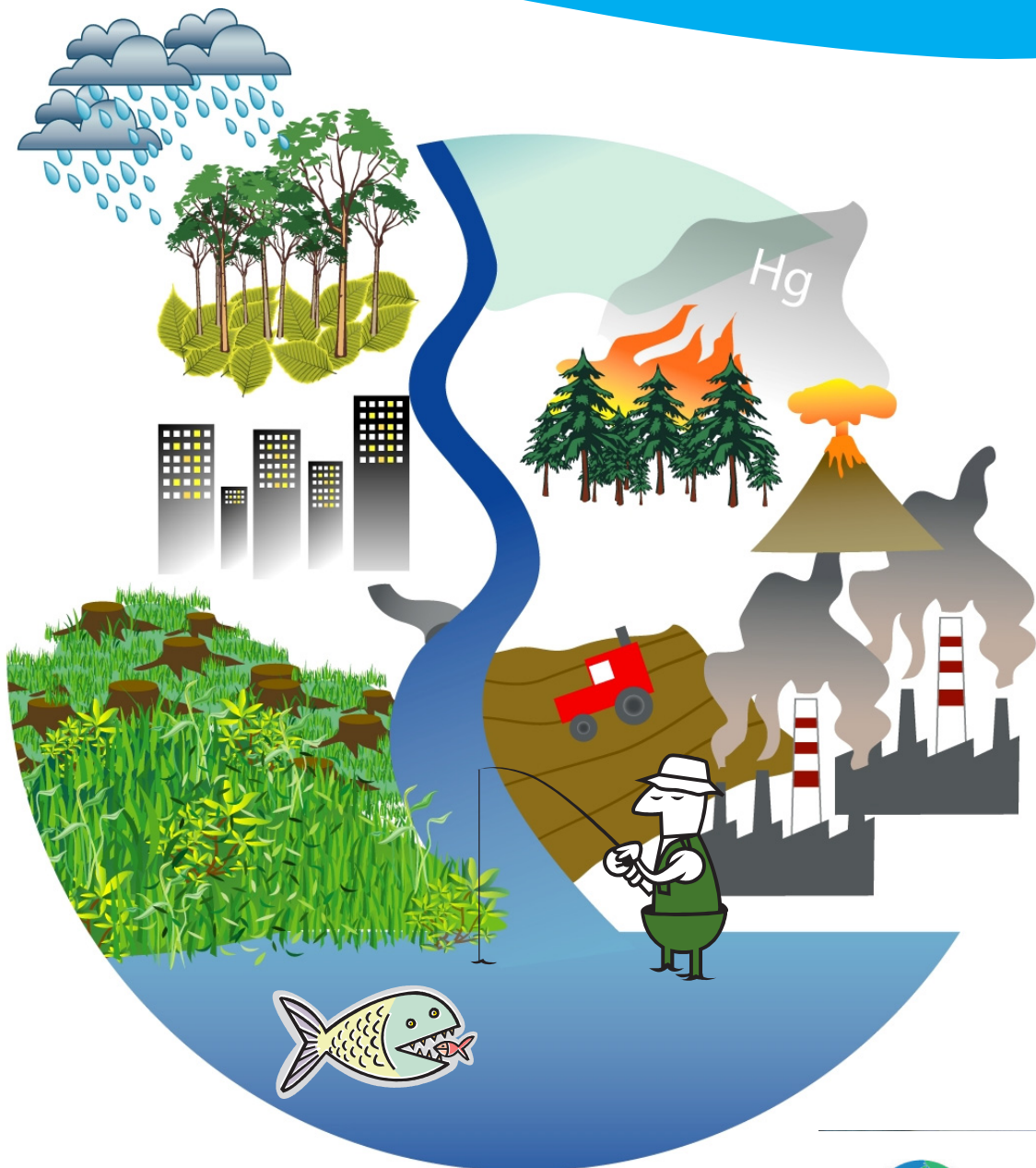


ICP Waters Report 97/2009

An assessment of Hg in the freshwater aquatic environment related to longrange transported air pollution in Europe and North America



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Abstract Long-range transboundary atmospheric transport of the pollutant mercury (Hg) (LTRAP-Hg) poses an ecological threat to aquatic ecosystems and biota. Through fish consumption, mercury and its highly toxic and bioaccumulative organic form methylmercury (MeHg) may cause harmful effects on human health. Factors controlling the bioaccumulation of Hg in aquatic ecosystems are not well understood. In Scandinavia an increased content of Hg in piscivorous fish has been observed the last decade. Today, legislation and guidelines on concentration levels of Hg for fish retail and private consumption are in place in most countries and the utility of the fishery as a resource is reduced. This report gives an overview of:

- relevant international agreements on Hg pollution
- factors that drive the spreading and accumulation of Hg in the environment
- current knowledge on the status of LRTAP Hg in freshwater aquatic ecosystems (fish, sediments, water) within the UNECE region (North America and Europe).
- recommendations for an outline of a monitoring network suitable for assessing the distribution and effects of LTRAP-Hg in freshwater aquatic environments.

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1. Kvikksølv	1. Mercury
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CONVENTION ON LONG-RANGE
TRANSBOUNDARY AIR POLLUTION

INTERNATIONAL COOPERATIVE PROGRAMME
ON ASSESSMENT AND MONITORING EFFECTS
OF AIR POLLUTION ON RIVERS AND LAKES

An assessment of Hg in the freshwater aquatic environment
related to long-range transported air pollution in Europe and
North America

Prepared at the ICP Waters Programme Centre
Norwegian Institute for Water Research
Oslo, November 2009

Preface

The International Cooperative Programme on Assessment and Monitoring Effects of Air Pollution on Rivers and Lakes (ICP Waters) was established under the Executive Body of the UNECE Convention on Long-Range Transboundary Air Pollution (LRTAP) in July 1985. Since then ICP Waters has been an important contributor to document the effects of implementing the Protocols under the Convention. Numerous assessments, workshops, reports and publications covering the effects of long-range transported air pollution has been published over the years.

The ICP Waters Programme Centre is hosted by Norwegian Institute for Water Research (NIVA), while the Norwegian Pollution Control Authority (SFT) leads the programme. The Programme Centres work is supported financially by SFT.

The main aim of the ICP Waters Programme is to assess, on a regional basis, the degree and geographical extent of the impact of atmospheric pollution, in particular acidification, on surface waters. More than 20 countries in Europe and North America participate in the programme on a regular basis.

The pollutant mercury (Hg) is addressed in the Aarhus Protocol under the LRTAP Convention. It is of particular interest in surface waters, as alarmingly high levels of Hg have been reported in fish in remote lakes. Currently, no international monitoring network for assessing distribution and effects of long-range transported Hg in freshwater ecosystems exists. The objective of this report was i) to assess the status of Hg in the aquatic environment based on published sources and ii) to make recommendations on the outline of a network suitable for monitoring Hg in freshwater ecosystems. The report supplies also an introduction to international agreements addressing Hg and factors that control distribution of Hg in the environment.

We are grateful for the contribution in the discussions of an earlier version of the report that was presented at the 24th Task Force meeting in Budapest October 6-8 2008 and for all written comments to the second draft of the report. In particular we want to thank Ton de Nijs, the Netherlands, Jens Fölster, Sweden, and Bjørn Olav Rosseland, Norway, who were appointed by the Task Force meeting to review the final draft.



Brit Lisa Skjelkvåle
ICP Waters Programme Centre

Oslo, November 2009

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Executive summary

The long-range transported pollutant mercury (Hg) is addressed in the Aarhus Protocol under the LRTAP Convention. It is of particular interest in surface waters, as alarmingly high levels of Hg have been reported in fish and sediments in remote lakes in northern boreal and arctic ecosystems with possible consequences for people and wildlife.

Currently, no international monitoring network for assessing distribution and effects of long-range transported Hg in freshwater ecosystems exists. The objective of this report is i) to assess the status of Hg in the aquatic environment based on published sources and ii) to make recommendations on the outline of a network suitable for monitoring Hg in freshwater ecosystems.

The ecologically most relevant form of Hg is methylmercury (MeHg), an organic form of Hg that is highly toxic and bioaccumulates in the foodweb. In fish, 95-99% of the Hg is the form of MeHg. Concentrations of Hg in fish in Europe and North America are often above the limit recommended for human consumption, especially in piscivorous fish. Recent studies from Scandinavia show increasing Hg levels in fish despite reduced Hg deposition. In South-Eastern Norway the concentration of Hg in perch has increased with 63% from 1991 to 2008.

Lake sediments are used as archives for atmospheric deposition. Regional lake sediment surveys in Northern America, Scandinavia, northern parts of Russia, and in the Arctic show that recent lake sediments are considerably enriched relative to sediments from pre-industrial times. The last 10-15 years, Hg contents in lake sediments have gradually declined, probably due to reduced emissions.

Concentrations of Hg in remote surface waters are generally low, usually below 40 ng L⁻¹ although slightly higher in waters affected by natural organic matter. Methylmercury (MeHg) usually comprises between 0.1 to 30% of total Hg in water. In surface waters, concentrations of Hg and MeHg are strongly controlled by catchment processes. Despite these low concentrations of Hg in the water, a considerable increasing concentration up through the food web is observed.

The threshold values for Hg in fish (0.3-1.0 mg Hg kg⁻¹), based on evaluations of human health risks through exposure via fish consumption, are considerably higher than environmental quality standards (EQS) for Hg in fish (0.02 mg Hg kg⁻¹) under the EU Water Framework Directive. Almost all fish in the compiled data in this report exceed the EQS-values, while a substantial number do not exceed the limit for human consumption. The balance between exposure to Hg through fish consumption versus its potential health benefits, is much discussed internationally.

Recommendations for a monitoring programme for Hg in freshwater ecosystems with the aim of assessing the effects of Hg originating from long-range atmospheric transport are made regarding site selection, receptors, methods, and the collection of supporting data.

Summary

Mercury, a long-range transboundary pollutant causing an environmental problem

Long-range transboundary atmospheric transport of the pollutant mercury (Hg) emitted to the atmosphere through industrial activity leads to increased exposure of humans and wildlife to the highly toxic organic form of Hg, i.e. methylmercury (MeHg). The major route of exposure of humans and many wildlife species to Hg and MeHg is through consumption of piscivorous fish. In the EU and the US the maximum levels of total Hg in freshwater fish allowed for retail is 0.5 mg kg^{-1} wet weight (ww) and 0.3 mg kg^{-1} ww, respectively. Since exposure to elevated concentration of MeHg may cause damage to the nervous system, heart, and other organs, advices related to fish consumption are established in most countries.

Objectives of this report

Currently, no international monitoring network for assessing distribution and effects of long-range transported Hg in freshwater ecosystems exists. The objectives of this report are i) to assess the status of Hg in the aquatic environment (fish, sediments, water) within the UNECE region (North America and Europe) based on published sources and ii) to make recommendations on the outline of a network suitable for monitoring Hg in freshwater ecosystems.

Several international agreements concern mercury in the environment

Under the Convention for Long-range Transboundary Air Pollution (LRTAP), the Aarhus Protocol on Heavy Metals was signed in 1998 and entered into force in December 2003. This Protocol targets Hg, cadmium (Cd) and lead (Pb), with the aim of reducing emissions of these metals to levels below those of 1990. Other important international Conventions are the OSPAR, HELCOM, and ACAP.

Concentrations of mercury species in surface waters are low

Few data and long term studies exist on aqueous concentrations of Hg and MeHg in surface waters from remote areas because of low concentrations and demanding analysis. Most data are as total Hg, even though MeHg is more important for evaluation of the environmental risk of Hg. In lakes and watersheds not directly influenced by point sources, total Hg concentrations are often in the range of $0.3\text{--}8 \text{ ng L}^{-1}$. In pristine lakes high in natural organic matter (NOM), concentrations can be considerably higher, in the range of $20\text{--}40 \text{ ng L}^{-1}$. Methylmercury usually comprises about 0.1-30% of total Hg in surface waters.

Lake sediments are enriched by long-range atmospheric transport of mercury

Lake sediment cores from remote, temperate and boreal areas and the Arctic are studied as archives for Hg deposition. In general, the cores reveal an increase in Hg from past to present, with a particular increase after the onset of the industrial revolution, peaking in the late 20th century. The last 10-15 years, Hg contents in lake sediments have gradually declined, probably due to reduced emissions. The ratio of Hg between surfaces to deep cores not influenced by industrial deposits is referred to as the enrichment factor (EF). Where $EF > 1$ this indicates influence by external sources. Enrichment factors of 1.1 to 30 are reported, but the most common values are roughly between 1.3 and 6. Generally, sediment enrichment of Hg is highest in regions with the highest atmospheric deposition.

