ACCUMULATION AND FLUXES OF MERCURY IN TERRESTRIAL AND AQUATIC FOOD CHAINS WITH SPECIAL REFERENCE TO FINLAND

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ABSTRACT

Mercury is known for its biomagnification especially in aquatic food chains and for its toxic effects on different organisms including man. In Finland mercury has formerly been used in industry and agriculture and in addition many anthropogenic activities may increase the mercury levels in ecosystems. Phenyl mercury was widely used as slimicide in the pulp and paper industry in the 1950s and 1960s. In the chlor-alkali industry metallic mercury was used as catalyst at three plants. The most toxic form of mercury, methyl mercury, may be formed in soils, water, sediments and organisms. Many factors, including microbial activity, temperature, oxygen status etc., affect the methylation rate. In the lake ecosystem bioaccumulation of methyl mercury is very strong. In early 1980s there was a restriction of fishing concerning approximately 4000 km^2 of lakes and sea areas because of mercury pollution. In aquatic systems we still find elevated concentrations near former emission sources. Long-range atmospheric transport and mechanical operations like ditching and water regulation may cause increased levels of mercury in the aquatic ecosystems. In the Finnish agriculture organic mercury compounds were used for seed dressing until 1992. Although the amounts used were substantial the concentrations in agricultural soils have remained rather low. In terrestrial food chains bioaccumulation is normally weak with low or moderate concentration at all ecosystem levels. Due to a weak uptake through roots terrestrial, vascular plants normally contain only small amounts of mercury. There is a bidirectional exchange of mercury between vegetation and atmosphere. Contrary to vascular plants, there is a very wide range of concentrations in fungi. Mercury may pose a threat to human health especially when accumulated in aquatic food chains.

Keywords: mercury, flux, food chain, terrestrial, aquatic

Mercury as a global pollutant

Mercury is one of the potentially most hazardous environmental pollutants. It is known for its biomagnification especially in aquatic food chains and for its toxic effects on different organisms including man. Mercury has formerly been widely used in industry and agriculture. In addition many anthropogenic activities may increase the mercury levels in ecosystems. Mercury may pose a threat to human health especially when accumulated in aquatic food chains. This bio-accumulation is based on the formation of methyl mercury, which is persistent and lipid soluble. The methylation process is possible in almost all environments and is affected by several physical and biological parameters. The disasters in Minamata and Niigata (methyl mercury poisoning or "Minamata disesase") were examples of the negative effects of industrialization and chemicalisation, which lead to increased environmental awareness.

Although the emissions of mercury to water and atmosphere have decreased substantially during the last decades we still have too high concentration in many areas. The efficient spreading of air-borne emissions makes the mercury problem global. Hg^0 is rather inert and has a long life-time in the atmosphere. Although the use has been banned or strongly restricted in most countries, emission control, research and monitoring are still needed. Anthropogenic emissions have at least doubled global atmospheric mercury deposition compared to pre-industrial times. In total, approximately one million tons of metallic mercury has been extracted from cinnabar and other ores during the past five centuries. Of this amount only a few percent have escaped to the atmosphere (Hylander and Meili 2003).

Ice core results from Wyoming, USA, indicate major atmospheric sources of both natural and anthropogenic mercury from regional and global sources over the past 270 years. Schuster et al. (2002) calculate the contribution from anthropogenic inputs to 52%, from volcanic events to 6%, and from background sources to 42%. However, during the last 100 years, anthropogenic sources contributed to 70% of the total Hg input. During the last period of this ice-core record, a decline in atmospheric deposition was observed.

Global direct emissions to the atmosphere amount 10–30 t per year currently (up to 10 from the Spanish Almadén gold mine alone), and probably exceed 10,000 t historically (Hylander and Meili 2003). One important source of both atmospheric and aquatic mercury emissions is the widespread use of this metal in gold mining. Mercury forms an amalgam with gold and is later released when the gold is purified by heating. This partly illegal process is practised in the Amazon but also in many African and south Asian countries. Western Europe exports each year about 100 tons of mercury for this purpose to Brazil (Hylander 2001).

Burning of coal is another important source of mercury pollution. The global average concentration of mercury in coal is 0.3 mg kg^{-1} which gives a potential worldwide flux of 1683 t in 2003 (Table 1). Part of this amount is removed from the flue gases by filters and is deposited with the ash. The remain is spread over large areas. The total anthropogenic mercury emissions have been estimated to ca. 2190 tons. Southeast Asia, particularly China, South Africa, Central and Eastern Europe, and the Eastern United States are major mercury polluters due to burning of coal without appropriate smoke cleaning devices, industrial use, waste incineration and gold mining. With more than 600 tons of Hg, China contributes about 28% to the global emissions of mercury (Mukherjee et al. 2000; Hylander 2001; Pacyna et al. 2003).

