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Atmospheric mercury in Norway: Contributions from different sources

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Abstract

The environmental loadings of national Norwegian mercury emissions compared to the loadings of atmospheric long range transported mercury have been estimated using national emission data and EMEP model data. The results indicate that atmospheric long-range transport to Norway is somewhat larger than the national Norwegian emissions of mercury. Atmospheric deposition of mercury has been studied using data from Norwegian monitoring programs on mercury in precipitation, mosses, natural surface soils, and lake sediments. Precipitation data show no significant time trend during 1990–2002, whereas moss samples show similar concentrations from 1985 to 1995, but a 30% decrease from 1995 to 2000. Concentrations of mercury in peat cores and reference sediments indicate that the current mercury levels measured in surface sediments, surface soils and mosses at background sites in Norway are substantially affected by long-range atmospheric transport.
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1. Introduction

Mercury is a toxic environmental pollutant that is among the metals most highly bioconcentrated in the human food chain. Once emitted, mercury may be deposited to environmental surfaces by dry and wet processes. In aquatic systems mercury is methylated by microorganisms and bioaccumulated through food chains, which may lead to human exposure.

Atmospheric levels of mercury are affected by natural as well as anthropogenic sources. The most important anthropogenic source of mercury is the combustion of fossil fuels, in particular coal. Additional anthropogenic sources of key significance are chlor-alkali production, cement production and waste incineration. European emissions of mercury from anthropogenic sources to the atmosphere have decreased from about 630 t in 1990 through 340 t in 1995 to about 200 t in 2000 (EMEP MSC-W, 2002). The global anthropogenic emissions have changed from about 3600 t year^{-1} during the 1980s to about 2000 t year^{-1} in the second half of the 1990s (Pacyna and Pacyna, 2002). It is estimated that between 200 and 250 t of Hg is emitted annually from natural emission sources in Europe (Pacyna et al., 2001).

Long-term monitoring programs are presently recognized as powerful tools for local as well as regional and global studies of atmospheric long-range transport processes (Kallenborn and Berg, in press). The pro-

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grams are designed to control and assess international regulatory measures, determine "natural" background levels, collect information on environmental behaviour (spatial and temporal trends), as well as to serve as "early warning tool". Even minimal changes in patterns and distributions may be a first signal of potentially severe environmental changes.

In the following work data from the EMEP program and various Norwegian monitoring programs have been used to estimate the environmental loadings of national Norwegian mercury emissions, study time trends in the mercury deposition, and assess background levels of mercury in Norway.

2. Material and methods

2.1. Emissions

Norwegian companies are divided in four categories according to pollutant. Those in three of these categories have to report to the environmental authorities their emissions to air and water each year. The official Norwegian emission data are based on the numbers reported from the companies, in addition to emission estimates made by Statistics Norway. The emission data for Norway as well as for other countries are relatively uncertain due to uncertainties in the procedures used to arrive at these data.

2.2. EMEP models

Heavy metals are transported across national boundaries in most parts of Europe, and the countries have joined their efforts to reduce the total emission of mercury and other heavy metals. Emission scenario used in modelling of mercury transport and deposition for 2000 is based on data officially submitted to the UNECE Secretariat from EMEP countries as well as on expert estimates. The EMEP-model has been used to make matrices showing transport of mercury to and from, e.g. Norway. The modelling has been carried out by EMEP-MSC-East and the work are presented at http://www.msceast.org/.