Concentrations of mercury in fish in Scandinavia and North America are frequently above the limit recommended for human consumption

Levels of Hg in fish vary with species, size, age, differences in Hg exposure, food web structure, and dietary strategy. Surveys of Hg in fish from various regions follow different designs, and the collection of supporting data that allows interpretation of results in relation to key controlling factors, is done to varying degrees. Thus, differences in Hg levels in fish sampled in different surveys may be controlled by fish size or position in the aquatic food web rather than by differences in Hg deposition. However, the highest concentration of mercury in fish is often found in piscivorous species and top predators. In Scandinavia and North America elevated concentrations of Hg are often found in northern pike and perch, and the concentrations are also often above the limit recommended for human consumption. Compiled results on Hg from Scandinavia have shown that Northern pike caught in Scandinavia have slightly elevated levels of Hg compared to North America. For perch, the concentrations are comparable or somewhat lower. Recent published reports from Scandinavia have showed that Hg content in fish has increased from the 90s to the 2008 despite reduced emissions. In south eastern Norway the concentration of Hg in perch has increased with 63% from 1991 to 2008.

Comparison of mercury in water and fish with environmental quality standards under the EU Water Framework Directive (WFD)

Mercury concentrations in surface waters are usually lower than the WFD environmental quality standard (EQS) of 0.05 µg/l. However, in many of these waters the Hg concentrations in fish are substantially higher than the recommended limits for human consumption (0.3-1.0 mg kg⁻¹) and the EQS-values for Hg in fish (0.02 mg kg⁻¹). Many of these lakes where concentrations of Hg in surface waters are lower than 0.05 µg/l and Hg content in fish above recommended limits for human consumption are found in areas where long range atmospheric deposited Hg is the main source. This situation is often found in Scandinavian aquatic ecosystems. Thus, the concern with regard to Hg pollution and human and wildlife exposure to Hg may not be addressed satisfactorily under the WFD.

Recommendations for monitoring Hg from LRTAP in aquatic ecosystems

A monitoring programme for Hg in freshwater ecosystems with the aim of assess effects of Hg originating from long-range atmospheric transport, should avoid sites affected by local sources or other disturbances in the catchments that could alter the fluxes of Hg to the rivers and lakes. Recommended Hg-receptors are water, sediments, and fish. Each receptor has its strengths and weaknesses with respect to insights into short-term or longer term processes controlling the spreading and accumulation of Hg in the aquatic environment. If only one receptor would be chosen, then both the ambition and the aim of the monitoring program, as well as the cost and analytical challenges, should be considered in order to make the best possible choice. Collection of relevant supporting data to facilitate cross survey comparison and interpretation is advised. Analysis of Hg species is recommended to be performed by dedicated laboratories.

1. Mercury - an environmental problem

Long-range transboundary transport of mercury (Hg) emitted to the atmosphere through industrial activity leads to increased exposure of humans and wildlife to the highly toxic organic form of Hg, i.e. methylmercury (MeHg, CH_3Hg^+). In remote areas, long-range atmospherically transported Hg is the main source of Hg, and elevated concentrations have been observed in sediments and fish. The major route of exposure of humans and many wildlife species to Hg and MeHg is through consumption of fish, especially piscivorous species. Exposure to elevated concentrations of MeHg may cause damage to the nervous system, heart, and, other organs (Clarkson, 1997; Magos and Clarkson, 2006).

Controlled feeding studies show that diets containing MeHg at environmentally realistic concentrations resulted in toxic effects in fish, birds, and mammals. Limited field-studies have corroborated these laboratory-based results, demonstrating significant relations between MeHg exposure and various indicators of MeHg toxicity, including reproductive impairment (Scheuhammer et al., 2007). For humans, reduced cognitive abilities have been observed for children exposed to elevated levels of MeHg in a prenatal stage (Grandjean et al., 1997). People who regularly consume fish, especially predatory fish species, are at risk for elevated exposure to MeHg with associated risks for their health (Mergler et al., 2007). In addition, elevated concentration of Hg in fish will adversely affect the quality and the economy of fisheries as a resource.

Today several countries have recommendations and legislation for fish sale and maximum human consumption due to concern for human exposure to Hg, and its impacts on human health. These advices depend on factors such as fish species, body size (age) (Lockhart et al., 1995), capture site, and target group (pregnant, nursing mothers, and young children). Today, the greatest concern for Hg is to the developing foetus and to small children through dietary exposure (Harris et al., 2007). In the EU and the US the maximum levels of total Hg in freshwater fish allowed for retail is $0.5 \text{ mg kg}^{-1} \text{ ww}$ and $0.3 \text{ mg kg}^{-1} \text{ ww}$, respectively. Canada was one of the first countries that established a maximum allowable level of Hg in commercially sold fish in 1970.

Monitoring of Hg levels in natural ecosystems, sediments, water, and aquatic organisms is carried out in many countries but at a considerably lower intensity and regional coverage than that carried out for monitoring the effects of acid deposition. There are no internationally agreed methods of how to monitor and evaluate the effects of Hg-deposition on aquatic ecosystems.

The objectives of this report are

- i) to assess the status of Hg in the aquatic environment (fish, sediments, water) within the UNECE region (North America and Europe) based on published sources
- ii) to make recommendations on the outline of a network suitable for monitoring Hg in freshwater ecosystems, including monitoring design, analytical methods and assessment of data.

Today, no data on Hg are reported to ICP Waters Programme. Therefore, the assessment is based on other existing surveys and datasets. Additionally, both an overview of international agreements regarding Hg as a pollutant and a review of factors that affect the distribution of Hg in aquatic ecosystems, are provided.

2. International agreements on mercury

2.1 The Aarhus Protocol

Similar to several other long-range transported pollutants, Hg is transported through the atmosphere and deposited far away from its emission source. The potentially harmful effects of Hg have been recognised and international measures taken to reduce emissions. Under the LRTAP Convention, the Aarhus Protocol on Heavy Metals was signed in 1998 and entered into force in December 2003. This Protocol targets three harmful metals, i.e. cadmium (Cd), lead (Pb) and Hg, and aims to reduce emissions of these metals to levels below those of 1990 (or an alternative year between 1985 and 1995). The Protocol aims at cutting emissions from industrial sources (the iron and steel industry, the non-ferrous metal industry and cement production), from combustion processes (power generation, road transport) and from waste incinerations. Stringent emissions limits from stationary sources are laid down and the best available techniques (BAT) for emission reduction are suggested, such as special filters or scrubbers for combustion sources or Hg-free processes. Scientific support for the development and implementation of this and other protocols under the CLRTAP, is provided by International Cooperative Programmes like ICP Waters that operate under the CLTRAP convention.

2.2 Other international conventions and agreements

There are several international Conventions that regulate the use of Hg and its emission to the environment, some of which are listed below. The most recent international agreement was signed by the United Nations Environment Programme (UNEP) in Nairobi, February 2009. In this agreement, a tightening of mercury regulation was approved. The partnership plan agreements were focused on improving the world-wide capability for nations to safely store stockpiled mercury, reduce the supply of mercury from primary mining, carry out awareness rising alongside projects to cut the use of mercury in artisanal mining, and reduce use of mercury in products.

The OSPAR Convention for the Protection of the Marine Environment of the North-East Atlantic. The objective of the OSPAR Convention to prevent and eliminate pollution is reflected in a strategy on hazardous substances which was agreed in 1998. This Convention has the ultimate aim of achieving concentrations in the marine environment near background values for naturally occurring substances (such as Hg) and close to zero for man-made synthetic substances, with every endeavour to be made to move towards the target of cessation of discharges, emissions, and losses of hazardous substances by 2020.

The Helsinki Convention (HELCOM) on the Protection of the Marine Environment of the Baltic Sea Area. This Convention aims to prevent and eliminate pollution in order to promote the ecological restoration of the Baltic Sea Area and the preservation of its ecological balance. A specific strategy on hazardous substances was adopted in 1998. Its objective is to prevent pollution by continuously reducing discharges, emissions and losses of hazardous substances towards the target of discharge cessation by 2020. The ultimate aim is to achieve concentrations in the environment near background values for naturally occurring substances and close to zero for man-made synthetic substances.

The Basel Convention on the Control of Transboundary Movements of Hazardous Wastes and their Disposal. This Convention strictly regulates the transboundary movements of hazardous wastes and establishes obligations for parties to ensure such wastes are managed and disposed of in an environmentally sound manner. Any waste containing, or contaminated by Hg or its compounds is considered hazardous waste and is covered by the provisions of the Convention. Hazardous wastes may not be exported from the EU or OECD for disposal, recovery or recycling in other countries.

The Rotterdam Convention on the Prior Informed Consent (PIC) Procedure for Certain Hazardous Chemicals and Pesticides in International Trade. The Convention establishes the principle that export of specified chemicals and pesticides can only take place with the prior informed consent of the importing party. At present, Hg compounds used as pesticides are covered by the PIC procedure, while Hg and its compounds intended for industrial use are not.

The Arctic Council Action Plan to Eliminate Pollution of the Arctic (ACAP). The Arctic Council is a high-level intergovernmental forum that provides a mechanism to address the common concerns and challenges faced by the Arctic governments and peoples. The ACAP, agreed to in 1997, prioritises a number of pollutants of special concern for the Arctic region, including Hg. Planned activities include identification and quantification of major point sources, with the aim of implementing concrete emission reduction pilot projects.

The Nordic Environmental Action Programme 2001-2004. This programme establishes environmental priorities within the framework of Nordic cooperation in the fields of nature and the environment. It follows up on commitments in a Nordic sustainable development strategy, which has as one of its objectives the discontinuation, within 25 years, of discharges of chemicals posing a threat to health and the environment.

International action relating to use of mercury in artisanal gold mining. A number of international bodies including the International Labour Organisation, the World Bank, and the United Nations Industrial Development Organisation (UNIDO) in particular, have worked on this issue. UNIDO hosts the Global Mercury Project, also supported by the United Nations Development Programme (UNDP) and the Global Environment Facility (GEF), which is undertaking projects in countries affecting international waters with Hg from artisanal mining.

EU Water Framework Directive. In the European Union (EU) and associated countries a joint strategy was developed to secure the Community's action towards a common water policy. The EU Water Framework Directive (WFD) aims to protect and restore clean water across Europe and to ensure its long-term and sustainable use. The major part on Community strategy against pollution of surface waters control policy is set out in Article 16. This Article requires the establishment of a list of priority substances and a procedure for the identification of priority substances and priority hazardous substances as well as the adoption of the specific measures against pollution with these substances. Mercury is within the WFD identified as a priority hazardous substance, and regulated in the Priority substances directive (2008/105/EC), which is a daughter directive of the WFD. Through the WFD, environmental quality standards (EQS) applicable to aquatic ecosystems have been established. These standards are based on various ecotoxicological parameters and should be set so as to limit the

quantity of certain chemical substances that pose a significant risk to the aquatic environment or to health in surface water in the EU. EQS-values for various receptors have been defined, and if the EQS-values are exceeded, risk assessment and remediation measures may be required. In the WFD, EQS-values for mercury are set at 0.02 mg kg^{-1} wet weight in freshwater fish, while in water samples, the annual average EQS of dissolved mercury in water is set at $0.05 \text{ } \mu\text{g L}^{-1}$.

3. Emissions, deposition and cycling of mercury

Hg occurs in the environment as a result of both natural and anthropogenic releases, but human activity has changed the extent of releases and the cycling of Hg. Since the onset of the Industrial Revolution an enrichment of Hg in the environment has been observed, and today Hg is a global pollutant. Elevated levels of Hg in the environment are caused by direct discharge to water as well as emission to air. Recent estimates suggest that anthropogenic emission of Hg to air accounts for one to two thirds of the total global Hg emissions (UNEP, 2008; UNEP/AMAP, 2008).

Anthropogenic Hg emissions originate from several sources, with fossil fuel combustion and metal production being the main contributors. Natural Hg emissions are mostly from geological activities and degassing from soil and oceans. **Figure 1** shows the estimated proportions of global anthropogenic emission of Hg by sectors in 2005. In 2005, emissions were estimated to 1220-2900 tonnes (UNEP/AMAP, 2008), indicating substantial uncertainties in the calculations.