Table 1 Some estimates of anthropogenic mercury fluxes.

Area	Year	Amount	Reference
Globally mined	2000	1849 t	Hylander and Meili 2003
Global atmospheric emissions – combustion of fossil fuels – industrial processes – Asia – Africa – Europe*	2000	2190 t 66% 30% 54% 18% 11%	Pacyna et al. 2006
Global amount in coals processed**	2003	1683 t	Mukherjee et al. 2008
Atmospheric emissions, Europe	1995	342 t	Pacyna et al. 2001
Atmospheric emissions, Finland	1997	0.62 t	Mukherjee et al. 2000

* Including European part of Russia.
** Including hidden flows of Hg through export and import of coal.

Use and emissions of mercury in Finland

In Finland phenyl mercury was widely used as slimicide in the pulp and paper industry in the 1950s and 1960s. This use ended in 1968 but there are still considerable amounts of mercury in the sediments of rivers and lakes downstream from these plants. In the chlor-alkali industry metallic mercury was used as catalyst at three plants (Äetsä, Kuusankoski and Oulu). The emissions were mainly to the atmosphere while lesser amounts were discharged to aquatic and terrestrial environments. The total emission of mercury to the atmosphere decreased from 1140 kg in 1990 to 620 kg in 1997. The reduction is a result of improved gas cleaning equipment, process changes, automation, installation of flue gas desulfurization in coal-fired power plants and strict pollution control legislation (Mukherjee et al. 2000). In Finland mercury has been extracted as a by-product of copper mining (Outokumpu and Kokkola).

Finnish lake sediments reveal that the first signs of increased levels of mercury and many other heavy metals can be seen during the 1800s. In the 1980s a subtraction of the background fluxes from the total fluxes gives 70–89% share for atmospheric deposition. This applies to southern and central Finland, areas that also have the highest deposition of acidic compounds. In northern Finland the increase in deposition starts later and has remained smaller than in the south (Verta et al. 1989).

In the Nordic countries mercury atmospheric concentrations and precipitation exhibit a strong south to north gradient. Trajectory analyses of episodes indicate main source areas in Central Europe. The wet deposition is influenced by oxidation of Hg^0 to water soluble species (RGM, Hg^{2+}) resulting in a smaller difference in gradient (Munthe et al. 2003). At least during the summer months the Baltic Sea, in particular its southern part and Gulf of Gdansk are also important sources of gaseous mercury (Urba et al. 2000). Also from the North Sea coastal region a decreasing trend in mercury wet deposition has been observed. The decrease in deposition is 10–30% when comparing the periods 1995–1998 and 1999–2002. This is obviously due to enhanced emission control in Europe. However, no decreasing trend in total gaseous mercury (TGM) could be observed during the same time periods (Wängberg et al. 2007).

Mercury deposition may be monitored by using mosses or epiphytic lichens which absorb both dry and wet deposition directly from the air. When studying the regional distribution of mercury in the lichen *Hypogymnia physodes* rather even concentrations were found in Finland with somewhat higher concentrations along the coasts in south and west, which indicated influence from Central Europe (Lodenius 1981). Also in Norway the concentrations in the moss *Hylocomium splendens* are quite even with no significant north–south gradient. This could be partly explained by a considerable supply of mercury from dry deposition of Hg^0 in addition to the retention of Hg^{2+} from wet deposition. Whereas the level and geographic distribution showed only small differences during 1985–1995 the data from year 2000 were approximately 30% lower (Steinnes et al. 2003).

Vegetation is an important factor for the removal of mercury from the atmosphere. Mercury is taken up by vegetation mainly from the air through the leaves and, to a lesser extent, from the soil through the roots. Using moss bag data Lodenius (1984) estimated the yearly background deposition to 8 μg m^{-2} while it was 1200 μg m^{-2} near (0–1 km) a chlor-alkali factory. Iverfeldt (1991b) estimated the bulk deposition over southern Finland in 1987–1989 to 11 μg m^{-2} (= g km^{-2}) while Porvari and Verta (2003) measured annual bulk deposition in the Lammi area to 5 μg m^{-2} in 1994–1995.

Approximately two-thirds of the mercury absorbed by moss bags originates from dry depostion and one-third from wet deposition (Lodenius 1998). In this measurement the deposition near a chlor-alkali plant was estimated to 480 μg m^{-2} per year. In a laboratory experiment mercury adsorption has shown to be rapid and strong for both moss (*Sphagnum girgensohnii*) and grass (*Loli-*

um perenne) at different temperatures (from +10 °C to +60 °C) and exposure times (from 1 h to 1 month) while the evaporation was negligible. Also the leaching of adsorbed mercury was small (Lodenius et al. 2003).