2.3. Sediments

Data on lake sediments is based on a nationwide survey from 1996 to 1997 (Rognerud and Fjeld, 2001). These lakes had been collected based on the following criteria: coverage throughout Norway, but with more lakes from areas with expected greater influence from atmospheric deposition, a wide range in lake size and water quality, and no significant local pollution. The lakes and the catchments covered a wide range of conditions regarding bedrock geology, thickness of overburden, amount of precipitation, and types of vegetation. Cores from the deepest part of each lake were collected. To document historical changes in concentrations of mercury in the sediments, the upper 0.5 cm surface section and a 1-cm section from the deepest part of the core, normally within 30–50 cm depth, were sampled. The deepest layer was defined as a "reference layer" likely to have been deposited in pre-industrial times, and therefore representing the natural background concentration of mercury in sediment. Sedimentation rate in Nordic subalpine lakes are about 0.5 ± 0.3 mm and about 1.2 ± 0.5 mm/year in Nordic forest lakes. Thus, the surface sediments represent the last 10 years in subalpine lakes and the last 4 years in forest lakes. Reference sediments are more than 400 years old.

2.4. Peat cores

Peat cores from ombrotrophic bogs represent an alternative method for looking at time trends in atmospheric deposition of metals, provided that the vertical leaching of the metal concerned is small. Peat cores down to a depth of 100 cm were sampled from six bogs situated in different parts of Norway (Steinnes and Sjøbakk, 2005). Samples were taken at 2.5-cm intervals down to 15 cm, 5–10 cm intervals at 15–30 cm depth, and 10-cm intervals at 40–100 cm depth.

2.5. Moss

Moss surveys are performed regularly in Norway (1977, 1985, 1990, 1995, 2000) to assess temporal

Table 1
Official Norwegian emissions of mercury to air, transport from Norwegian sources to EMEP countries (modelled by EMEP-MSC-East), transport from Norwegian sources to areas outside EMEP (modelled by EMEP-MSC-East), transport to Norway (modelled by EMEP-MSC-East), and National emissions compared to contribution from other countries

Official Norwegian emissions to air	Transport from Norwegian sources to EMEP countries	Transport from Norwegian sources outside EMEP	Transport to Norway	National emissions compared to contribution from other countries
960 kg	281 kg (30%)	671 kg (70%)	2594 kg	37%

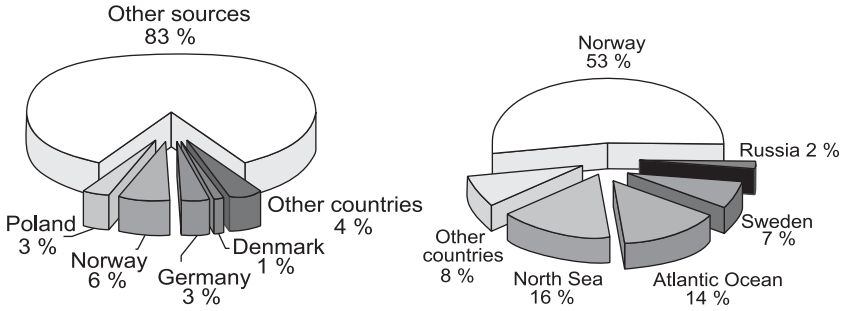

Fig. 1. Left: Transport of Hg to Norway from the EMEP area. Right: Transport of mercury from Norwegian sources to EMEP countries. Source: www.msceast.org/EMEP.html.

and spatial changes in heavy metal atmospheric deposition (Steinnes and Andersson, 1991; Berg et al., 1995; Berg and Steinnes, 1997; Steinnes et al., 2003). Similar investigations are carried out simultaneously in Sweden, Finland and Denmark, and also in other parts of Europe (Rühling et al., 2002). The basis of the moss technique as a mean of surveying atmospheric deposition of metals is that the carpet-forming species obtain much of their supply of chemical substances directly from precipitation and from dry deposition of airborne particles. Furthermore, mosses have a high capacity to sorb and retain a number of trace elements. This means that mosses have found a large number of applications as biomonitors. In the Norwegian surveys samples of *Hylocomium splendens* have been collected at 500 sites throughout the country. The samples were collected at least 300 m from main roads and populated areas, and at least 100 m from local roads, single houses and agricultural areas.