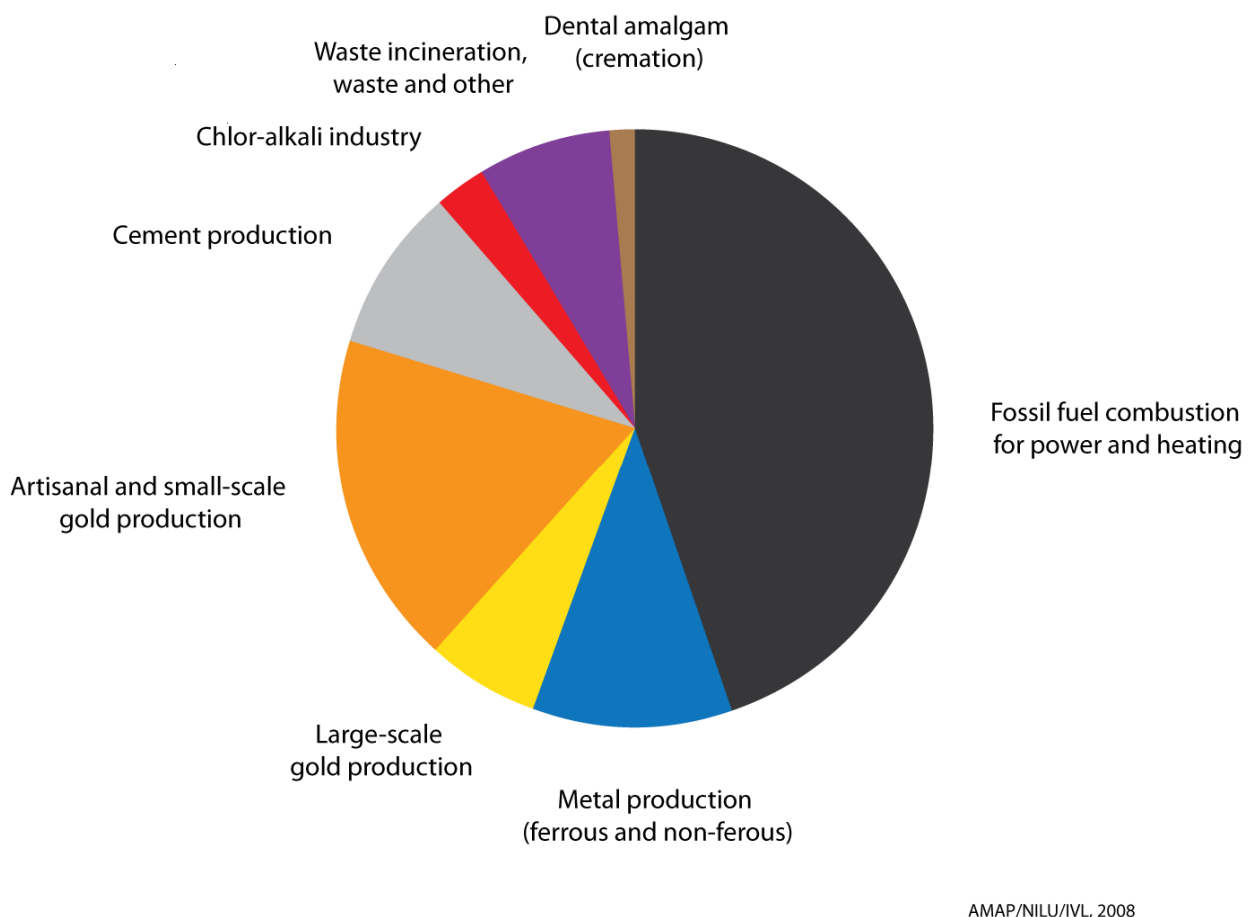


Figure 1. Estimated global emissions of anthropogenic mercury to air in 2005 by proportions and sectors (UNEP/AMAP, 2008).

Hg is emitted to the atmosphere in three different chemical forms:

- gaseous elemental Hg (GEM, Hg⁰),
- oxidised Hg (Hg(II)) in the form of so called reactive gaseous Hg (RGM),
- associated with particles and defined as total particulate Hg (TPM)

The lifetime of GEM in the atmosphere is currently estimated to be 0.8-1.3 year, while RGM and TPM have a lifetime of hours to days (UNEP/AMAP, 2008). As a consequence, RGM and TPM are generally deposited closer to its sources, while GEM is subjected to long-range transport, making Hg a global pollutant. Chemical reactions of Hg in the atmosphere are complex and involve kinetics that are not completely understood (Lindberg et al., 2007). According to UNEP (2008), knowledge gaps exist in the understanding of key chemical processes, kinetics of Hg removal from the atmosphere, seasonal variation in emission, information of natural emissions, and the exact chemical composition of RGM and TPM.

It is generally agreed that the global trend of anthropogenic Hg has increased after the onset of the Industrial Revolution. Global Hg emission inventories and emission measurements depict somewhat different trends in the recent years. Inventories of Hg emissions estimate a reduction in the 1980s and almost constant level afterwards (UNEP/AMAP, 2008), while measurements of Hg suggest an increase in the 1980s and a decrease in the 1990s. According to (Pacyna et al., 2006), a recent reduction in Hg emission has been observed in Europe and North-America, while in Asia, the emissions have increased. Today, Hg in air and deposition/precipitation are monitored by several regional and national networks around the world. Initially, only wet deposition was measured manually. Today GEM is analysed automatically (Slemr et al., 2003), while RGM and TPM are not measured on a regularly basis (UNEP, 2008). A comparison of trends in concentration of airborne total GEM measured at several monitoring stations in the Northern and Southern Hemispheres is shown in **Figure 2**.

Monitoring stations are sparse in remote areas (**Figure 2**), and few have data series that are more than 30 years long. Concentrations of Hg in air on the Southern Hemisphere appear to be about one third lower than the Northern Hemisphere. Concentrations on each Hemisphere are evenly distributed, even if stations are located at remote or relative rural areas. The station at Rørvik, close to Gothenburg in Sweden, showed a peak in TGM in the late 1980s. According to Lindberg et al. (2007), this peak is probably due to the close-down of regional sources in Eastern Europe, connected to the structural changes in the state administration in Eastern Europe, and not a global trend. If the station at Rørvik was excluded, no changes in TGM from late 1970s to 2002 would have been observed. Today, a discernible trend in the global atmospheric Hg concentration over the last 20-30 years is difficult to detect, due to the low number of monitoring stations and uncertainties in the monitoring data (Lindberg et al., 2007; UNEP, 2008).

Future emissions of Hg depend on several factors, such as economical development, improvement of emission reducing technology, regulatory changes, etc., making it challenging to develop scenarios. Several models have been applied for predicting emission scenarios (UNEP, 2008), and an overview of future trends is given in a report by UNEP/AMAP (2008).

Hg in the atmosphere is deposited to terrestrial and marine ecosystems. Various processes transfer Hg between and within the different compartments of the ecosystems (soil, water, sediments, air, and biota), resulting in complex biogeochemistry (Harris et al., 2007; Sigel and Sigel, 1997). In relation to aquatic ecosystems, which are the main focus in this report, factors affecting the cycling and subsequent bioaccumulation of Hg in the aquatic food web are discussed in the following chapters.

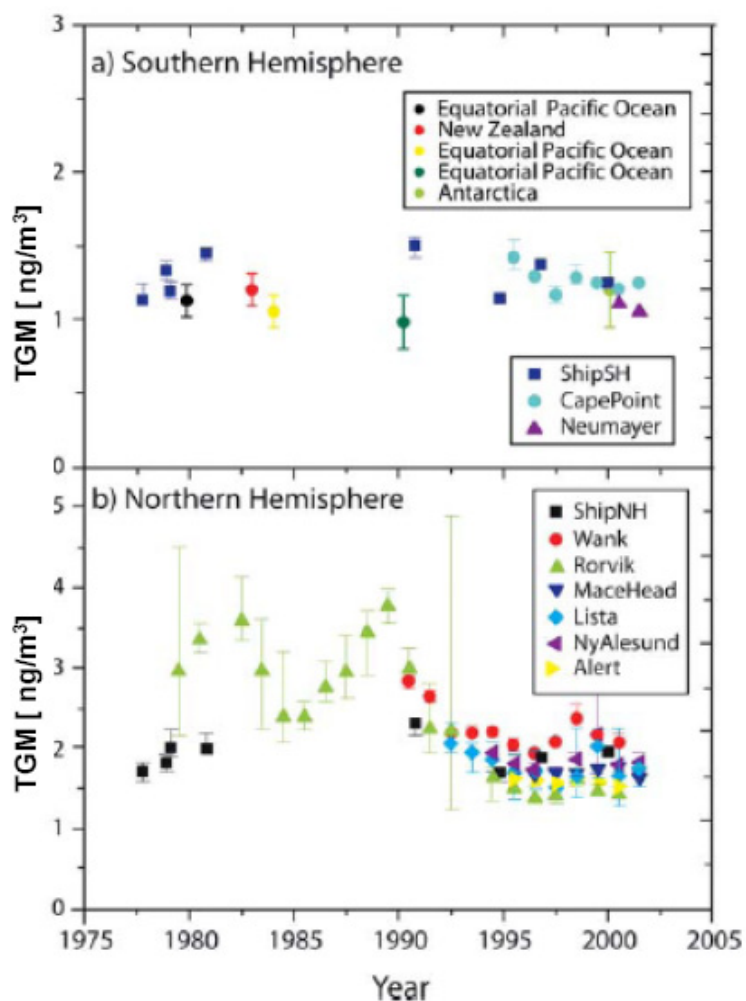


Figure 2. Trends in airborne TGM (Hg^0) measured at several land-based monitoring stations and ship cruises in the Southern and Northern Hemispheres. Legends:

- ShipNH/SH, ship cruises at Northern/Southern Hemispheres;
- Rörvik, South-Western Sweden;
- Wank, Southern Germany;
- Lista, Southern Norway;
- MaceHead, Western Ireland;
- Alert, Nuavut Canada;
- Ny Ålesund, Svalbard/Norway;
- Cape Point, South-Africa, and Neumayer, Antarctica.

From (Slemr et al., 2003).

4. Factors affecting mercury and methylmercury in aquatic ecosystems

Hg distributed via atmospheric processes is deposited as dry and wet deposition to terrestrial and aquatic ecosystems. Aquatic ecosystems in remote areas receive Hg deposition directly from the atmosphere, but transportation from terrestrial watersheds is generally believed to be the most significant Hg source. It is estimated that 5-25% of atmospheric deposited Hg in the watershed is transported to lakes from the corresponding catchment, and this is highly dependent on the lake surface to catchment area ratio (Mierle and Ingram, 1991). This implies that catchment processes play an important role for the distribution and accumulation of Hg and MeHg in organisms, sediments and waters of remote aquatic ecosystems. Mobilisation and transport of Hg and MeHg in catchment soils to lakes and rivers is controlled by catchment characteristics and processes which in turn are affected by climate, direct human perturbation such as forest management, road building, and other more or less natural events such as forest fires. In this chapter, we aim to give an overview of several factors influencing the fate of Hg in aquatic ecosystems.

4.1 Dissolved organic matter (DOM) and mobility of mercury

Solubility and mobility of metals are to a great extent determined by their partitioning between dissolved and solid phases. Several environmental factors affect this partitioning, but for Hg the interaction with dissolved organic matter (DOM) is pronounced. Hg binds to various functional groups in DOM, with thiol/sulphur and acidic functional groups as major complexation sites. The complexation with these thiol/sulphur-groups, through a covalent bond, is considered to be the main explanation for the strong binding of Hg to DOM (Ravichandran, 2004; Skyllberg et al., 2006).

The strong binding between Hg-DOM is believed to be one of the most important factors controlling the mobilisation of Hg in soil, and the speciation and bioavailability of Hg in aquatic ecosystems. This strong complex with DOM facilitates mobilisation of Hg from soils and sediments into aquatic ecosystems, increasing the Hg concentration in the water and sequestration of Hg to suspended sediments in lakes. Strong correlations between Hg and DOM concentrations are often found in surface waters. Although DOM is ubiquitous in the aquatic environment, special attention has been given to wetlands and peatlands, enriched with DOM. In these ecosystems Hg is sequestered through DOM, while anaerobic microbial processes facilitate the formation of MeHg. These ecosystems play an important role in the mobility of Hg, and are identified as a sink for total Hg, and a net source of MeHg. In this way, peatlands/wetlands function as storage sites, leaking substantial amounts of both Hg accumulated over decades and MeHg, to downstream aquatic ecosystems (Grigal, 2002; Larssen et al., 2008; Skyllberg et al., 2003).

4.2 Catchment characteristics

Watershed characteristics play an important role for the transport and fate of Hg in aquatic systems. In boreal catchments, input-output budgets suggest a high retention of deposited Hg in soil and vegetation (Grigal, 2002). As discussed in the previous paragraph, dissolved organic material is the main carrier of Hg and MeHg from soils to surface waters.

Deposition of atmospheric Hg is increased by the presence of forest, since foliage functions as a filter for the passing air, thereby increasing the surface area for dry deposition. Deposition of Hg in forests can be 2 to 3 times larger than wet deposition alone (St.Louis et al., 2001). Hg is collected through direct adsorption, assimilation of Hg⁰ by leaf stomata and through recycling. Litterfall and throughfall transport are the main inputs of Hg to the soil and only a minor part of soil Hg will eventually reach surface waters (St.Louis et al., 2001). However, a large part of the Hg is also likely to re-volatilise to the atmosphere without reaching the surface water

Because organic top soils, wetlands and forested areas have accumulated wet and dry atmospherically deposited Hg over a long period of time, the reduction in emissions seen in Europe the last 15 years have not yet had any profound impact on the concentrations observed in boreal lake ecosystems. This suggests that transport and bioaccumulation of the stored Hg can continue to pose a threat. Since deposition is higher than leaching, Hg is still accumulating and increasing levels of Hg are expected. For example, Johansson et al. (2001) estimated that the atmospheric deposition in Sweden had to be reduced by 80% to achieve a decrease in soil concentrations.

4.3 Disturbance by forest management and forest fire

There are strong indications of increased transport of Hg and MeHg to surface water in catchments affected by forest harvest operations, forest fires and similar disturbances.