Terrestrial environment

Bedrock and soil

In most areas mercury occurs in very low concentrations in the bedrock with higher amounts in sedimentary rocks and organic-rich soils. The most important mineral is cinnabar (HgS). Mercury ores are often situated in geologically young areas with volcanic activity. Three main mercury mines in Europe have been in operation in Spain (Almadén), Italy (Amiata) and Slovenia (Idrija). Highly elevated concentrations have been found in soil and biota around these mines.

In the Finnish agriculture organic mercury compounds were used for seed dressing until 1992. E.g. in 1981, 5.6 t of metoxy ethyl mercury was used for this purpose. Although the amounts used were substantial, the concentrations in agricultural soils have remained rather low. In terrestrial food chains bioaccumulation is normally weak with low or moderate concentration at all ecosystem levels. In the soil mercury is mainly attached to organic substances with bounds of various strength but with a great affinity to SH-groups. Mercury is often strongly bound to the soil organic matter (Lodenius et al. 1987, 2003).

As mercury (partly in methylated form) has been spread over continents, considerable amounts have been deposited in forests soils and lake sediments also in remote areas. This mercury may be released as a result of changes in physical, chemical or biological conditions. Pools and fluxes of mercury have been studied in catchments in e.g. NE Bavaria, Germany and in Sweden. The soil storage of total mercury for a soil depth of 60 cm was calculated to approximately 890 g ha^{-1} in the coniferous and 190 g ha^{-1} in the deciduous catchment in Germany. The corresponding value for the Swedish catchment was 280 g ha^{-1}. Only 0.3–0.6% of the mercury was methylated (Munthe et al. 1998; Schwesig and Matzner 2000).

When studying soil concentrations in European city parks Rodrigues et al. (2006) found generally low but highly variable concentrations (from 0.015 to 6.3 mg kg^{-1}) of total mercury. The variability obviously reflected contributions from natural and anthropogenic activities and differences in the ages of cities and land use, local environmental conditions.

Vascular plants and fungi

Elemental mercury is easily evaporated and spread by winds. It has a long residence time in the atmosphere but may be attached to particles and/or oxidized and washed out by precipitation. It can also be directly absorbed by vegetation. Due to a weak uptake through roots terrestrial, vascular plants normally contain only small amounts of mercury. However, in a strongly polluted environment the uptake through leaves may be considerable. There is a bidirectional exchange of mercury between vegetation and atmosphere.

Mercury concentrations in vascular plants are normally very low, but in the vicinity of pollution sources plants may absorb significant amounts of mercury mainly from the air but also from the soil (Ellis and Eslick 1997; Lodenius et al. 2005). As this mercury is strongly retained by vegetation (Lodenius et al. 2003), forests and other vegetation cover form a sink for atmospheric mercury.

In a northern mixed-hardwood forest in the Lake Huron Watershed, USA, Rea et al. (2001) estimated the annual throughfall deposition flux of mercury to 10.5 μg m^{-2} compared to an annual precipitation flux of 8.7 μg m^{-2}. The difference may be explained by wash-off of dry deposition and foliar leaching. The calculated dry deposition flux (12–14 μg m^{-2}) to the canopy indicated that atmospheric deposition could account for all of the mercury deposited in net throughfall (mean 1.9 μg m^{-2}; Table 2). The authors conclude that atmospheric mercury may account for all of the mercury deposited in litterfall (11.4 μg m^{-2}). In eight small forest catchments in southern Finland Porvari and Verta (2003) estimated the output fluxes in runoff water of total mercury to 0.92–1.8 g km^{-2} a^{-1} and that of methyl mercury to 0.03–0.33 g km^{-2} a^{-1}.

Results from a spruce forest in the catchment of Lake Gårdsjön, south-western Sweden (Iverfeldt 1991a) show that annual dry deposition of mercury in throughfall water can be 50% of the wet deposition. A strong seasonal trend in deposition via throughfall water was observed with increased levels during the growing season and with a monthly maximum in August. During part of the winter, dry deposition in throughfall water seems to be negligible.

Contrary to vascular plants, there is a very wide range of concentrations in fungi. Many macrofungi show a strong ability to accumulate mercury and other heavy metals. The interspecific differences are great with generally low concentrations in mycorrhizal forest species and often considerably higher amounts in lawn decomposing species. The uptake and binding mechanisms in these species are still poorly known but methyl mercury is found in small quantities only. In contaminated areas also mycorrhizal fungi show elevated concentrations (Kuusi et al. 1981; Lodenius and Herranen 1981). Based on analyses on mushrooms near the city of Koszalin in North-central Poland Falandysz et al. (2004) estimated that the flesh of edible mushrooms may not pose hazards to human health even at a maximum consumption rate (28 g/day).