2.6. Natural surface soil

Nationwide sampling of natural surface soils was carried out in 1977 (Steinnes et al., 1997), 1985 (Njåstad et al., 1994) and 1995 (Nygård, 2000) according to a similar network as for the moss sampling. Soils from 2 to 5 cm depth of the humus layer at 170 localities sampled in 1985 were analysed for Hg (Steinnes et al., 1997). There exists almost no data for Hg in mineral soils from Norway.

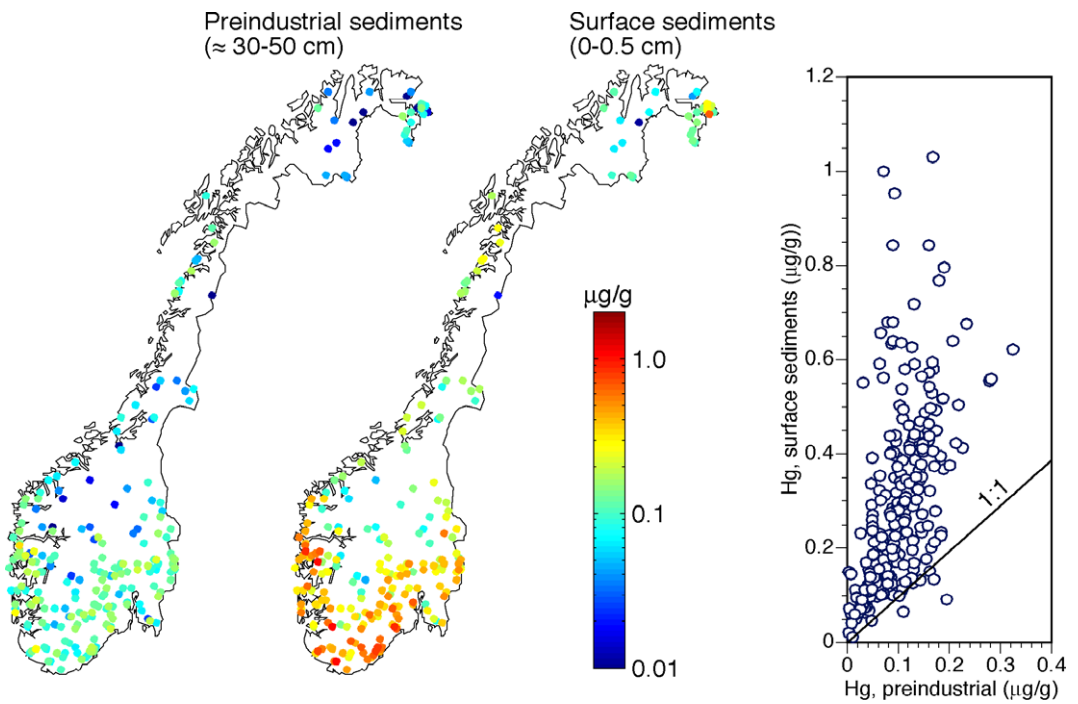

Fig. 2. Hg concentration (µg/g) in surface and reference sediments collected in 1996–1997.

2.7. Precipitation

Mercury in precipitation has been determined at the extreme coastal station Lista in the southernmost part of Norway since 1990 as part of the OSPAR program. This is so far the only station with long time precipitation data on Hg in Norway. The samples were collected on a monthly basis using duplicate IVL bulk samplers and analysed using acid digestion followed by Cold vapour atomic fluorescence spectroscopy (CV-AFS). Details on sampling and analysis are given in Berg et al. (2001).

