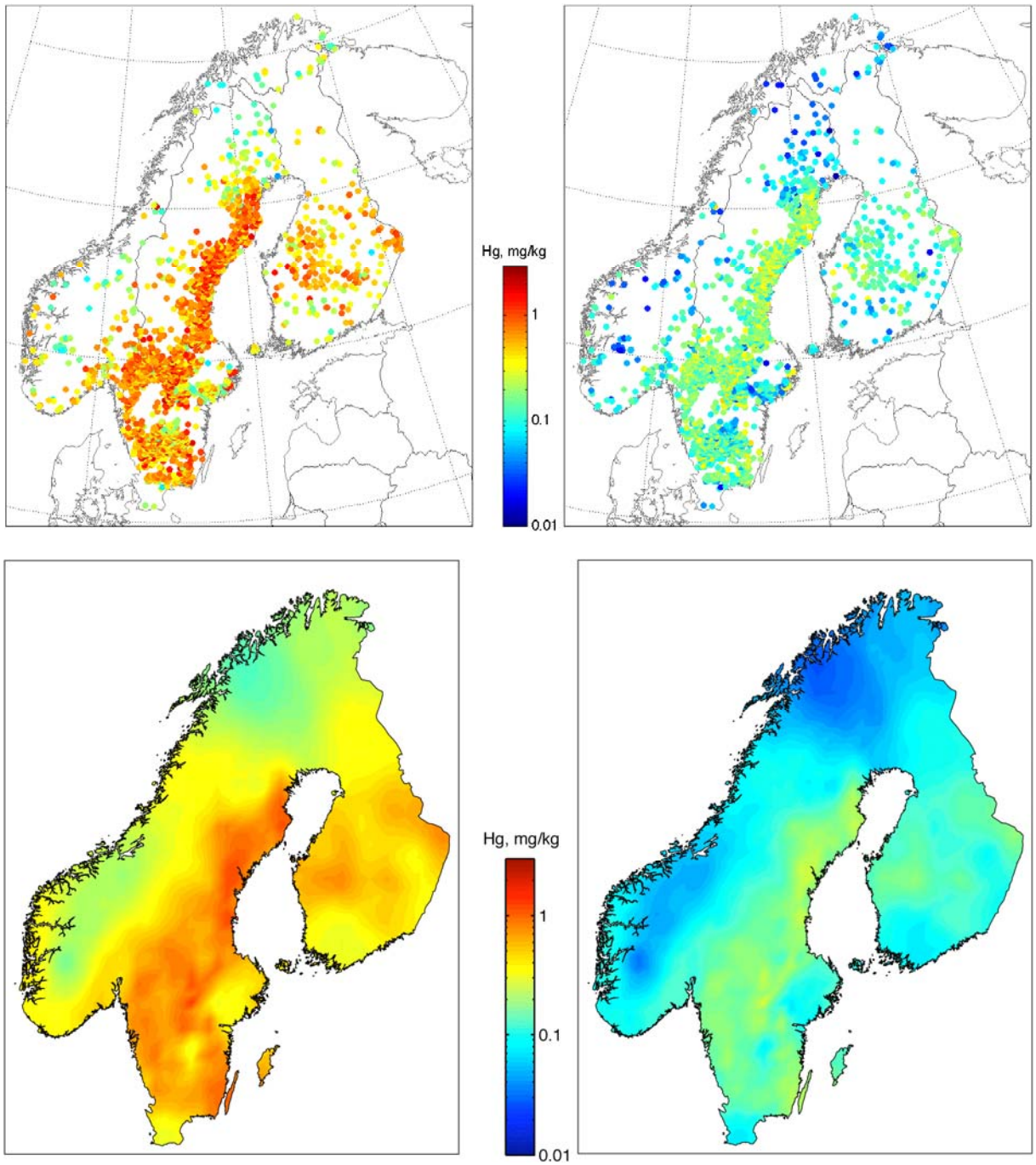

Figure 21. Nordic maps showing mercury concentrations (mg kg^{-1} ww) in Nordic freshwater fish after standardization to a 1-kg pike (left) and to a 25-cm trout (right), shown both as site-specific arithmetic means of selected individuals (upper) and as interpolated maps (lower). The map to the left is representative of a 1-kg pike, a 0.3-kg perch, a 3.2-kg trout, and a 1.4-kg char, whereas the map to the right is representative of a 50-g pike, a 10-g perch, a 160-g trout, and a 70-g char (Meili et al. 2004). The maps to right shows values that are on average four times lower than those in the maps to the left.