Several studies comparing logged, burned and unaffected catchments have found that levels of MeHg in biota are significantly higher in logged and burned catchments (Garcia and Carignan, 2000; Garcia et al., 2007). Compared to reference lakes, higher concentrations of Hg were found in fish from lakes with partially burned watersheds (Kelly et al., 2006). In Lake Moab in Canada the concentrations of nutrients increased after parts of the watershed burned in 2000 compared to pre-fire conditions, stimulating the lake productivity. Increased productivity led to alterations of the ecosystem and caused enhanced accumulation of MeHg. For example, increased growth rates were shown to lead to an earlier shift to the piscivorous stage of lake trout (*Salvelinus namaycush*) (Kelly et al., 2006).

Several studies have shown increased leaching of especially MeHg after forest harvest (Desrosiers et al., 2006; Porvari et al., 2003) and after physical disturbance of soils through use of heavy machinery (Munthe and Hultberg, 2004). Higher concentrations of MeHg were also seen after a large storm uprooted large areas of forest in southern Sweden (Munthe et al., 2007a). The severity of the response to the disturbance depends on the forest management methods used and how much of the catchment area that is affected (Porvari et al., 2003). Effects of clear-cutting may last up to several years after the disturbance took place (Munthe et al., 2007a).

The mechanisms behind how forest management and wildfires affect the export of Hg and MeHg from soils to surface waters are uncertain, and several explanations have been proposed. For example, that the removal of vegetation causes a substantial reduction in evapotranspiration, and thus may lead to a heightening of the water table in both logged and burned catchments (Munthe et al., 2007a; Porvari et al., 2003). This will change the local hydrological regime and increase erosion and transport of coloured water to lakes (Porvari et al., 2003). Additionally, the production of MeHg in soil may increase, especially in clear-cut catchments because rising water levels and soil compaction may induce anoxic conditions favourable for methylating bacteria (Munthe et al., 2007a; Porvari et al., 2003).

5. Results from monitoring of Hg in the aquatic ecosystems

Over the last few decades, substantial progress has been made in our understanding of Hg in aquatic environments. This includes analytical procedures for measuring of Hg and MeHg in various matrixes, insights into cycling and bioaccumulation of Hg and MeHg and the sources of Hg. Together these advances provide a solid basis for the implementation of monitoring programmes for Hg in aquatic ecosystems. This chapter presents an overview of results from monitoring programmes from the UNECE region concerning the fate of long-range transported Hg in aquatic ecosystems, including surface waters, sediment, and fish.

5.1 Mercury in surface waters

Monitoring of Hg in surface waters provides important information on levels of Hg in areas receiving various loadings of atmospheric Hg with the possibility to detect regional patterns and temporal trends. Aqueous concentrations of Hg and MeHg are affected by deposition, catchment- and in-lake processes. Frequently, discussion of Hg in waters is limited to total Hg (HgT) which measures all forms of mercury lumped together even though HgT alone is a limited indicator of the environmental risk of Hg in the environment.

In lakes and watersheds not directly influenced by anthropogenic sources, HgT concentrations are often in the range of 0.3–8 ng L⁻¹ (Krabbenhoft et al., 2007). In pristine lakes high in natural organic matter (NOM), considerably higher HgT concentrations are found. Since Hg (II) readily forms stable complexes with NOM, its presence in humic lakes may be in the range 20–40 ng L⁻¹. In streams and lakes influenced by mining or geological sources, HgT concentrations in the range 2500–12 000 ng L⁻¹ have been observed (Ganguli et al., 2000; Gray et al., 2000).

Tributaries to Lake Michigan influenced by urban and agricultural land use have been shown to have elevated HgT concentrations (particulate and filterable) compared to tributaries coming from rural and forested areas (Krabbenhoft et al., 2007). In most samples of this study, Hg was found in the particulate phase, presumably bound to natural organic matter (NOM).

Long-term studies that show spatial and temporal variations in Hg concentrations in surface waters are few and most often only results from HgT are reported. In a recently published paper by Eklof et al., (2009), concentrations of HgT from 19 Swedish streams monitored since 2000 were presented. Only one stream showed an upward trend in HgT concentrations, in all other streams no such trend was observed. In a thirteen year study from Sweden (Lake Gårdsjön) Hg was monitored before, during, and after construction of a roof covering a whole catchment (Munthe and Hultberg, 2004). No upward or downward trends in HgT concentrations and HgT fluxes were observed. A similar lack of trends were found in the ongoing METALLICUS-project in Canada/USA (Krabbenhoft et al., 2007). In the remote Experimental Lake Area (ELA) in the boreal ecoregion of north west Ontario, Hg has been monitored since the early 1990s. Until 2001, no long-term trends were observed (St.Louis et al., 2004).

From Lochnagar in Scotland monthly monitoring data from end of 1997 to 2007 showed that Hg in lake water declined over this period. Mean Hg concentration for the first full year of monitoring in 1998 was 14.1 ng L^{-1} , 6.4 times higher than that for the final full year of monitoring (2.2 ng L^{-1}) in 2007 (Rose and Yang, 2009 (in prep.))

Since complex processes control the occurrence, formation, distribution, and redistribution of HgT and MeHg, it is difficult to detect and quantify changes attributed to alterations in the deposition. According to Driscoll et al. (2007), multi-year studies are needed in order to distinguish changes in Hg loadings and real trends in surface waters. In the long-term Hg monitoring project ACME from the Everglades in Florida (US) data series are 8 years long. Results from this project showed that although HgT concentrations in water varied considerably, a significant (95% confidence) declining trend was observed, from 1.9 ng L^{-1} to 1.2 ng L^{-1} . In the same period, Hg concentrations of piscivorous birds' feathers and largemouth bass (*Micropterus salmoides*) were reduced in this area. According to the authors, these reductions were linked to declines in the atmospheric deposition (Krabbenhof et al., 2007).

In the METALLICUS-project, Hg was added experimentally to selected lakes. In Lake 658, a doubling of the Hg load to the surface water nearly doubled the concentration in the lake. This observation was repeated in 2002 and 2003, and shows that small lakes may respond quickly to increased Hg loadings (Krabbenhof et al., 2007).

Methylmercury comprises 0.1- 5% (Krabbenhof et al., 2007) to 0.5-30% (Meili, 1997) of total Hg in surface water, is the most toxic and bioaccumulative form of Hg. The concentration levels of MeHg depend on methylation, demethylation, and formation of complexes, stabilising MeHg. Concentrations appear to vary locally and regionally, but factors like high temperature and content of organic matter have shown to support the formation and stabilisation of MeHg (Meili, 1997). Long-term records of MeHg monitoring are limited, and early results may suffer from analytical obstacles related to high detection limits compared to today's methods.

Under the WFD, the environmental quality standard (EQS) for Hg is set at $0.05 \mu\text{l}$, and differentiation between different species of Hg – total, dissolved or MeHg is not carried out. Given the overview above, most surface waters in watersheds without direct influences of run-off from mining and other major point sources are likely to achieve this EQS. This appears to suggest that no environmental problems are expected within these concentration ranges. However, many of the lakes where aqueous concentrations of Hg and MeHg have been measured were selected because concentrations in fish in these lakes had reached levels well above thresholds recommended for human consumption. Thus, the concern with regard to Hg pollution and human and wildlife exposure to Hg is not addressed satisfactorily with the current EQS established for the WFD.

5.2 Mercury in lake sediments

Lake sediments are considered to be an important part of the Hg cycle. In aquatic ecosystems, sediments function as a sink and a source for Hg. Processes in the lake and its catchment form

organic Hg-complexes that precipitate in the lake sediment where it may be trapped or/and transformed into bioavailable species. As sediments serve as the ultimate repository for much of the particulate matter that moves through the watersheds, sediments are well studied environmental matrixes.

One of the most importance pathways in the Hg cycle is the transformation of Hg into MeHg, the highly toxic and bioaccumulative Hg specie. In anoxic sediments Hg can be converted to MeHg via a mechanism which is not thoroughly understood, but believed to be mediated by sulphate-reducing bacteria (Pak and Bartha, 1998; Skyllberg et al., 2007). An important aspect of this process appears to be NOM, acting as a transport medium of Hg, promoting sedimentation of Hg and the formation of anaerobic conditions in the sediments, and finally acting as an energy source for sulphate-reducing bacteria. All these factors may stimulate the methylation process (Pak and Bartha, 1998; Skyllberg et al., 2007). As a consequence, lake sediments are considered to be an important source of MeHg, which it is partitioned between organisms, the aquatic phase, and sediments (Jackson, 1997). According to (Munthe et al., 2007b) the highest amount of Hg in piscivorous fish in Fennoscandia has generally been found in areas where the concentration of Hg in surface sediments is high.

5.2.1 Sediments are archives for mercury deposition

Hg deposited from the atmosphere to lake surfaces and in the run-off water from catchments is scavenged by particles and deposited to the lake sediments, where it is retained and isolated from the biogeochemical cycle. This makes it possible to use deep lake sediments as a proxy for spatial and temporal trends in Hg deposition. Since a network of Hg monitoring stations is missing, core samples of sediments are frequently used as archives for Hg deposition (Munthe et al., 2007b). The use of cores for construction of historical deposition is complicated, since several factors may influence the sedimentation process. In addition, Hg is abundant in minerals, and weathering of rocks may release Hg to the sediment, indicating that results should be interpreted with caution. Several studies include both surface and deep core sediments, where the deep core contains deposition from times before the onset of the Industrial Revolution. The ratio of Hg between surface sediments to deep cores, not influenced by industrial deposits is often interpreted as the increase in Hg deposition. This ratio is frequently referred to as the enrichment factors (EF), where $EF > 1$ indicates influence by external sources. In order to detect trends in Hg inputs to a lake, the sediment cores have to be dated. Radiometric dating is used to estimate the sedimentation accumulation rate (fluxes) and the age of the various depths (Curtis et al., 2008; Jackson, 1997). It has been shown that natural organic matter (NOM) is probably the most important carrier of Hg to the sediments; in the lake itself and from the catchment to the lake. As a result, Hg concentrations in the sediment cores are strongly correlated to the content of organic matter through the core profile. In cores where changes in Hg deposition are adjusted for the content of organic matter, the deposition history is more correctly depicted (Munthe et al., 2007b).

5.2.2 Regional surveys of mercury in lake sediments

A considerable number of papers document $EF > 1$ for Hg in sediments cores from lakes in the Northern Hemisphere, which have not been influenced by local point sources. In general, the cores reveal an increase in Hg content from past to present, with a particular increase after the onset of the Industrial Revolution, and a peak in the late 20th century. The last 10-15 years the Hg content in most lake sediments has gradually declined, probably due to reduced Hg-emissions. In the Arctic, some lakes do not show this recent decline in mercury concentration.

High Hg concentrations found in sediments of remote and pristine headwater lakes demonstrate the importance of long-range atmospheric transport. Deposition of Hg is influenced by meteorological phenomena, causing temporal and spatial variations within different countries, at remote and as well urban areas (Jackson, 1997; Lindberg et al., 2007).

5.2.3 Scandinavia

In Scandinavia, atmospheric Hg deposition originates primarily from sources in Central Europe. A considerable number of deep sediments cores have been collected in Scandinavia (Jackson, 1997; Munthe et al., 2007; Munthe et al., 2007b). In the work by Munthe et al. (1995), classic Hg profiles describing the well-known rise in Hg after the start of the Industrial Revolution are depicted (**Figure 3**). These results are of great interest due to: i) the sharp increase in Hg loading from the atmosphere over time, followed by a sudden and dramatic reduction, starting at some time in the recent past and continuing to the time of the sample collection; and ii) the similarity in shape of all Hg profiles with a peak at practically the same depth, although they represent different lake environments. Unfortunately, radiometric dating by use of ^{210}Pb and ^{137}Cs was not performed on these cores making it impossible to date the samples.

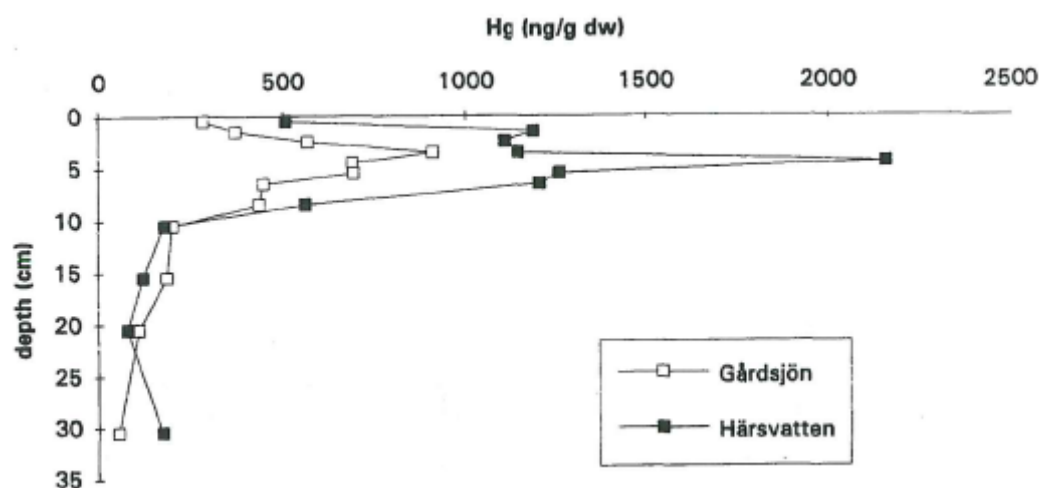


Figure 3. Hg profiles in sediment cores from two Swedish lakes (Gårdsjön and Härsvatten, (Munthe et al., 1995).