3. Results and discussion

3.1. Norwegian emissions of mercury and fluxes from other countries

EMEPs budgets for transport of Hg to and from Norway for 2000 are shown in Table 1 together with

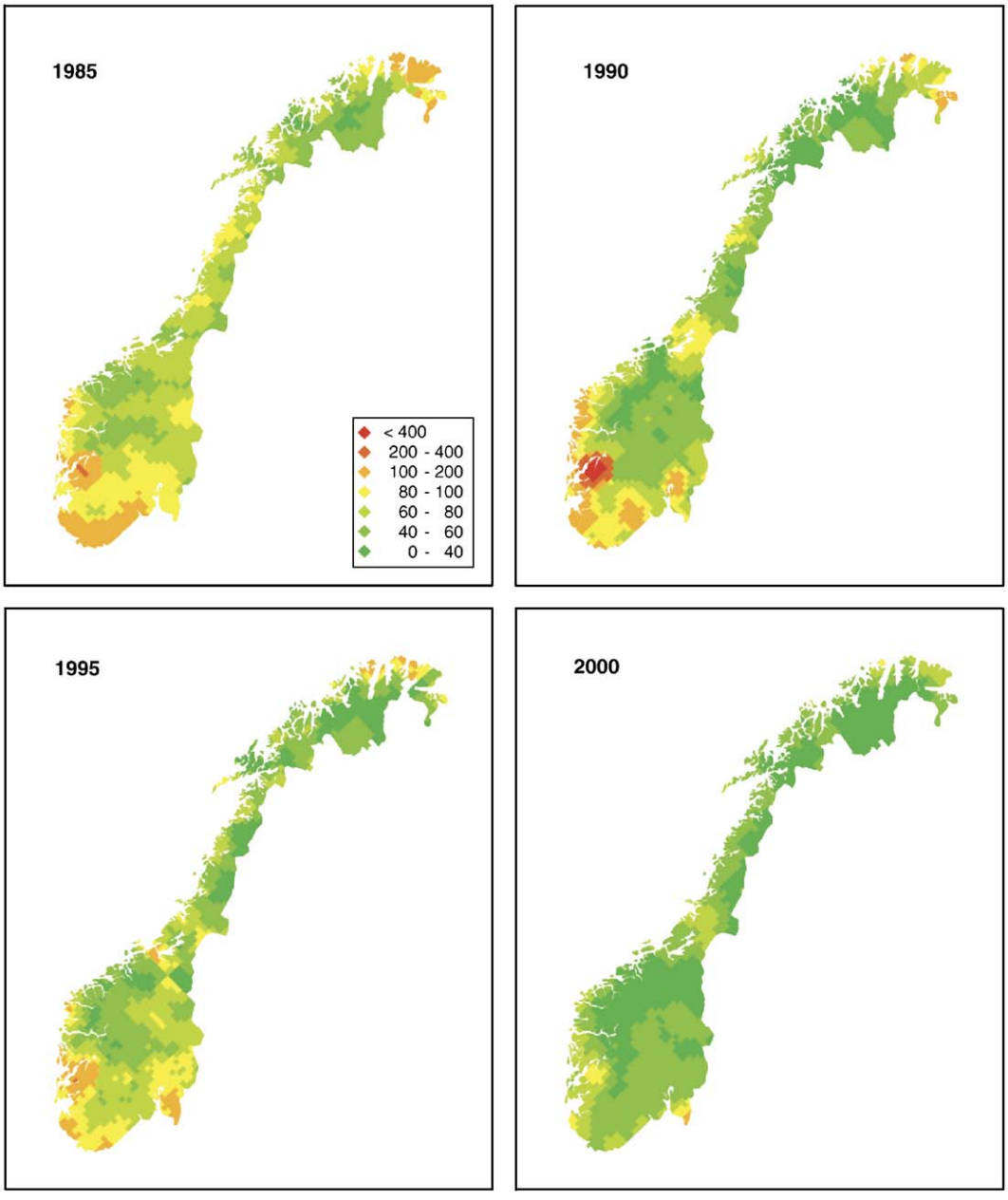

Fig. 3. Hg concentration (ng/g) in moss collected in 1985, 1990, 1995, 2000.