3.3.1 Fish mercury concentrations

For the six species, the Hg concentration level was highest in Northern pike (arithmetic mean 0.73 mg/kg), followed by perch (mean 0.40 mg/kg); trout (mean 0.13 mg/kg); Coregonus sp. (mean 0.12 mg/kg); and char (mean 0.11mg kg). The median concentrations were lower by some 0.1 mg/kg for pike and perch and less for other species (Table 4).

For a "standard fish" (1-kg pike, 0.3-kg perch, 3.2-kg trout or 1.4-kg char), the median mercury concentration was 0.69 mg/kg (Table 5). Classified by country, the highest Hg concentration for a standard fish was in Sweden (median 0.71 mg/kg) and clearly lower in Finland (0.51 mg/kg) and even more so in Norway (0.34 mg/kg) being less than half (48 %) of that in Sweden. By latitude, the highest mean concentration was in central Sweden, followed by southern Sweden, southern Norway (low-altitude lakes) and southern Finland. In all countries, the concentration level was lowest in the northern regions (>65°N), with a median ranging from 0.23 mg/kg in Norway to 0.31 mg/kg in Finland and Sweden. The lowest concentrations were, however, observed further south in high-altitude lakes (> 800 m) in Norway (median 0.14-0.15 mg/kg, Table 5). We produced taxon-specific maps showing arithmetic mean values for each site (Figure 19 and Figure 20). For whitefish (*Coregonus lavaretus* etc.) and vendace (*Coregonus albula*), we combined data to produce a *Coregonus* sp. map. These scatter plots (dot maps) show site-specific mean fish mercury concentration regardless of individual weight or length. Also fish with no known weight or length were accepted for these maps to maximize the number of observations for a general overview.

The maps show different spatial distributions for each species, reflecting both natural fish occurrence and sampling efforts. Pike and perch data are most common in the forested regions, whereas salmonid species have been analyzed frequently only in Norway and coregonids in Finland, even though these are relatively abundant also in other regions of Fennoscandia. Thus, the maps largely reflect a pattern of fishing interests, consumption preferences, and monitoring traditions that are influenced among other by the location of suspected point sources of mercury to the atmosphere (particularly in Sweden).

We also produced maps for "standard fish", both by site and as interpolated (kriged), focusing on 1-kg pike and 25-cm trout (Figure 21). As for country-specific data, concentrations are highest in southern and central Sweden, somewhat lower in central and eastern Finland and decline to the north (Figure 19, Figure 20, Figure 21). However, low concentrations are also common in the south, particularly in coastal areas of Sweden and Finland where the geological setting is dominated by marine deposits and where agriculture is common, and in mountain regions.

For all salmoniform taxa (trout, char and whitefish), the mercury levels are generally much lower than in pike and perch. Notably, whitefish species show low levels in Finland also in lakes were pike levels are high (Figure 19 and Figure 20). A similar pattern can be found for trout in SE Norway. For trout and char, high values were found in single lakes where mainly large piscivorous individuals were analyzed. Fish size is an important factor influencing fish mercury levels, regardless of the geographic scale at which data are collected, since larger fish eat larger prey, which in general is more enriched in mercury.

3.4 Discussion

3.4.1 Spatial distribution of normalized mercury concentrations

The main features of the geographical pattern for mercury in fish is decreasing levels from south to north with the high concentrations also found in central Sweden and along the Swedish coast of the Gulf of Bothnia and in central and eastern Finland. The elevated concentrations in Central and Northern Coastal Sweden may be influenced by historical emissions of mercury to both water and air as suggested e.g. by Lindquist et al. (1991). However, specific ecosystem characteristics also contribute to the variability in fish mercury between regions. This is also true for the southernmost part of Sweden and south western Finland where low concentrations were found despite a large influence of atmospheric pollution. These areas are mainly agricultural and the freshwater bodies mainly eutrophic.

In general the areas with highest concentrations in Sweden and in Finland follow the distribution of boreal coniferous forests. Regions with forests and or peatland seem to have higher mercury in fish than regions with predominantly cultivated soils or high-elevation regions. The exception is parts of northern Scandinavia from the Bothnian Bay to the arctic where fish mercury is low even at forested regions.