Table 2 Some estimates of annual total mercury fluxes µg m^{-2} to and from forests.

Measurement	Area	Flux	Reference
Total depositional flux	Background, spruce canopy, Sweden	40	Iverfeldt 1991a
Dry deposition	–"–	4–7	–"–
Dry + wet deposition	Chlor-alkali plant, moss bag, Finland	1200	Lodenius and Tulisalo 1984
–"–	Background, moss bag, Finland	8	–"–
Dry deposition	Chlor-alkali plant, moss bag, Finland	320	Lodenius 1998
Throughfall + stemfall	Upland, Minnesota, USA	13	Grigal et al. 2000
Litterfall	–"–	12.3	–"–
Wet deposition	–"–	160	–"–
Total throughfall	Background, mixed hardwood forest, USA	10.5	Rea et al. 2001
Precipitation	–"–	8.7	–"–
Litterfall	–"–	11.4	–"–
Runoff water	Upland, Minnesota, USA	2.8	Grigal et al. 2000
–"–	Peatland, Minnesota, USA	4.4	–"–
Runoff water	Upland, Wisconsin, USA	0.04	Hurley et al. 1995
–"–	Wetland - forest, Wisconsin, USA	5.5	–"–
Runoff water	Upland, Ontario, Canada	1.2–2.3	St Louis et al. 1996
Runoff water	Upland, wetland, SW Sweden	1.8	Hultberg et al. 1995
Runoff water	Forest catchment, S Finland	0.92–1.8	Porvari and Verta 2003

Animals

Like in terrestrial plants, the mercury concentrations are normally very low in mammals belonging to terrestrial food chains. Free ranging Alaskan reindeer had mean mercury levels of 55 mg/kg in their hair while, interestingly, reindeer fed a pollock-based fishmeal diet had only 19 ng/g. Younger reindeer (2 years of age or less) showed lower levels (0.8 ng/g) compared to adult reindeer (Duffy et al. 2005). When Sobanska (2005) studied hair samples of wild boar (*Sus scrofa*) from four differently polluted regions of Poland, she found the highest mercury concentrations (mean 0.16 mg/kg in middle part of mane hair) in young individuals between 1 and 2 years of age. Mercury concentrations in feathers of Sparrow hawk (*Accipiter nisus*) seemed to follow the industrial use of mercury in Finland (Solonen and Lodenius 1984).

In an area near the Idrija mine Gnamus et al. (2000) showed that food intake of mercury in roe deer (*Capreolus capreolus* L.) is much more important than inhaled mercury, which represents only up to 0.2% of ingested mercury. Higher accumulation of methyl mercury was observed in environments polluted with high concentrations of inorganic mercury compared to less contaminated and control areas.

Waste

The global primary production of mercury was 1800 t in the year 2000 and that of Finland 45 t (UNEP 2002). Mercury use is declining both globally and in the European Union (EU-15) accounting for 440 tonnes in 2005. European Commission has proposed legislation to ban all European Union exports of mercury from 2011 (European Commission 2006). The amount of mercury in waste for the year 1995 in the EU has been estimated at around 990 t (including coal combustion products, landfills, chlor-alkali waste and incinerator slag). If complete information would be available for the 15 member states, the amount would be 2–4 times larger. Mercury is occasionally recovered from waste, but this is often discouraged for economic reasons. The use of mercury in lamps and batteries is declining, and several countries have strict regulations on the use mercury in e.g. dentistry and electrical equipment Mukherjee et al. 2004).

Aquatic environment

Mercury pollution in Finnish watercourses

In early 1980s there was a restriction of fishing concerning approximately 4000 km^2 of lakes and sea areas because of mercury pollution (Lodenius 1985). In aquatic systems we still find elevated concentrations near former emission sources. In addition, the fish concentrations may be high also in seemingly unpolluted areas. Long-range atmospheric transport and mechanical operations like ditching and water regulation may cause increased levels of mercury in the aquatic ecosystems. As a consequence of significantly reduced direct emissions to the watercourses concentration of mercury in fish has decreased in many formerly polluted areas (Lodenius 1991). Recent research has given much new information on the importance of water quality parameters and on methylation and bioaccumulation processes.