Norwegian official figures on emissions to air. In all 960 kg Hg was emitted to the atmosphere from Norwegian sources. About 280 kg (30%) of this amount is deposited in EMEP countries, whereas about 670 kg (70%) is deposited outside the EMEP area. About 2600 kg is transported to Norway from sources outside the Norwegian borders. Norway receives more Hg (approximately a factor of 3) from other countries than domestic emissions. Matrices for Hg transport to and from Norway are shown in Fig. 1. Countries outside Norway contributing significantly to the Hg loadings in Norway are Poland, Germany, and Denmark. "Other countries" in Europe also contribute. Due to the uncertainty in the data material a significant part of the atmospheric long-range transported Hg cannot be ascribed to emissions from specific countries, but has to be placed in the category "other sources". In this category reemission and contributions from countries outside Europe are also included. The matrices show that of the 280 kg deposited to EMEP countries, approximately half are deposited within own borders. About 30% are deposited in the Atlantic Ocean and the North Sea. Norway also contributes significantly to Sweden and Russia. It is important to keep in mind that both emission figures and modelled data are associated with uncertainties, and should be viewed as indications only.

3.2. Atmospheric deposition of long-range transported mercury

The nationwide survey on lake sediments from 1996 to 1997 (Fig. 2) showed typical surface sediment Hg concentrations in the range of 0.14–0.42 ug/g (25th to 75th percentile). The highest degree of impact was observed in the southern part of the country, with a pronounced decrease with latitude and altitude (Rognerud and Fjeld, 2001). The highest concentrations were found in the low-land coastal areas in South-Norway (forest lakes), with concentrations usually in the range of 0.3–0.6 ug/g, but concentrations greater than 1 ug/g was also observed. In Central to Northern Norway and in mountain areas in Southern Norway the concentrations were usually in the 0.1–0.2 ug/g range. Compared to the pre-industrial sediments had the surface sediments significantly elevated Hg concentrations, usually by a factor of 2–4 in the country as a whole. However, in forest lakes in Southern Norway were the surface sediment concentrations commonly 4–6 times higher than in the pre-industrial sediments. We attribute the elevated Hg concentrations to be mainly caused by anthropogenic emissions.

Results from the analysis of peat cores provided even stronger indications of increased atmospheric deposition of Hg in Norway during industrial times (Steinnes and Sjøbakk, 2005). Concentrations in the surface layer were in the range 200–400 ng/g with the highest levels far south and far north. Concentrations at depth were of the order 10% of the surface peat levels. The results indicate that the Hg accumulation over the last 100 years is about 15 times higher on average than the pre-industrial level.

The national surveys on Hg in moss showed similar concentrations from 1985 to 1995, whereas the results from 2000 showed a general decrease (30%) for the whole country (Fig. 3). In 2000 the normal background concentrations in Norway were less than 40 ng/g. The 25–75 percentile were 40–66 ng/g, whereas the concentrations in the southernmost areas most influenced by atmospheric long-range transport were about 80 ng/g. The distribution trend in the 2000 samples is very similar to those observed in previous years (Steinnes and Andersson, 1991; Steinnes et al., 2003). The Hg level in moss is relatively uniform all over the country, with slightly higher values in the far south and an apparent decrease from the coast inland. The mosses show a less significant north–south gradient than for sediments and precipitation (see later in this article). The less pronounced gradient could be explained by a considerable supply of Hg to the moss

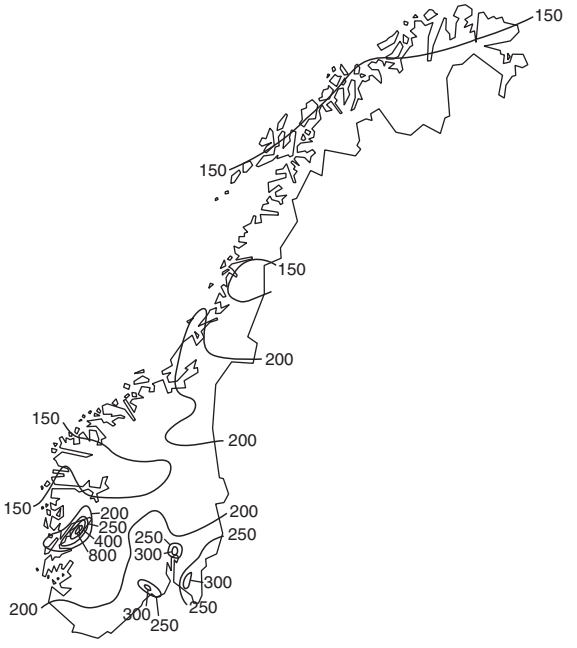

Fig. 4. Hg concentration (ng/g) in natural surface soil collected in 1985.