Mercury concentrations in dated sediments cores from Scandinavia show that there has been an increase from the 1800's to the 1960's, followed by a decrease to the early 2000's (Bindler et al., 2001; Fjeld and Rognerud, 2001; Kaste et al., 2001; Mannio, 2001; Munthe et al., 1995).

A considerable amount of sediment core data referred to in a review by Jackson (1997) showed the existence of a spatial gradient in Scandinavia, with high Hg levels in the south and low levels in the north, ascribed to long-range atmospheric transport of anthropogenic Hg. This has also been confirmed by Munthe and co-workers (2007b). In that study, 389 lake surface sediments and 210 subsurface sediments from Fennoscandia were analysed for Hg. A scatter-map showing the Hg concentrations in the sediments is shown in **Figure 4**.

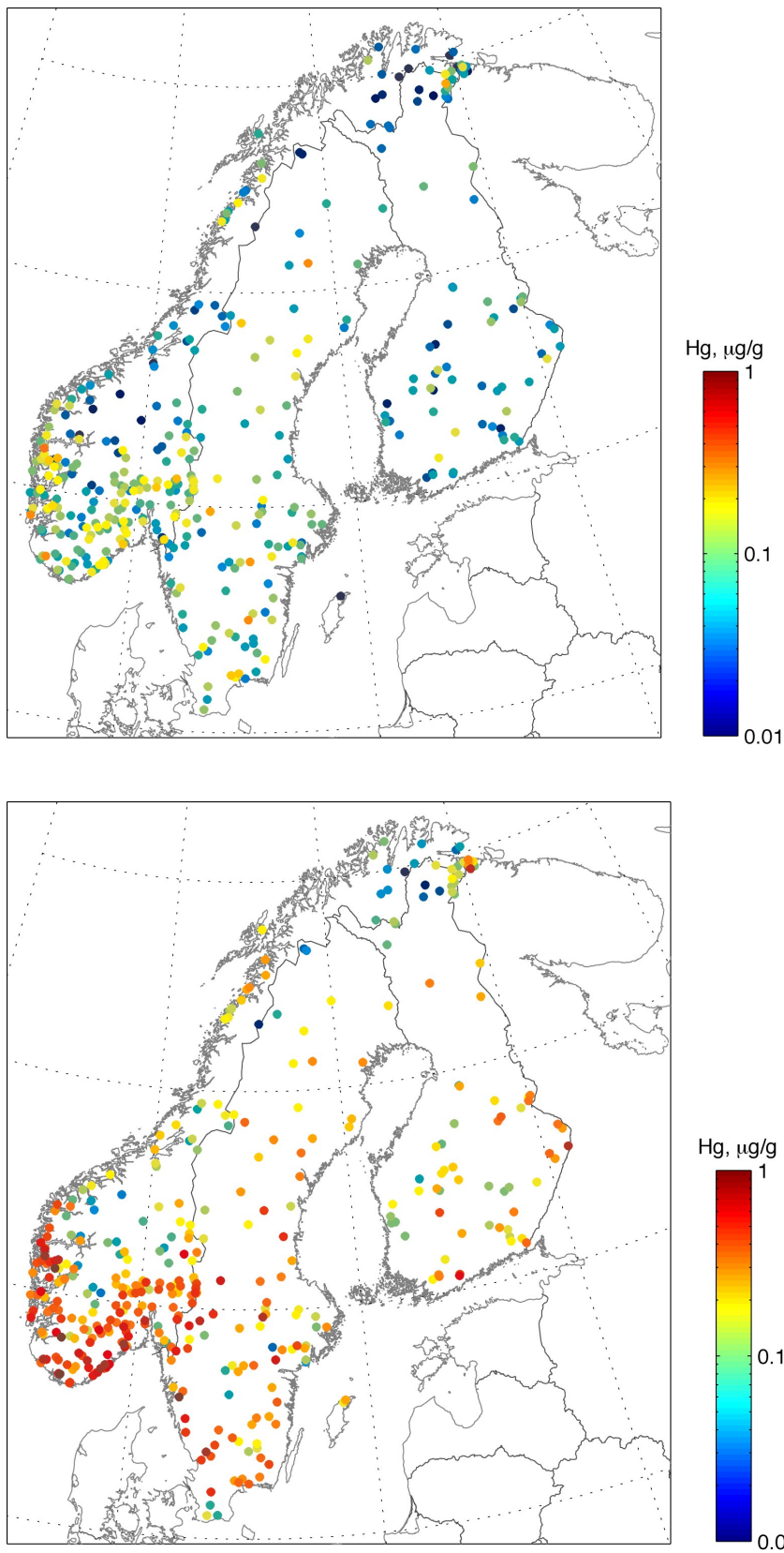


Figure 4. Hg concentration (mg kg^{-1} dry weight, dw) in reference sediments (top) and surface sediments (bottom) from Fennoscandia. The Norwegian, Finnish, and Swedish sampling were performed 1996-1997 (Rognerud and Fjeld, 2001), 2002-2003 (Mannio, 2001), and 1998-2002, respectively. Figure is taken from (Munthe et al., 2007).

Surface sediments from this lake survey showed elevated concentrations of Hg compared to the reference sediments (median EF = 2.8), which was most likely caused by long-range transported anthropogenic atmospheric deposition. In general, the concentration of Hg was highest in the surface sediments in coastal areas in Southern Norway, Southwest/Central Sweden, and Eastern Finland. These concentrations were above $0.3 \mu\text{g Hg g}^{-1} \text{ dw}$. Low concentrations ($< 0.15 \mu\text{g Hg g}^{-1} \text{ dw}$) were found in mountain areas in Southern Norway, and in Central, and Northern Scandinavia. Concentrations of Hg in lake and catchments situated close to each other showed occasionally large variation. This was indicated as a result of the local lake and catchment capability to modify the influence of atmospheric deposited Hg. From the organic matter adjusted data (not shown) it appeared that Hg was closely associated to the organic sediment fraction, which, together with water quality and morphology of the aquatic system (e.g. depth of the lake) influences the spatial distribution of Hg in sediments. This is consistent with other studies (references in (Munthe et al., 2007)) showing the importance of NOM as a carrier of Hg in ecosystems.

A Norwegian national lake survey conducted in 2004-2006 included measurements of several heavy metals, PCB, and PAH in 274 surfaces, sub-surfaces, and pre industrial time (deep core) sediments. In this survey, surface sediments showed considerably higher Hg values than the pre-industrial sediments, and the highest concentrations were observed along the coast and in southern Norway. Coastal areas in northern Norway also revealed elevated levels of Hg, while inland and mountainous area had the lowest values. Here, differences between sub-surface and surface sediments were minor, but indicated that there had been a reduction in the Hg deposition in the south and an increase in the north the last 15-20 years. Median enrichment factors for Hg in these sediments were 2.6 (Rognerud et al., 2008). Similar results for the northern Norway were found by Christensen et al. (2008). In this survey, part of the the region north of the Polar circle reporting to the Arctic Monitoring and Assessment Programme (AMAP), was examined. Sediment samples from 123 lakes were taken during 2004-2006 and analysed for Hg and several inorganic and organic pollutants. Highest Hg values were observed in surface sediments along the coast and in the eastern part of the county Finnmark (Sør-Varanger). A recent increase in Hg deposition was also observed in this area (measured as the difference between the sub-surface and surface sediment). The median values for all the lakes were $0.15 \mu\text{g Hg g}^{-1} \text{ dw}$, while the mean concentrations from the mainland and the Spitsbergen archipelago in the north, were $0.16 \mu\text{g Hg g}^{-1} \text{ dw}$ and $0.13 \mu\text{g Hg g}^{-1} \text{ dw}$, respectively.

5.2.4 The Arctic

The Arctic Monitoring and Assessment programme (AMAP) was established 1991 to monitor identified pollution risks and their impacts on Arctic ecosystems. An extensive report on Hg in the Arctic has been published by AMAP in 2005 (AMAP, 2005). Results from AMAP shows that the Arctic is closely connected to the rest of the world, that contamination from long range sources are deposited in the Arctic.

AMAP (2005) draws the following conclusions:

- EFs are generally 1.5 to 3.0 throughout the Arctic, including northern Scandinavia, Siberia, Arctic Canada, and South-Western Greenland. This indicates that the sediments are influenced by long-range transported anthropogenic Hg.

- In Eurasia, elevated Hg-levels in lake sediments are generally found in regions with industrial/urban emissions.
- In the Murmansk and Kola region, a mixture of long-ranged transported Hg and local emission to lake sediments was observed.
- Arctic lakes along the Siberian coast had considerable higher concentrations of Hg in surface sediments than in pre-industrial sediments. The differences between the two layers decreased with increasing longitude to the east. This indicates that there is a recent Arctic-wide increase in Hg in lake sediments.
- A transect of lakes from the coastal inland to the Greenland ice sheet, found in general a three-fold increase in EF in coastal lakes and a two-fold increase in inland lakes. The highest EF-values (4 to 11) were observed in lakes adjacent to the inland ice sheet and within the ice sheet.
- Lake sediments in Canadian Arctic and sub-arctic showed considerably higher levels than pre-industrial times. In several of these southern Arctic lakes, the traditional decline in recent Hg deposition was not revealed, in contrast to observations done at lower latitudes. This may indicate that recent Hg deposition has not declined in southern Arctic regions.
- In the North Alaskan coastal plain and the Ellesmere Island, concentrations of Hg in upper surface sediments were similar to those of pre-industrial times, which suggest an absence of anthropogenic Hg in these regions.

5.2.5 Mountain lakes in Europe

In the EU-project EMERGE (<http://www.mountain-lakes.org>), 275 sediment samples were collected from remote lakes in mountain areas across Europe (including Greenland). Surface and pre-industrial sediments were sampled from these lakes. Results showed that the overall EF for mercury from the survey was 1.6, and median mercury concentration in the surface sediments was $0.20 \mu\text{g g}^{-1}$ (dw). Among the sites studied, the highest EF was found in the Tatras (Poland/Slovak) and Rila Mountains (Bulgaria), 2.6 and 3.0 respectively (Lluís Camarero, Spain, in preparation).

5.2.6 North America

Late in 2000, a group of scientists formed the Northeastern Ecosystem Research Cooperative (NERC, www.ecostudies.org/nerc/), in order to promote collaboration among scientists in north-eastern US and Canada. A compilation of Hg data in lake sediments in Northeast America was carried out under the NERC umbrella. The data compilation showed heterogeneous levels of Hg across the north-eastern North America, dictated by topography, hydrology, biogeochemistry, land-use, and point source impacts (Evers and Clair, 2005).

Based on the NERC database, an extensive survey of Hg in the surface sediments of lakes, rivers and reservoirs of North-eastern North America (US-Canada border) was performed by Kamman et al. (2005a). From 579 reported sites the Hg concentration ranged from 0.01 (limit of detection) to $3.7 \mu\text{g g}^{-1}$ (dw), and the overall average concentration was $0.19 \mu\text{g g}^{-1}$ (dw). In this survey MeHg was also analysed, which is frequently missing in other reports. Sediment MeHg ranged from 0.15-21 ng g^{-1} (dw), and the mean concentration was 3.83ng g^{-1} (dw). Their data indicate that 44% of waters across the region have total HgT concentration in excess of guidelines for the protection of aquatic biota.

Surface sediments across the entire Great Lakes basin were collected by Marvin and co-workers in 2004 (Marvin et al., 2004). In general, Lakes Huron, Superior, and Michigan exhibited low levels of Hg in the sediments, while higher values were found in the lakes St. Clair, Ontario, and the western basin of Lake Erie. A spatial distribution in contamination in the area was observed, with decreasing concentration from the east to the west. Considerable point sources of Hg appeared to be the Detroit area, influencing the western basin of Lake Erie and waste water previously discharged into Lake Ontario. Average concentrations in deep lake sediments varied from 0.043-0.585 $\mu\text{g g}^{-1}(\text{dw})$. The elevated levels of Hg in the sediments were ascribed to local point sources and global influences

In a study by Bookman et al. (2008), sediments from urban/suburban lakes in New York, US were presented. Local, regional, and long-ranged emissions were ascribed as the source for the elevated Hg in the sediments. Hg fluxes peaked during 1971-2001, and were three to more than 30 times greater than pre-industrial deposition. The contribution of local and regional emission sources to central New York lakes was estimated to about 80% of the total atmospheric Hg deposition, while 20% was ascribed to long-ranged atmospheric deposition. This shows that long-range pollution has a considerable impact on urban areas as well.

On the North American west coast, long-range pollution sources were responsible for most of the Hg found in the upper sediments. Results indicated that enrichment in the sediments had started before local municipal and industrial sources could have influenced the sediments. In the lakes studied, global sources of Hg were primarily responsible for the increase in the lakes, starting about 1900 (Paulson and Norton, 2008).