In central and eastern Finland, relatively high mercury concentrations are observed in lakes with high humus concentrations. Also in Norway higher fish mercury is mainly observed in areas with higher DOC (Henriksen et al. 1998, Skjelkvåle et al. 1996, Skjelkvåle et al. 2001). Our findings establish basic geographic and waterbody-specific trends, and corroborate prior findings relating to controls exerted on mercury by waterbody DOC and pH.

The maps for size- and taxon-standardized mercury concentrations may reflect a combination of current mercury deposition, historical mercury deposition, and ecosystem susceptibility, whereby the latter probably accounts for most of the observed variability. Accordingly, lakes and regions with high levels at present are likely those where the critical (atmospheric) load of mercury is low.

3.4.2 Comparison of sediment and fish mercury patterns

Standardized fish concentrations show a similar pattern to surface sediment concentrations (on dry weight basis) in some regions (central Sweden, Norway, eastern Finland and in the north, 1-kg pike, Figure 21 and Figure 16). In other regions, this pattern is not evident e.g. in the southern areas of Finland and Sweden. Here sediment concentrations are elevated but fish mercury concentrations are relatively low. In contrast to these examples, relatively high fish mercury is observed in western Finland where sediment mercury concentrations are generally low. This illustrates the complexity of the biogeochemical cycling of mercury and highlights the importance to take into account ecosystem characteristics, in addition to mercury loading, when assessing the degree of contamination of fish by mercury.

3.4.3 Comparison with mercury in fish from US and Canada

Our data from Northern Europe show a strong similarity with data from a large survey in NE North America, when considering the mean values for different taxa (Table 6). A comprehensive study from north eastern North America provides data from 15,305 records of fish tissue mercury data from New York State to Newfoundland (Kamman et al. 2005). Detailed analyses were carried out using data for 13 species. Relevant species for comparison with the Nordic fish species are listed in Table 6. The comparison between these two data sets is somewhat difficult because of different mean length/weight of the species in different regions. However it seems probable that the concentrations in Northern pike in Scandinavia are notably higher and even more so if these data was length/weight adjusted. For perch (*Perca fluviatilis vs. Perca flavescens*) the mean concentrations and size are comparable, although somewhat lower in Scandinavia. For the rest of the species (brown trout, whitefish) the mean length/weight (and mean mercury concentration) are clearly lower in Scandinavia compared to N-E America.

US-EPA Fact Sheet (2001) provides mercury data from 90 000 records of freshwater fish collected in 43 states from 1980s to early 2001. Mean concentrations and range were (mg Hg/kg ww): Yellow perch (0.25, 0,005-2.14), Lake trout (0.27, 0.005-2.0), Northern pike (0.36; 0,005-4.4). The EPAs 1987 National Study of Chemical Residues in Fish (NSCRF) also provides data of brown trout from 374 sites. Mean concentration was 0.14 mg Hg/kg ww.

Mercury levels in standard-length fillets varied significantly with waterbody type for 7 of 13 species analyzed. Generally, several species displayed elevated mercury concentrations in reservoirs, relative to lakes and rivers. Brook trout mercury concentrations were lower in rivers than in lakes, whereas no significant differences were observed for northern pike, brown trout and yellow perch.

There is a large variation in mercury concentrations in Arctic char in the northern hemisphere in the AMAP area. Since Arctic char (similar to other potentially piscivorous fish) can occupy different trophic levels, its mercury concentrations may vary considerably even within a given lake, which makes site comparisons quite difficult (AMAP 2002, 2005). However, much of this variation can be accounted for by taking advantage of known patterns within lakes.

Table 6. Mean values of data from large fish surveys in North America and the Nordic countries.

Species (common name and *latin*)		Counts n	Length cm	Weight* kg	Hg mg/kg
N.E. North-America:					
Northern pike	*Esox lucius*	1 065	62	1.3	0.64
Yellow perch	*Perca flavescens*	2 888	20	0.09	0.44
Brown trout	*Salmo trutta*	412	42	0.8	0.30
Brook trout	*Salvelinus fontinalis*	1 104	27	0.2	0.18
Lake trout	*Salvelinus namaycush*	1 076	57	2.0	0.60
Lake whitefish	*Coregonus clupeaformis*	234	39	0.5	0.21
N. Europe:					
Northern pike	*Esox lucius*	24 520	48	1.0	0.73
Eurasian perch	*Perca fluviatilis*	4 782	22	0.18	0.40
Brown trout	*Salmo trutta*	2 422	29	0.39	0.13
Arctic char	*Salvelinus alpinus*	829	26	0.22	0.11
Whitefish sp.	*Coregonus spp.*	563	26	0.21	0.12

* Estimated from reported fish length and length/weight relationship for species.