Methylation

Methylation is a key process in the bioaccumulation of mercury. The most toxic form of mercury, methyl mercury, may be formed by biotic and abiotic processes in soils, water, sediments and organisms. Many factors, including microbial activity, temperature, oxygen status etc., affect the methylation rate. Sulphate-reducing bacteria are important methylators of inorganic mercury to methyl mercury (MeHg) in many environments (Gilmour and Henry 1991). Methyl mercury formed in the drainage area of a lake contributes to the total load in aquatic systems. In the lake ecosystem bioaccumulation of methyl mercury is very strong: although the concentrations in water are near detection limit, concentrations in predatory fish and fish eating birds may be very high.

In the soil methylation may take place under favourable conditions. In a laboratory study Matilainen et al. (2001) found organic surface layer, especially living moss to be the dominant scavenger of the added mercury. Methylation was also most intensive in this layer. Increased precipitation mobilized part of mercury from the moss layer to the deeper layers and leachate water. Fertilizing of the soil seemed to increase the availability of mercury for methylation. Humic substances seem to be main carriers of methyl mercury in lake water but not active methylation agents (Porvari and Verta 1995). The results from south Finnish boreal catchments of Porvari and Verta (2003) indicate a more efficient methylation in peatlands compared to uplands with mineral soils.

Acidification and humic matter

Acidification of lakes often results in elevated mercury concentrations in biota. Many lake surveys have documented a negative correlation between lake water pH and fish mercury concentrations (Watras et al. 2006). This is related to increased methylation in the water column and at the water–sediment interface and changes in fish communities and fish growth. Decreased pH may also decrease the loss of volatile mercury and increase binding of mercury to particles. Liming of the water body and/or the catchment may thus mercury concentrations in fish (Meili 1995; Hrabik and Watras 2002; Rask et al. 2007).

Finnish lakes are often small and polyhumic with low pH and low alkalinity. Porvari and Verta (2003) studied total mercury and methyl mercury concentrations in runoff from eight small boreal forest catchments. Runoff waters of the studied lakes were very humic (TOC 7 – 70 mg^{-1}) and the total mercury concentrations varied between 0.84 and 24 ng^{-1} and methyl mercury between 0.03 and 3.8 ng^{-1}. The concentrations and output fluxes of total mercury were similar to those measured in other boreal regions. The fluxes from catchments of total mercury ranged from 0.92 to 1.8 g km^{-2} a^{-1} and those of methyl mercury from 0.03 to 0.33 g km^{-2} a^{-1}. Peatland catchments released more methyl mercury than pure mineral soil.

Ditching and water level control

Although the binding to organic matter normally is strong, mercury may be released from the soil by mechanical operations like forest or peatland ditching (Simola and Lodenius 1982; Leinonen 1989), agricultural operations (Bash and Miller 2007) or fluctuations in the water level. Clearly elevated concentrations of mercury have been found from man-made lakes in northern Finland. The concentrations often exceeded the Finnish safety limits of 0.5 and 1 mg kg^{-1} fresh weight. These concentrations were also reflected in elevated concentrations in human hair in these areas (Lodenius and Seppänen 1982; Lodenius et al. 1983; Porvari et al. 2003).

Mercury concentrations in fish were monitored in 18 reservoirs impounded in 1964–1980 in western and northern Finland over a period from 1979 to 1994. In most cases fish mercury concentrations in reservoirs exceeded those in natural lakes. Shortly after inundation, fish mercury concentrations clearly increased and remained above background concentrations for 15–25 years. The 1 mg kg^{-1} fish Hg level recommended by health authorities as the upper limit for human consumption was still exceeded in two reservoirs 20 years old or more (Porvari 1998).

Bioaccumulation

Aquatic plants normally show very low concentrations of mercury but in the vicinity of emission sources, the concentrations may be much higher (Lodenius 1980). As mercury is readily accumulated in aquatic food chains, high concentrations may be found besides predatory fish, also in fish eating birds and mammals (Särkkä et al. 1978; Solonen and Lodenius 1990). The seals in Lake Saimaa (*Phoca hispida saimensis*) had elevated concentrations of mercury in liver, muscle and kidney. The concentrations dropped in the middle of the 1980s. Mercury pollution might have been one reason for the decrease in population density of the Saimaa seal in this century (Hyvärinen et al. 1998).

Baltic Sea

The mercury problem in Baltic Sea is generally lower compared to Finnish lakes. However, Marks (2002) found on a research expedition on the Baltic Sea two areas with distinctly elevated mercury saturation were found: one south of Bornholm and another over the ammunition disposal region south-east of Hoburgs Bank. These data indicate that certain maritime areas can emit gaseous mercury from surface waters into the atmosphere and contribute

to long-range atmospheric transport. In fish, the concentrations are often lower than in lake ecosystems with a less pronounced bioaccumulation in food chains. Only in few cases the concentrations exceed the maximum levels stipulated for human consumption (Voigt 2004).