Prairie lake sediments in central Alberta, Canada, revealed that local coal-fired electricity plants were the main sources of Hg. Dated cores showed that the Hg emissions started after the construction and operation of the plants. Since 1950 the levels of Hg in the sediments have increased 1.2 to 4-fold (Donahue et al., 2006).

Lake sediments from Arctic Alaska indicated that anthropogenic Hg impacts in the Arctic are of similar magnitude as in temperate latitudes. A threefold increase in atmospheric Hg deposition since the pre-industrial times was revealed ($\text{EF} > 3$) (Fitzgerald et al., 2005). In the WACAP project, 8 national parks on the West coast of the USA were examined from 2002 through 2007 for a considerable number of contaminants in multiple ecosystem compartments (Landers et al., 2008). The temporal records from sediment cores indicated that in nearly all parks, Hg deposition increased in the twentieth century because of anthropogenic sources. Both declines and increases in the fluxes of mercury were observed in the most recent sediments from the parks.

5.2.7 Russia

Considerable numbers of remote sites in Russia have been studied in the AMAP project. Results from the AMAP indicated that sediments in Arctic lakes along the Siberian coastline had elevated levels of contemporary long-ranged transported Hg, while in the area Kola/Murmansk the elevated Hg was described as a mixture of recently long-range transported and local sources. The differences in Hg between surface to pre-industrial sediments decreased with latitude and increasing longitude to the east (AMAP, 2005).

Smelters in the Noril'sk region along the Siberian coast are among the worlds' largest emitters of sulphur dioxide (SO₂). Several authors have suggested that a substantial proportion of air pollutants reaching the Arctic can be linked to this area. Mercury EFs in this area ranged from 1.39-3.64, and there was no significant decline in Hg concentration with distance from the smelters. Levels of Hg in the Noril'sk area are clearly elevated, but EFs are comparable to other values found in northern areas (Blais et al., 1999).

5.2.8 United Kingdom

Six lake sediment cores were analysed in order to study the temporal distribution of Hg across the UK. All sites had been contaminated with Hg, and there had been a rapid increase in the last 200 years at all rural sites. Peak concentrations ranged from 162 to 1602 ng g⁻¹ and EFs from 2.2 to 26.7. All cores except for one fell into the upper range for European rural areas (Yang and Rose, 2003), showing a relatively high degree of pollution in the UK. An additional 10 sites across the UK were analysed by Yang and Rose (2005). In this study, peak concentrations for sites only receiving atmospheric inputs of Hg ranged from 165-460 ng/g, while peak fluxes ranged from 11-55 ng cm⁻² yr⁻¹.

5.2.9 Summary: Hg in lake sediments

Lake sediments are an important part of the Hg cycle, functioning both as a sink and a source for Hg. Deposition of Hg on the lake surface or through catchment processes form organic complexes that precipitate in the lake sediment where it may be trapped or/and transformed by microbial processes into the bioavailable and highly toxic specie, MeHg. This process that occurs in the sediments and in anoxic sites in the catchment is mediated through sulphate-reducing bacteria. Most of the Hg deposited in sediments is retained and isolated from the biogeochemical cycle, and as a result, deep lake sediments are used as a proxy for spatial and temporal trends in Hg deposition. Core samples of sediments are used as archives for Hg deposition, and the ratio of Hg between surfaces to deep cores not influenced by industrial deposits is referred to as the enrichment factor (EF), where an EF > 1 indicates influence by external sources. An overview EFs from different surveys in Europe, North America, and Arctic is given in **Table 1**.

Table 1. Summary of concentration ranges and enrichment factors (EF) of Hg in lake sediments from surveys in Europe, North America, and the Arctic.

	No of lakes	Surface sediments range ($\mu\text{g g}^{-1}$)	EF	Reference
Scandinavia	389		1.3-6.1 (2.8)	(Munthe et al., 2007b), 10 th - 90 th percentile, median is given in bracket
Arctic			1.5-3.0	AMAP (2005)
Finland	9		1.2-8.9	(Verta et al., 1989)
Alaskan coastal lakes	15		1.8-2.1	(Engstrom and Swain, 1997)
Yukon and Northwest Territories in Canada	18		1.1-7	(Lockhart et al., 1995)
Canada (Quebec)			~2.3	(Lucotte et al., 1995)
Alberta (Canada)	3		1.5-7	Donahue et al. (2006)
UK	6		2.2-26.7	Yang and Rose (2003)
Northeast North America	478	< 0.01-3.7		Kamman et al. (2005b)
Western US, Whatcom County	6	0.130-0.229		Paulson and Norton (2008)
New York (US)	4		3-30	Bookman et al. (2008)
Northern Sweden	12		~ 2	(Lindeberg et al., 2007)
Great Lakes	6	0.043-0.59		Marvin et al. (2004)
Russia (Norilsk)	10		1.39-4.65	(Blais et al., 1999)

Summarizing all of the surveys covered in this chapter from temperate, boreal and arctic regions, EFs of 1.1 to 30 are reported, but the most common values are roughly between 1.3 and 6. Large variability in Hg enrichment is found even between lakes that are located with the same region, probably related to catchment characteristics and variations in meteorology. In the Arctic, lakes situated close to emission sources can show high EFs whilst lakes without any apparent Hg enrichment are also found in more remote areas. In Scandinavia, a strong north-south gradient is seen where lowest enrichment is found furthest north. This pattern appears to be controlled by atmospheric Hg deposition. In North America, a similar gradient but from west to east is visible with highest lake sediment enrichment in northeast USA and southeast Canada. Also here, this pattern seems to be controlled by atmospheric Hg deposition. However, data from the Great Lakes and from urbanized areas indicate that sediment Hg levels are affected by a combination of local emission sources and long-range transported Hg pollution. In Russia, lakes with the highest EFs are located in a region with a high activity of smelters. In the UK, some lakes with very high EFs were found which could suggest a substantially higher Hg deposition than in Scandinavia. Also in remote alpine lakes in Europe, enrichment of Hg in sediments was found.

5.3 Mercury in fish

Aquatic organisms at all trophic levels from algae, zooplankton, benthic invertebrates to fish and wildlife are all potentially affected by Hg. Little is known about the mechanisms that control and influence the Hg content in organisms at lower trophic level, but the uptake and transfer through the food web is well documented (Wiener et al., 2007). Data on Hg from organisms at the base of the food web are still limited today, but considerable work on fish and other wildlife has been performed. Historical records on piscivorous fish from North America date back to early 1970's, while in Scandinavia surveys were started in the 1960's. The considerable amount of work carried out on fish and Hg has been driven by concerns for human health associated with consumption of Hg contaminated fish. Very few, if any, regional datasets exist for other aquatic organisms. In this chapter we present results from surveys on Hg in fish from Europe, North America and the Arctic.

Environmental problems with Hg in aquatic ecosystems are related to MeHg, a compound that readily accumulates in biota, and biomagnifies to high levels in organisms at the top of the food web, and can cause neurological damage to organs. About 95% or more of the Hg found in fish is in the form of MeHg and is obtained almost entirely from dietary uptake.

The entry of MeHg into the base of the food web is not well understood. It is believed that most of the Hg is transformed into MeHg through microbial processes. Sulphate-reducing bacteria, preferring anoxic conditions are thought to be responsible for this transformation. As a result, elevated levels of MeHg are frequently found in anaerobic locations such as lake-bottom sediments and water logged areas in catchments (Pak and Bartha, 1998).

MeHg binds strongly to protein and peptides, especially those enriched with S-groups, and organisms will absorb MeHg at a rate greater than it is lost from the organisms, causing bioaccumulation. Through the food web the MeHg is biomagnified, and the highest levels are thereby found in organisms at the top of the food web. This occurs across trophic levels, and the pattern is similar among aquatic systems that differ in water type, Hg source, and pollution intensity. The greatest increase in MeHg concentration occurs in the trophic step between water and algae/ seston, with bioaccumulation factors in the range 10^5 to 10^6 , whereas ratios of MeHg concentration between successive trophic levels above algae are generally lower than 10^1 (Wiener et al., 2007).

The concentration of MeHg and the fraction of total Hg present as MeHg usually amplifies in fish with increasing trophic level and age (size). But when growth rates are high, this relationship may diminish, due to biomass dilution (Campbell et al., 2006; Greenfield et al., 2000; Sharma et al., 2008). At lower trophic levels, such as invertebrates, the MeHg fraction can constitute from 10 to 90% of the total Hg fraction. Due to the tight bond between S-enriched proteins and MeHg, considerable amounts of MeHg are found in the muscle tissue/proteins (Wiener et al., 2007). In **Figure 4** the correlation between Hg content in fish muscle and fish length in perch (*Perca fluviatilis*), pike (*Esox lucidus*), and brown trout (*Salmo trutta*) is shown (Fjeld and Rognerud, 2004). Other factors like top trophic position in the food web and inefficient growth (energy is spent on maintenance) may enhance the concentration of Hg in fish (Gantner et al., 2009; Jenssen et al., 2009; Rognerud et al., 2002b).

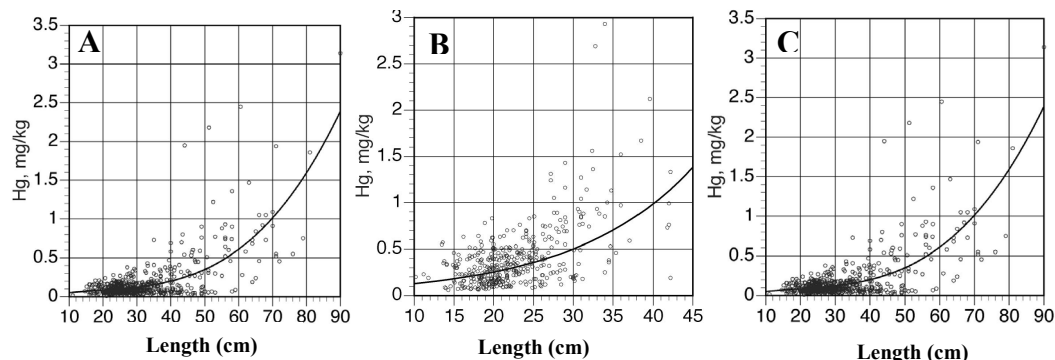


Figure 4. The relation between Hg concentration in muscle tissue and fish length in Northern pike **A** (*Esox lucius*), European perch **B** (*Perca fluviatilis*), and brown trout **C** (*Salmo trutta*), (Fjeld and Rognerud, 2004).

5.3.1 Normalisation of Hg in fish for comparison across species and trophic level

Although monitoring of Hg in fish in some countries has a tradition of several decades, no internationally agreed methodology and reporting standard exist. Analytical procedures for measurement of Hg and MeHg have been developed and detection limits for Hg and MeHg have decreased over time. This poses some challenges for establishing base line concentrations and comparison of results over time and between data sets.

Additionally, analytical procedures for measurement of MeHg are complicated and expensive compared to analysis of HgT. As a result, HgT is more frequently analysed and reported than MeHg. Since it is apparent that MeHg constitutes more than 95% of total Hg in fish, analysis of HgT can be used as a substitute for MeHg analysis. Analyses of Hg in fish are usually performed on muscle tissue since this part is usually consumed and fish muscle contains high proportions of MeHg, also compared to fish liver.

As can be seen from **Figure 4**, the amount of Hg in fish is highly dependent on fish species and length. This makes it necessary to have data on length, weight or age on individual fish in order to compare and interpret trends in Hg concentrations. In order to normalise data, a 1 kg pike (*Esox lucius*) has been widely used as a standard organism (Fjeld and Rognerud, 2004). Problems with normalising data may occur at high growth rates, since deviation from the “Hg concentration and length (age/weight) curve” may occur due to biomass dilution (Campbell et al., 2006; Greenfield et al., 2000; Sharma et al., 2008). As a result, normalising data should only be performed when growth curves of the organisms are known.

In order to decide whether differences in Hg concentration in the same species from different lakes are due to differences in Hg pollution between lakes or differences in the trophic position of the sampled fish, stable-isotope analyses of nitrogen in the muscle tissue is needed. Determination of the relationship between $^{14}\text{N}/^{15}\text{N}$ ($\delta^{15}\text{N}$) gives the sampled fish an isotope signature, which is correlated to its position in the food web. The stable isotope signature $\delta^{15}\text{N}$ increases with 3.4‰ at each trophic level resulting in the highest $\delta^{15}\text{N}$ -values at the top of the food web. Application of $\delta^{15}\text{N}$ gives valuable time-integrated information with respect to feeding strategy and the influence of trophic position in relation to Hg concentration (Fjeld and Rognerud, 2004; Rognerud et al., 2002b; Rosseland et al., 2003;

Wiener et al., 2007). Unfortunately, stable isotope analyses for determination of trophic level are often missing in analyses of Hg in fish.

In the following overview, data on Hg and MeHg in fish are grouped according to region. Wherever possible, normalised numbers for each fish species with regard to fish size and Latin name are given. Where no standardisation was carried out, absolute concentration ranges of Hg and MeHg in g/dw or in g/ww are given.