3.4.4 Comparison with mercury in fish with different guidelines

In Table 5 the exceedance of the fish mercury concentrations compared with the different guideline values are presented for different regions. The table indicates that for the whole Scandinavia the exceedance of the guideline for a predatory fish (1.0 mg/kg, pike) is 20 percent with the highest frequency in South-Central Sweden and Norway and South Finland. In the case of large perch (300 g) the EU guideline of 0.5 mg/kg would be exceeded in 61 % of the lakes (table 5), while the mean and median concentrations for the "normal" weight perch of less than 200 g is well below the EU guideline (mean 0.40 mg/kg, median 0.30 mg/kg, table 4). The USEPA criterion of 0.3 mg/kg would be exceeded in 81 % of the lakes in the case of standard fish.

In North-East America the exceedance of a standard perch (length 20 cm) of the USEPA criterion was 42 percent (Kamman et al. 2005). Given that the size of the US standard perch is almost equal with the mean of our data for perch (22 cm) it seems probable that the variation of mercury concentration and the exceedance of the guideline levels are almost identical with these two species inhabiting similar ecological niche at different continents, but at similar climatological and geological (precambric shield) regions with almost identical mercury load from the atmosphere.

3.5 Concluding remarks - fish

This new compilation of fish mercury data from Sweden, Norway and Finland shows that large geographical areas are affected by mercury contamination, which is enhanced in regions of high susceptibility. The historical background of fish mercury in the Scandinavia is highly uncertain, but a general level of less than 0.2 mg/kg for the Swedish lakes was hypothesized by Lindqvist et al. 1991. Verta (1990) estimated, based on sediment mercury/organic matter relation, that pike mercury concentrations in the most humic Finnish lakes may have reached the level of 0.4-0.5 mg/kg at natural prehistoric conditions.

The general pattern of fish contamination follows a similar pattern to current and previous atmospheric deposition and pollution discussed in sections 2 and 3. Large areas have fish with Hg concentrations exceeding the WHO-based EU health advisory guideline of 0.5 mg/kg or 1.0 mg/kg

(northern pike), thus restricting their use for human consumption. Furthermore the species specific median concentrations are identical with concentrations in the North America at regions with similar level of atmospheric mercury pollution and geographic characteristics of bedrock and soils.

4 Overall conclusions

An evaluation of geographical variations in mercury concentrations in precipitation indicates that the influence from anthropogenic sources in Central European areas is still significant. The interannual variability of deposition is large and dependent of precipitation amounts. An evaluation of data from stations around the North Sea has indicated a significant decrease in mercury concentrations in precipitation indicating a continuous decrease of emissions in Europe (Wängberg et al., 2007). For mercury in air (TGM), the geographical pattern is less pronounced indicating the influence of mercury emissions and distribution over a larger geographical area (i.e. hemispherical transport).

Comparison of surface and historical lake sediments from 1600's-1800's show significantly elevated concentrations of mercury most likely caused by anthropogenic atmospheric deposition over the past century. The highest degree of pollution impact was observed in the coastal areas of southern Norway, in south western Finland, and in Sweden from the coastal areas in the southwest across the central parts to the north-east. The general increase in recent versus old sediments was 2-5 fold.

The general pattern of fish contamination follows to some extent a pattern to current and previous pollution. Large areas have fish with Hg concentrations exceeding the WHO-based EU health advisory guideline of 0.5 mg/kg or 1.0 mg/kg (northern pike), thus restricting their use for human consumption.

A more comprehensive assessment of factors influencing levels of methylmercury in fish has to include a number of parameters not included in this evaluation such as catchment characteristics (*e.g.* relative size, presence of wetlands), contents and fluxes of DOC in soil run-off and surface waters as well as methylation potential within ecosystems.

This report provides a first comprehensive compilation and assessment of available data on mercury in air, precipitation, sediments and fish in the Nordic countries. One of the main conclusions is that mercury levels in Nordic ecosystems continue to be affected by long-range atmospheric transport. The geographical patterns of mercury concentrations in both sediments and fish are also affected by ecosystem characteristics and possibly also by historical emissions. The evaluation of mercury in precipitation clearly shows that long-range transport from European sources continues to deliver mercury to Nordic ecosystems. The data on air concentrations, however, suggest that sources on the hemispheric scale are also contributing to the long-range transport.

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