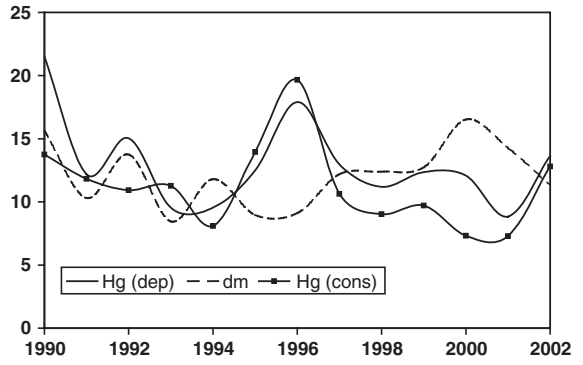

Fig. 5. Hg in precipitation. Hg (dep): mercury in deposition (μg m^{-2} $year^{-1}$), Hg (conc): mercury concentrations in precipitation (ng l^{-1}), dm: precipitation amount in dm (mm*100).

from dry deposition of Hg^0 in addition to the retention of Hg^{2+} from wet deposition. Presumably the cool summer climate particularly in the middle and northern parts of the country explains that some of the Hg^0 retained in the moss is not lost by re-volatilisation.

Hg concentrations in natural surface soil were quite similar throughout the country, but a somewhat higher level in the south indicates a possible contribution from atmospheric long-range transport (Fig. 4). A representative level for Norway is in the range 137 ng/g (25 percentile) to 202 ng/g (75 percentile). It is difficult to say how much this value is affected by a possible increased level of elemental mercury in the atmosphere in modern time. In addition to atmospheric deposition from local and long range transport sources, contributions may also come from sources such as atmospheric deposition from natural processes and possibly also from upward transport from mineral soil, although existing data indicate that the Hg content of mineral soils are generally very low (Steinnes, 1995).

Fig. 5 shows the time trend for Hg deposition at Lista for the period 1990–2002 (annual averages). The Hg deposition has varied between 21 μg m^{-2} $year^{-1}$ and 9 μg m^{-2} $year^{-1}$, but the use of Mann-Kendall test yields no statistically significant trend. Hg observations in air at six sites in the Northern Hemisphere, two sites in the Southern Hemisphere, and during eight ship cruises over the Atlantic Ocean provided a consistent picture suggestion that Hg concentration increased since the first measurements in 1977 to a maximum in 1980s and then decreased to a plateau in 1996–2001 (Slemr et al., 2003). No significant trend was, however, found for Hg in air measured at the Zeppelin station, Svalbard during 1994–2002 (Berg et al., 2004). On average the deposition values at Lista for the years 1999–2002 were 30% higher than at other OSPAR-CAMP sites (Wängberg and Munthe, 2004). This is not due to significant differences in concentrations, but owing to the higher precipitation rate at Lista (1200 mm). Iverfeldt (1991) found a steep south–north gradient in the Hg deposition varying from 27 μg m^{-2} $year^{-1}$ at Birkenes in southern part of Norway to 3 μg m^{-2} $year^{-1}$ at Jergul in the far north of the country for the period 1987–1989. The annual precipitation rates at those stations were 1730 and 340 mm, respectively.

To conclude, the atmospheric long range transport to Norway are larger than the national Norwegian emissions of Hg. Concentrations of Hg in peat cores and reference sediments indicate that the current Hg levels measured in surface sediments, surface soils and mosses at background sites in Norway are substantially affected by long-range atmospheric transport.

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