Trends and perspectives

Pacyna et al. (2006) predict that future changes of mercury emissions from anthropogenic sources worldwide until the year 2020 should be within ±20% of the current estimates, "although this assessment should be treated with great caution". Mercury has been globally used for many different purposes, but nowadays economically viable mercury-free alternatives exist for practically all applications. Thus the production and use of mercury can be further reduced and all primary production of Hg other than by-production terminated. Global mercury pollution would significantly diminish if the mercury trade in Europe and North America was totally banned (Hylander 2001; Hylander and Meili 2003). However, on a global scale, gold mining will remain an important source of mercury to both terrestrial and aquatic ecosystems.

Lakes in the boreal areas remain sensitive for mercury pollution because of cool climate with slow degradation, slow fish growth, acidic and polyhumic water, atmospheric deposition and pools accumulated in forest soils. Direct anthropogenic pollution will continue to decrease but combustion of coal will continuously be an important energy source and obviously also an important source of mercury emissions. Modern technology can significantly reduce particle-bound and gaseous mercury in flue gases, but the costs certainly retards the introduction of these methods e.g. in China and other developing countries.

REFERENCES

Bash JO, Miller DR (2007) A note on elevated total gaseous mercury concentrations downwind from an agriculture field during tilling. Sci Total Environ 388: 379–388.

Duffy LK, Duffy RS, Finstad G, Gerlach C (2005) A note on mercury levels in the hair of Alaskan reindeer. Sci Total Environ 339: 273–276.

Ellis RW, Eslick L (1997) Variation and range of mercury uptake into plants at a mercury-contaminated abandoned mine site. Bull Environ Contam Toxicol 59: 763–769.

European Commission (2006) Commission proposes ban on EU mercury exports. IP/06/1481 Brussels, 26 October 2006.

Falandysz J, Jezdrusiak A, Lipka K, Kannan K, Kawano M, Gucia M, Brzostowski A, Dadej M (2004) Mercury in wild mushrooms and underlying soil substrate from Koszalin, North-central Poland. Chemosphere 54: 461–466.

Gilmour CC, Henry EA (1991) Mercury methylation in aquatic systems affected by acid deposition. Environ Pollut 71: 131–149.

Gnamus A, Byrne A, Horvat M (2000) Mercury in the soil-plant-deer-predator food chain of a temperate forest in Slovenia. Environ Sci Technol 34: 3337–3345.

Grigal DF, Kolka RK, Fleck JA, Nater EA (2000) Mercury budget of an upland-peatland watershed. Biogeochem 50: 95–109.

Hrabik TR, Watras CJ (2002) Recent declines in mercury concentration in a freshwater fishery: Isolating the effects of de-acidification and decreased atmospheric mercury deposition in Little Rock Lake. Sci Total Environ 297: 229–237.

Hultberg H, Munthe J, Iverfeldt Å (1995) Cycling of methyl mercury and mercury – Responses in the forest roof catchment to three years of decreased atmospheric deposition. Water Air Soil Pollut 80: 415–424.

Hurley JP, Benoit JM, Babiarz CL, Shafer MM, Andren AW, Sullivan JR, Hammond R, Webb DA (1995) Influences of watershed characteristics on mercury levels in Wisconsin rivers. Environ Sci Technol 29: 1867–1875.

Hylander L (2001) Global mercury pollution and its expected decrease after a mercury trade ban. Water Air Soil Pollut 125: 331–344.

Hylander L, Meili M (2003) 500 years of mercury production: global annual inventory by region until 2000 and associated emissions. Sci Total Environ 304: 13–27.

Hyvärinen H, Sipilä T, Kunnasranta M, Koskela JT (1998) Mercury pollution and the Saimaa ringed seal (*Phoca hispida saimensis*). Marine Pollut Bull 36: 76–81.

Iverfeldt Å (1991a) Mercury in forest canopy throughfall water and its relation to atmospheric deposition. Water Air Soil Pollut 56: 311–330.

Iverfeldt Å (1991b) Occurrence and turnover of atmospheric mercury over the Nordic countries. Water Air Soil Pollut 56: 251–265.

Kuusi T, Laaksovirta K, Liukkonen-Lilja H, Lodenius M, Piepponen S (1981) Lead, cadmium and mercury contents of fungi in the Helsinki area and in unpolluted control areas. Z Lebensm Unters Forsch 173: 261–267.

Leinonen K (1989) The influence of soil preparation on the levels of aluminum, manganese, copper, zink, cadmium and mercury in *Vaccinium myrtillus*. Chemosphere 18: 1581–1587.