5.3.2 Scandinavia

Hg concentrations in Nordic freshwater fish have been monitored regularly since the 1960s. Compiled results from surveys are registered in national databases, but there is also a considerable amount of scattered data not included in these databases. Results from these studies revealed that Hg is particularly elevated in several predatory fish species, and that levels of Hg in fish are not necessarily linked to elevated concentrations of Hg in the watershed.

Recently published reports from Scandinavia have shown that Hg content in fish increases despite the decline in deposition of Hg. In Sweden, a decline in Hg content in fish was observed at the end of the 1980ties, which was ascribed to the reduced emissions caused by the closure of factories in the former east block countries. During the 1990s the Hg levels in fish stabilised, while an increase has been observed this decade. According to the authors, the concentrations of Hg in fish have increased with a few percent each year (Åkerblom and Johanson, 2008). Similar work has been carried out in Norway (Fjeld and Rognerud, 2009). In this study, the Hg content in perch ($n = 565$) from 28 lakes caught in 1991 and 2008 was compared, and results showed that the Hg concentrations had increased with 63 %. The mechanisms behind these observations are not understood. Several explanations have been suggested, such as climate change with elevated temperatures and longer growing seasons, increased runoff of NOM-Hg, and in some areas changes in forest/silviculture practices.

In a report by Munthe et al., (2007b), covering 2,758 lakes in Fennoscandia, Hg content in fish was examined (**Figure 6**). Data compiled in this study were generated between 1965 and 2004. Due to considerable variability, standardisation of the monitored data was required. The following points were considered:

- Lakes with known point sources of Hg were not included
- Samples of fish fillet (muscle tissue) or whole fish were used
- Outliers from the weight-length/ Hg–size relationship were discarded
- Small perch (< 25 g) were excluded, since they were overrepresented
- Lakes with less than three individuals were rejected
- Hg concentrations were related to ww, or dw converted to ww
- MeHg results were converted to HgT (98% of total Hg in fish was regarded as MeHg)
- Analytical procedures for measurement of Hg were checked (but no results were rejected on this basis)

In this survey, six fish species were analysed. Northern pike (*Esox lucius*) had the highest Hg concentration (arithmetic mean 0.73 mg kg^{-1}), followed by perch (*Perca fluviatilis*) (0.40 mg kg^{-1}); trout (*Salmo trutta*) (0.13 mg kg^{-1}); whitefish (*Coreonus lavaretus/Coreonus albula*) (0.12 mg kg^{-1}), and charr (*Salvelinus alpinus*) (0.11 mg kg^{-1}). For a “standard fish” (1-

kg pike, 0.3-kg perch, 3.2-kg trout or 1.4-kg charr), the median Hg concentration was 0.69 mg kg^{-1} . The highest concentration of Hg for a standard fish was found in Sweden (median 0.71 mg kg^{-1}), followed by Finland (0.51 mg kg^{-1}), and Norway (0.34 mg kg^{-1}). By latitude, the highest concentrations were found in Central Sweden, followed by Southern Sweden; Southern Norway (low-altitude lakes), and Southern Finland. Fish caught in regions with forest and/or peatlands had higher levels of Hg than cultivated soils and high elevated regions. This has also been observed by several other studies, indicating that there is a link between levels of dissolved organic matter and Hg accumulation in fish.

In this Nordic survey, surface sediments were also included in the analyses. According to the authors, patterns of standardised Hg concentrations in fish were almost identical with patterns in concentrations of Hg in bottom surface sediments after adjustment to dry weight.

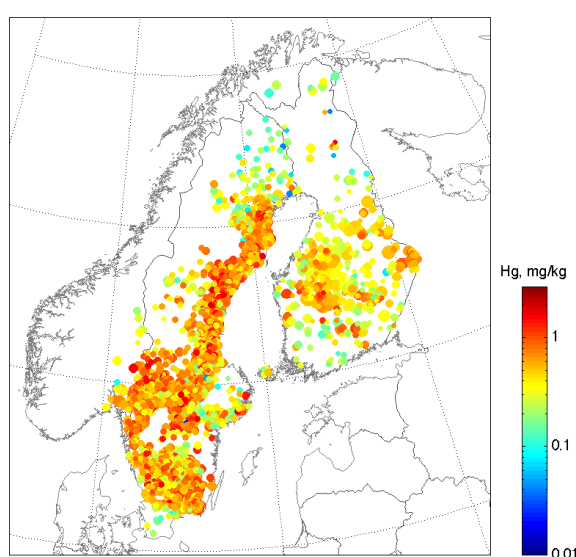


Figure 6. Nordic map showing the site-specific arithmetic means of observed mercury concentrations (mg kg^{-1} fw, without any adjustments), in pike (*Esox lucius*) collected 1965-2004 in 2517 lakes and rivers. The dot size indicates the number of individuals analysed. Figure is from (Munthe et al., 2007b).

5.3.3 Europe – remote mountain lakes

Some data exist concerning Hg in fish caught from remote mountain lakes. Ecological studies of remote mountain lakes in Europe (from the Arctic to the south of Spain) situated above or beyond the tree-line were reported in the EU-projects AL:PE 1992-1995; (Rosseland et al., 1996), MOLAR 1996-1999; (Wathne and Rosseland, 2000), and EMERGE 1999-2003; (Rosseland et al., 2003). In these projects Hg (not ALPE) and several other pollutants were measured in various fish organs. Results from MOLAR, although from a limited number of samples compared to other surveys, showed no seasonal changes in Hg concentration in fish and Hg concentrations were lower than 0.4 mg kg^{-1} in all samples (brown trout, Arctic charr, and other Salmonoidae). Similar Hg values were reported from the MOLAR project, where five European high mountain lakes and one lake from the Svalbard archipelago were examined. Mercury concentration in Arctic charr and brown trout did not exceed 0.50 mg kg^{-1}

(Rognerud et al., 2002a). In Lochnagar in Scotland, the HgT concentration in brown trout (0.23 mg kg⁻¹ ww) was the highest found in a non-piscivorous fish in the study.

5.3.4 North America

In North America historical records of MeHg in piscivorous fish dates back 35 years (Wiener et al., 2007).

USEPA (EPA, 2001) has compiled records of Hg content in fish caught in 43 states from the 1980s to 2000. Results are shown in **Table 2**.

Table 2. Mercury concentration (mg kg⁻¹ ww) for selected fish species from 43 states in the U.S. from 1980s to 2000 (EPA, 2001).

Species	Mean concentration	Range
	Hg (mg kg ⁻¹ ww)	Hg (mg kg ⁻¹ ww)
Largemouth bass	0.52	0.0005-8.94
Smallmouth bass	0.32	0.005-3.34
Yellow perch	0.25	0.005-2.14
Eastern chain pickerel	0.61	0.014-2.81
Lake trout	0.27	0.005-2
Walleye	0.43	0.005-16
Northern pike	0.36	0.005-4.4

A comprehensive survey of freshwater fish in Northeast North America was performed by Kamman et al. (2005b). This dataset comprised of 15,305 records of fish tissue from New York State to Newfoundland. Results showed that Hg in fish varied with geographic area, waterbody type. Highest Hg content was found in muskellunge (*Esox masquinongy*), walleye (*Sander vitreus*), white perch (*Morone americana*), and northern pike, and elevated concentrations were found in reservoirs relative to lakes and rivers. Certain geographical regions showed higher or lower than average concentrations of Hg in fish, but in general a highly heterogeneous pattern was observed. Considering all the waterbodies examined, 14 % of standard-length brook trout and 42% of standard-length yellow perch, exceeded the US EPA's criteria for MeHg in fish (0.3 mg kg⁻¹).

In another US survey, 2707 fish from streams/rivers in Western US states were collected and analysed for Hg. Levels in large fish (>120 mm total length) were correlated to both fish length and trophic level. The mean Hg concentrations in piscivorous and non-piscivorous were 0.260 mg kg⁻¹ and 0.090 mg kg⁻¹, respectively. Salmonids, the most common fish in this survey, exceeded the US EPA's criteria for fish fillet, equivalent of 0.3 mg kg⁻¹ in 2.3% of cases, while 57% of piscivorous exceeded this limit. According to the authors, atmospheric transport is the key factor determining the Hg content in fish across the western US (Peterson and van Sickle, 2007). Mercury content in fish from the WACAP project, covering 8 national parks on the US West coast, increased with fish age in all species up to approximately 15 years. Fish older than 15 years had lower contents of Hg. In-lake processes, biological processes, including fish age, Hg methylation, watershed biogeochemical characteristics, and food web efficiency influenced the higher rates of bioaccumulation found in Alaska lakes (Landers et al., 2008).

In Canada, the Sport Fish Contaminant Monitoring Program was established in 1976, and it is the largest program of its kind in North America. This programme collects fish samples every second year, and today approximately 120 000 analyses of Hg and other contaminants on 12 000 samples have been conducted. Based on these results, size-specific consumption advice for each species tested from each location is provided in a guide. Today, consumption advices for approximately 1860 locations have been developed and the guide is still developing further, by adding new sites, species, and updated information on the contaminants (www.ene.gov.on.ca/en/water/fishguide/guides.php).

Fish from lakes in Northern Canada were examined in a report from 2005 by Lockhart and co workers (Lockhart et al., 2005). Hg was found in measurable levels in all fish muscle tissues sampled. In this survey 7974 fish samples covering 25 species were examined for Hg. Walleye, northern pike and lake trout usually exceeded the subsistence consumption guideline of 0.3 mg kg⁻¹ HgT, and often the higher guideline of 0.5 mg kg⁻¹. Levels of Hg in whitefish were in general low. Results from this survey are shown in **Table 3**. A conclusion drawn from this study is that there is a widespread problem with Hg in subsistence fisheries for predator species. This will affect Native People in northern Canada, where fish play a key role in meeting nutritional needs.

Table 3. Numbers of length-adjusted means of levels of Hg in muscle found in concentration range with averages of the means and standard deviations in parentheses from Northern Canada (Lockhart et al., 2005).

Species	Sites with mean Hg in fish < 0.2 mg kg ⁻¹	Sites with mean Hg in fish between 0.2 -0.5 mg kg ⁻¹	Sites with mean Hg in fish > 0.5 mg kg ⁻¹
Lake trout (n = 94)	29 (0.14 ± 0.04)	46 (0.32 ± 0.09)	19 (0.79 ± 0.29)
Whitefish (n = 81)	69 (0.08 ± 0.04)	12 (0.27 ± 0.06)	0
Arctic charr (n = 55)	53 (0.05 ± 0.04)	1 (0.26)	1 (1.78)
Walleye (n = 29)	11 (0.14 ± 0.04)	11 (0.35 ± 0.07)	7 (0.92 ± 0.30)
Northern pike (n = 75)	21 (0.14 ± 0.05)	42 (0.35 ± 0.07)	12 (0.70 ± 0.25)
Burbot (n = 39)	24 (0.12 ± 0.04)	14 (0.30 ± 0.08)	1 (0.89)

5.3.5 Russia

Literature concerning Hg in fish from Russia is limited and scattered, and national monitoring programmes appear to be lacking. Some work was published in the 90's from Karelia and the Darwin national reserve 300 km north of Moscow (Amundsen et al., 1997; Haines et al., 1992; Haines et al., 1995; Stepanova and Komov, 1997). In a study by from the Pasvik River, fish were sampled from three locations at the border between Russia and Norway. Together with Hg, several other heavy metals were included. Results showed that Hg concentration in fish were independent of proximity to the large smelters at the Kola Peninsula. The mean concentration of Hg in the muscle was 0.16-0.89 mg kg⁻¹ dw, considering all species and localities. Concentrations of Hg in the liver were in general higher or equal to that in the muscle. Elevated concentrations were found in piscivorous fish. The levels of Hg found were as follows; Burbot > Perch > Brown trout ≈ Pike > Vendace ≈ D.r. whitefish ≈ S.r. whitefish. According to the authors all results from the Kola Peninsula were comparable to “metal-contaminated” lakes (Amundsen et al., 1997). In another study from North Western Russia by Moiseenko et al. (2006), several fish species were sampled during 1998-2003, and Hg levels

in various organs were measured. No fish species had levels of Hg above 0.5 mg kg^{-1} . Size and trophic level were not reported.

Arctic Russia is covered in the AMAP programme, and results are presented below.

5.3.6 The Arctic

Arctic charr, brown trout, burbot (*Lota lota*), northern pike, and whitefish (*Coregonus* spp. and *Prosopium* spp.) were designated as indicators for monitoring of Hg accumulation in the Arctic Monitoring and Assessment Programme (AMAP, 2005). Arctic charr is considered as the key species for monitoring of Hg in Arctic fish, due to its circumpolar distribution, its likelihood to occupy different trophic positions, and its importance for consumption. In Greenland, Arctic charr and threespine stickleback (*Gasterosteus aculeatus aculeatus*) are the only freshwater fish found. Results have shown that landlocked charr contain considerably higher levels of Hg than sea-running Arctic charr. Hg concentrations in landlocked charr caught in Canada, Iceland, and northern Fennoscandia were in generally considered low. Higher levels were found in Greenland, Chukotka-Lavrentiya (Russia), the Faeroe Islands, and lakes in Canadian High Arctic. The clear relationship between increasing size (age) and elevated concentrations of Hg in the fish described above, was not observed in Arctic charr. A possible reason for this is that Arctic charr within one lake may have a complex feeding strategy; ranging from benthic to piscivorous habitats, independent on size (age).