Lodenius M (1980) Aquatic plants and littoral sediments as indicators of mercury pollution in some areas in Finland. Ann Bot Fennici 17: 336–340.

Lodenius M (1981) Regional distribution of mercury in *Hypogymnia physodes* in Finland. Ambio 10: 183–184.

Lodenius M (1985) The mercury problem and fishing in Finland. In: Hall DO, Myers N, Margaris NS (eds): Economics of ecosystem management: pp 99–103. Junk Publ., Dordrecht.

Lodenius M (1991) Mercury concentrations in an aquatic ecosystem during twenty years following abatement the pollution source. Water Air Soil Pollut 56: 323–332.

Lodenius M (1998) Dry and wet deposition of mercury near a chloralkali plant. Sci Tot Environ 213: 53–56.

Lodenius M, Herranen M (1981) Influence of a chloralkali plant on the mercury contents of fungi. Chemosphere 10: 313–318.

Lodenius M, Seppänen A (1982) Hair mercury contents and fish eating habits of people living near a Finnish manmade lake. Chemosphere 11: 755–759.

Lodenius M, Tulisalo E (1984) Environmental mercury contamination around a Finnish chloralkali plant. Bull. Environ. Contam Toxicol 32: 439–444.

Lodenius M, Nuorteva P, Nuorteva SL (2005) Mercury in vascular, terrestrial plants Comparison between Finland and Slovenia in the years 1979–1981. Acta Univ Carolinae Environmentalica 19: 91–96.

Lodenius M, Seppänen A, Autio S (1987) Leaching of mercury from peat soil. Chemosphere 16: 1215–1220.

Lodenius M, Seppänen A, Autio S (1987) Sorption of mercury in soils with different humus content. Bull Environ Contam Toxicol 39: 593–600.

Lodenius M, Seppänen A, Herranen M (1983) Accumulation of mercury in fish and man from reservoirs in northern Finland. Water Air Soil Pollut 19: 237–246.

Lodenius M, Tulisalo E, Soltapour Gargari A (2003) Exchange of mercury between atmosphere and vegetation under contaminated conditions. Sci Total Environ 304: 169–174.

Marks R (2002) Preliminary investigation of the mercury saturation in the Baltic Sea winter surface water. Sci Total Environ 229: 227–236.

Matilainen T, Verta M, Korhonen H, Uusi-Rauva A, Niemi M (2001) Behavior of mercury in soil profiles: Impact of increased precipitation, acidity, and fertilization on mercury methylation. Water Air Soil Pollut 125: 105–119.

Meili M (1995) Liming effects on mercury concentrations in fish. In: Henrikson L, Brodin YW (eds.) Liming of Acidified Surface Waters. Springer-Verlag, Berlin-Heidelberg, pp 383–398.

Mukherjee AB, Melanen M, Ekqvist M, Verta M (2000) Assessment of atmospheric mercury emissions in Finland. Sci Total Environ 259: 73–83.

Mukherjee AB, Zevenhoven R, Bhattacharya P, Sajwan KS, Kikuchi R (2008) Mercury flow via coal and coal utilization by-products: A global perspective. Resour Conserv Recy 52: 571–591.

Mukherjee AB, Zevenhoven R, Brodersen J, Hylander LD, Bhattacharya P (2004) Mercury in waste in the European Union: sources, disposal methods and risks. Resour Conserv Recy 42: 155–182.

Munthe J, Lee YH, Hultberg H, Iverfeldt Å, Borg GC, Andersson I (1998) Cycling of mercury and methylmercury in the Gårdsjön catchment. In: Hultberg H, Skeffington R (eds): Experimental reversal of acid rain effects: The Gårdsjön roof project. John Wiley and Sons Ltd., 1998, pp 261–276.

Munthe J, Wängberg I, Iverfeldt Å, Lindqvist O, Strömberg D, Sommar J, Gårdfeldt K, Petersen G, Ebinghaus R, Prestbo E, Larjava K, Siemens V (2003) Distribution of atmospheric mercury species in Northern Europe: final results from the MOE project. Atmos Environ 37 Suppl 1: S9–S20.

Pacyna EG, Pacyna JM, Pirrone N (2001) European emissions of atmospheric mercury from anthropogenic sources in 1995. Atmos Environ 35: 2987–2996.

Pacyna EG, Pacyna J M, Steenhuisen F, Wilson S (2006) Global anthropogenic mercury emission inventory for 2000. Atmos Environ 40: 4048–4063.

Pacyna EG, Pacyna JM, Fudala J, Strzelecka-Jastrza E, Hlawiczk S, Panasiuk D (2006) Mercury emissions to the atmosphere from anthropogenic sources in Europe in 2000 and their scenarios until 2020. Sci Total Environ 370: 147–156.

Pacyna JM, Pacyna EG, Steenhuisen F, Wilson S (2003) Mapping 1995 global anthropogenic emissions of mercury. Atmos Environ 37 Suppl 1: 109–117.

Porvari P (1998) Development of fish mercury concentrations in Finnish reservoirs from 1979 to 1994. Sci Total Environ 213: 279–290.

Porvari P, Verta M (1995) Methylmercury Production in Flooded Soils: A Laboratory Study. Water Air Soil Pollut 80: 765–773.

Porvari P, Verta M (2003) Total and methyl mercury concentrations and fluxes from small boreal forest catchments in Finland. Environ Pollut 123: 181–191.

Porvari P, Verta M, Munthe J, Haapanen M (2003) Forestry practises increase mercury and methyl mercury output from boreal forest catchments. Environ Sci Technol 37: 2389–2393.

Rask M, Jones R, Järvinen M, Paloheimo A, Salonen M, Syväranta J, Verta M (2007) Changes in fish mercury concentrations over 20 years in an acidified lake subject to experimental liming. App Geochem 22: 1229–1240.

Rea AW, Lindberg SE, Keeler GJ (2001) Dry deposition and foliar leaching of mercury and selected trace elements in deciduous forest throughfall. Atmos Environ 35: 3453–3462.

Rodrigues S, Pereira ME, Duarte AC, Ajmone-Marsan F, Davidson CM, Grcman H, Hossack I, Hursthouse AS, Ljung K, Martini C, Otabbong E, Reinoso R, Ruiz-Cortés E, Urquhart GJ, Vrščaj B (2006) Mercury in urban soils: A comparison of local spatial variability in six European cities. Sci Total Environ 368: 926–936.

Särkkä J, Hattula M-L, Paasivirta J, Janatuinen J (1978) Mercury and chlorinated hydrocarbons in the food chain of Lake Päijänne, Finland. Holarctic Ecol 1: 326–332.

Schuster PF, Krabbenhoft DP, Naftz DL, Cecil LD, Olson ML, Dewild JF, Susong DD, Green JR, Abbott ML (2002) Atmospheric mercury deposition during the last 270 years: a glacial ice core record of natural and anthropogenic sources. Environ Sci Technol 36: 2303–2310.

Schwesig D, Matzner E (2000) Pools and fluxes of mercury and methylmercury in two forested catchments in Germany. Sci Total Environ 260: 213–223.

Simola H, Lodenius M (1982) Recent increase in mercury sedimentation in a forest lake attributable to peatland drainage. Bull Environ Contam Toxicol 29: 298–305.

Sobanska M (2005) Wild boar hair (*Sus scrofa*) as a non-invasive indicator of mercury pollution. Sci Total Environ 339: 81–88.

Solonen T, Lodenius M (1984) Mercury in Finnish sparrowhawks *Accipiter nisus*. Ornis Fennica 61: 58–63.

Solonen T, Lodenius M (1990) Feathers of birds of prey as indicators of mercury contamination in southern Finland. Holarct Ecol 13: 229–237.

St. Louis V, Rudd JWM, Kelly CA, Beaty KG, Flett RJ, Roulet NT (1996) Production and loss of methylmercury and loss of total mercury from boreal forest catchments containing different types of wetlands. Environ Sci Technol 30: 2719–2729.

Steinnes E, Berg T, Sjøbakk TE (2003) Temporal and spatial trends in Hg deposition monitored by moss analysis. Sci Total Environ 304: 215–219.

UNEP (2002) Global mercury assessment. UNEP Chemicals, Geneva, Switzerland.

Urba A, Kvietkus K, Marks R (2000) Gas-phase mercury in the atmosphere over the southern Baltic Sea coast. Sci Total Environ 259: 203–210.

Verta M, Tolonen K, Simola H (1989) History of heavy metal pollution in Finland as recorded by lake sediments. Sci Total Environ 87–88: 1–18.

Voigt HR (2004) Concentrations of mercury (Hg) and cadmium (Cd), and the condition of some coastal Baltic fishes. Environmentalica Fennica 21: 1–22.

Wängberg I, Munthe J, Berg T, Ebinghaus R, Kock HH, Temme C, Bieber E, Spain TG, Stolk A (2007) Trends in air concentration and deposition of mercury in the coastal environment of the North Sea Area. Atmos Environ 41: 2612–2619.

Watras CJ, Morrison KA, Regnell O, Kratz TK (2006) The methylmercury cycle in Little Rock Lake during experimental acidification and recovery. Limnol Oceanogr 51: 257–270.