Whitefish (*Coregonus* spp and *Prosopium cylindraceum*, and several other species often treated together in the AMAP assessment) are widely distributed across circumpolar Arctic. Recent and numerous data from Canada, Russia, Finland, Norway, and US (Alaska) exist, which give a good spatial coverage of the Hg content in whitefish. Most data report values between $0.05 - 0.20 \text{ mg Hg kg}^{-1} \text{ ww}$. Elevated Hg concentrations are most often found in fish high up in the food chain (piscivorous fish), but deviations from this pattern are found at several sites, and spatial variability between sites and species are high. Highest levels of Hg are generally found in walleye, northern pike, lake trout, inconnu, and perch. For comprehensive information on Hg accumulation in various species and sites, reference should be made to the AMAP (2005).

Assessments of temporal trends in fish are limited, since most data sets are too short. Data from Lake Storvindeln, a forest lake in Sweden, spanned 1968-1996, but does not show any significant trend in Hg in pike muscle tissue. Similar results were found in a study from 1981-2001 in Abiskojaure (Sweden), where Arctic charr were sampled. According to results from the Swedish study, 10 - 20 years of monitoring data is required to detect average trends of 5% change per year for Hg in fish (AMAP, 2005).

5.3.7 Summary: Hg in fish

Levels of Hg in fish vary with species, size and age, differences in Hg exposure and in food web structure and dietary strategy. The complexity of the mechanisms that control Hg levels in fish make it difficult to do a meaningful comparison of different surveys and regions that results in conclusions on the correlation between atmospheric Hg loading and Hg levels in fish.

Accumulation of Hg in fish for human consumption has received considerable attention internationally and many countries have fish consumption advices related to Hg. The most

commonly used recommended limit for consumption is 0.5 mg kg^{-1} , although there are other limits in some countries and species. The US EPA recommends 0.3 mg kg^{-1} , while Sweden 0.5 mg kg^{-1} for most species, but 1 mg kg^{-1} for pike. These recommended values are all based on data from one epidemiological study (from the Faeroe Islands, Grandjean et al. 1997), but are adapted to suit local conditions, risk acceptance and politics. The choice of such limits for fish consumption, and the communication to the public of the balance between the potential risks and benefits of eating fish with elevated Hg concentrations, have been discussed internationally (Hoyt, 2008; Schwartz, 2006; Trasande et al., 2006).

An overview of data from large surveys from Scandinavia and North America is taken from the work of Munthe and collaborators (2007) (**Table 4**). The data show that Northern pike caught in Scandinavia has slightly elevated levels of Hg compared with those caught in North America. For perch, the concentrations are comparable or somewhat lower in Scandinavia, while for the other species the mean Hg concentration is considerable lower in Scandinavia compared to North East America (Munthe et al., 2007b). Levels of Hg in these fish species frequently exceed the recommended values for consumption. Another interesting observation is that virtually all fish analysed for Hg in Scandinavia exceed the environmental quality standard (EQS) established by the EU WFD of $0.020 \text{ mg Hg kg}^{-1} \text{ ww}$.

Table 4. Data from large fish surveys in North America and Scandinavia. Mean values are presented. Table is copied from (Munthe et al., 2007).

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

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

6. Monitoring mercury in aquatic ecosystems

The main aim of the ICP Waters Programme is to assess, on a regional basis, the degree and geographical extent of the impact of atmospheric pollution, on surface waters. Presently, no agreed guidelines exist for the monitoring of distribution and effects of Hg from atmospheric deposition in surface waters. In the WFD, guidelines and environmental quality standards (EQS) for Hg in water, sediments, and biota have been established. Monitoring of Hg in the WFD will supply information on Hg on a general basis in the aquatic ecosystem. However, the guidelines and threshold values are not specifically adapted to collect data relevant for risk evaluation of exposure of humans and wildlife to Hg through consumption of fish. A monitoring programme designed and adapted exclusively for Hg, with the aim to measure the distribution and effects of long-range transported Hg in aquatic environment, is necessary.

Special care has to be taken with respect to selection of appropriate sampling sites, frequencies, receptors (indicators), etc., and the main focus will be sampling from locations that are minimally affected by local point sources or other disturbances in the catchments that could alter the Hg fluxes.

Results from the monitoring programme shall provide documentation and information to policymakers and stakeholders, who will consider the societal effects of reducing Hg-emissions.

Here we present a general outline of a monitoring programme that is suitable for supporting the aims of the LRTAP Convention.

6.1 Recommended receptors

In the ICP Waters programme we recommend using water, sediments, and fish as receptors for monitoring Hg in the aquatic environment. Each of the receptors has strengths and weaknesses. If only one receptor would be chosen, both ambition and aim of the monitoring program, as well as price and analytical challenges should be considered to make the best choice.

- Water
 - **Strengths:** appropriate for monitoring the general level of Hg and MeHg in the water, trends over time, and the geographical extent of contamination when many lakes in a region are monitored at the same time. Frequent sampling (e.g. weekly) in a lake or brook can give insight in catchment processes and in-lake processes controlling the Hg level in the water.
 - **Weaknesses:** variability in water chemistry over time calls for more analysis to be able to set the level of Hg in the water body, analytical challenges due to low concentrations, and problems with contamination of (due to low concentrations). Determining the 'natural' background level (baseline) is challenging due to detection limits, between-lake and seasonal variations, but is important in order to assess future changes.

- Sediments

- **Strengths:** integrates major processes in the catchment chronologically, and can be used to reveal both spatial pattern and temporal trends in lakes. Dated cores may be used to compare present day levels with pre-industrial levels, and thereby the whole pollution history can be resolved. Sediments are also good for evaluating geographical patterns when many lakes in a region are sampled in the same time period. Sediments are stable receptors and not sensitive to short term fluctuations in water chemistry.
 - **Weaknesses:** Sediments can not be used to obtain detailed insight in processes in the catchment, as the accumulation of Hg in sediments depends on many variables. Care should be taken during sampling in order to avoid artefacts. Spatial variation in one lake may require that several sediment cores have to be collected in order to cover the natural variability.
- Fish
 - **Strengths:** fish, and especially piscivorous fish at the top of the food web aggregate processes in the catchment and in the lake that eventually lead to magnification of Hg. Compared to small sized prey fish, large and often piscivorous fish are a major dietary route for exposure to MeHg by humans. Knowledge about Hg concentrations in piscivorous fish is therefore necessary. Reasonably long records of Hg levels in fish makes it possible to elucidate eventual trends in concentrations. To be able to evaluate such trends, correct placement of organisms in food chains by stable isotope analysis, is required.
 - **Weaknesses:** as a monitoring tool, fish sampling is more time consuming and thereby more expensive than either sediment or water sampling. Another problem could be that sampling may have an impact on the population. Also, it is difficult to establish cause-effect mechanisms because it is such an integrated variable.

6.2 Recommendation for selection of sites

To obtain the best possible results from the monitoring, sites should be carefully chosen. ICP Waters has the following guidelines for choosing sites to monitor any pollutant from long-range transboundary air pollution.

- The sites should preferably be headwater lakes, or first order streams, without local pollution sources or any other mechanical disturbance in the catchment, such as forest harvesting, or large water level fluctuations due to e.g. hydroelectric power generation.
- The catchment to lake ratio should be sufficiently large to secure a good sedimentation rate, in order to obtain suitable samples for sediment analysis.
- The lake should have, if possible, a well-defined sedimentation basin in order to obtain suitable samples for sediment analysis.
- There should be a sustainable fish population in the lake in order to obtain suitable samples for analyzing Hg in fish.

6.3 Recommended supporting data

In order to be able to interpret, harmonise and compare the data from one lake to another and from one region to another, supporting data in addition to Hg concentration in the various receptors is necessary.

- **Catchment data:** catchment size, lake size, lake depth, water flux, vegetation cover

- **Lakewater data:** General water chemistry; all major ions including DOC/TOC. These data are necessary to interpret the observed Hg-concentrations.
- **Sediment data:** length of sediment core and position of sample, TOC, LOI, water content/porosity. These data are needed to normalise the measured values, to be able to compare data from different lakes and across regions. Sediments should be dated and fluxes measured.
- **Fish data:** Fish species, sex, weight, length, age, maturity stage, and $\delta^{15}\text{N}$ -analysis. All these data are necessary to be able to interpret the results and compare the results from different lakes and regions with each other

6.4 Recommendations for analytical procedures for measurement of Hg

Mercury is a ubiquitous element which is found at varying concentrations and forms in the environment. Except for fish, long term monitoring studies of Hg in the environment are scarce. This is largely ascribed to methodological detection limits having been higher than the levels found in environmental samples. Due to the low concentrations often found in aquatic samples, clean sampling and handling procedures are required along with correct preservation techniques.

Analyses of Hg in fish and sediments are to a large extent, considered as straight forward, and performed routinely in many laboratories. In fish, 95% or more of the Hg is in the form of MeHg, which makes it possible to measure total Hg instead of MeHg and use this as an estimate of MeHg (Munthe et al., 2007). In sediments, determination of MeHg is often problematic, due to preservation of the original Hg species, variability of the matrix, and the low concentrations often present (Margetínová et al., 2008).

Although analytical methods have improved considerably recently with respect to improvement of detection limits and advances in speciation, accurate determination of low levels of Hg and MeHg is still an analytical challenge in aquatic samples (Díez and Bayona, 2008; Margetínová et al., 2008). In water, the concentration of Hg is often low, and an up-concentration/extraction step is often required. Such complicated work-up results in enhanced analysis costs. Currently, there is no commonly accepted method for detection of either Hg or MeHg in aquatic samples (Díez and Bayona, 2008; Leermakers et al., 2005); Margetínová et al., 2008). At present, a method developed by the US Environmental Pollution Agency (www.epa.gov; Method 1630 is widely used routinely for determination of Hg in aquatic samples. The limit of detection (LOD) for this method is 0.5 ng L^{-1} , although laboratories having sufficient facilities for conducting low level Hg analyses may achieve LODs of about 0.01 ng L^{-1} .

In order to secure accurate and precise results, all Hg analyses should only be performed by dedicated laboratories. Adequate facilities for the performance of Hg analyses are required, and the laboratory should participate in interlaboratory calibrations frequently, where certified reference materials are analysed.

7. Conclusions

The long-range transported pollutant mercury (Hg) is addressed in the Aarhus Protocol under the LRTAP Convention. It is of particular interest in surface waters, as alarmingly high levels of Hg have been reported in fish and sediments in remote lakes in northern boreal and arctic ecosystems with possible consequences for people and wildlife.

Currently, no international monitoring network for assessing distribution and effects of long-range transported Hg in freshwater ecosystems exists. The objective of this report is i) to assess the status of Hg in the aquatic environment based on published sources and ii) to make recommendations on the outline of a network suitable for monitoring Hg in freshwater ecosystems.

The ecologically most relevant form of Hg is methylmercury (MeHg), an organic form of Hg that is highly toxic and bioaccumulates in the foodweb. In fish, 95-99% of the Hg is the form of MeHg. Concentrations of Hg in fish in Europe and North America are often above the limit recommended for human consumption, especially in piscivorous fish. Recent studies from Scandinavia show increasing Hg levels in fish despite reduced Hg deposition. In South-Eastern Norway the concentration of Hg in perch has increased with about 60 % from 1991 to 2008.

Lake sediments are used as archives for atmospheric deposition, and most sediments indicate a decline in atmospheric Hg deposition in recent decades. Regional lake sediment surveys in Northern America, Scandinavia, northern parts of Russia, and in the Arctic show that recent lake sediments are considerably enriched relative to sediments from pre-industrial times.

Concentrations of Hg in remote surface waters are generally low, usually below 40 ng L⁻¹ although slightly higher in waters affected by natural organic matter. Methylmercury (MeHg) usually comprises between 0.1 to 30% of total Hg in water. In surface waters, concentrations of Hg and MeHg are strongly controlled by catchment processes. Despite these low concentrations, a considerable up-concentration through the food web is observed.

The threshold values for Hg in fish (0.3-1.0 mg Hg kg⁻¹), based on evaluations of human health risks through exposure via fish consumption, are considerably higher than environmental quality standards (EQS) for Hg in fish (0.02 mg Hg kg⁻¹) under the EU Water Framework Directive. Almost all fish in the compiled data in this report exceed the EQS-values, while a substantial number do not exceed the limit for human consumption. The balance between exposure to Hg through fish consumption versus its potential health benefits, is much discussed internationally.

Recommendations for a monitoring programme for Hg in freshwater ecosystems with the aim of assessing the effects of Hg originating from long-range atmospheric transport are made regarding site selection, receptors, methods, and the collection of supporting data.

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