

SOURCES OF MERCURY IN THE ARCTIC

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Abstract. *Global and regional emission inventories of mercury are reviewed with special emphasis on the source regions with potential impact on the Arctic environment. These sources are located mostly in Eurasia and North America and emit almost 1300 t of Hg to the air annually. Combustion of fossil fuels to produce electricity and heat is the major source of Hg. Major portion of the element emissions from this source is in a gaseous phase. A small portion of Hg emissions in Eurasia and North America is deposited in the Arctic region, perhaps 60 to 80 t annually. Additional amounts of Hg in the Arctic air originate from natural sources, although it is very difficult to quantify them. A small decrease of anthropogenic Hg emissions is observed in Europe at present. These emissions are expected to increase again in the near future.*

1. Introduction

Mercury has been measured in environmental samples collected in the Arctic and sub-Arctic region (e.g. SFT, 1994). Although the most of these measurements were carried out in the aquatic and terrestrial environments, atmospheric Hg deposition was also studied, particularly in northern Scandinavia (e.g. Iverfeld, 1991). A summary of recently reported concentrations, including those in the northern regions and deposition rates has been prepared by Munthe (1993). The atmospheric pathway is clearly contributing to the contamination of other compartments of the environment by Hg, also in the Arctic region. Both, direct and indirect estimates of total deposition of the element proved this hypothesis. Direct estimates of the total deposition can be carried out on the basis of information on emissions which are then used in various dispersion models. The indirect estimates are based on the element concentrations in peat bogs, lake sediments, and mosses.

During the last few years long-range transport of Hg within air masses has been studied in Europe and North America (e.g. Petersen, 1993). It was concluded that Hg deposition predicted by a Lagrangian trajectory model applied in Europe agrees with observations from the Scandinavian deposition network (Iverfeld, 1991) within a factor of two (Petersen *et al.*,

1994). Even better agreement can be obtained through the improvement of mercury emission data, particularly with respect to the chemical speciation of the element.

Recent Hg measurements in peat bogs in Norway show a decrease of the element concentrations with depths corresponding to a time period of about 100 to 200 years (Steinnes and Andersson, 1991), thus, clearly indicating the effect of anthropogenic activities. The effect of long-range transport of Hg was also investigated using the element concentrations in moss as indicator of the atmospheric deposition over Scandinavia (Steinnes and Andersson, 1991). A south to north decreasing concentration gradient was observed except for the regions far north. The authors speculated that the enhanced dry deposition of Hg in the north can be related to a cold climate.

It can be concluded from the above mentioned and other studies that Hg can be transported within air masses to the Arctic and sub-Arctic regions and at least partially deposited there. How much of Hg can be brought to and deposited within these regions depends on many parameters. One of the most important parameters is the amount of emissions of the element, particularly in the regions with a potential impact on the Arctic environment. The purpose of this work is to assess emission fluxes of Hg with focus on emission regions having possible impact on the contamination of the Arctic and sub-Arctic air. Location of these sources is also described.

2. Global Fluxes of Mercury

Although quite extensive information exists on environmental and health effects of mercury and its behavior in the environment, much less information is available on the fluxes of the element to the air, water, and soils. The first quantitative worldwide estimate of the annual industrial inputs of 16 elements to the environment, including Hg has been prepared by Nriagu and Pacyna (1988). Summary of these estimates for the mid 1980's is presented in Table I. It can be concluded that the Hg emission to the air is comparable with direct inputs of the element to the aquatic environment and are almost a half of the direct releases to the terrestrial environment. No re-emission of Hg from the aquatic and terrestrial surfaces to the air was considered in these estimates.

Combustion of fossil fuels to produce electricity and heat is the major source of atmospheric emissions of Hg on a global scale. Major portion of Hg emissions from combustion of fuels is in a gaseous phase. In the combustion zone Hg present in coal or other fossil fuels evaporates in elemental form and then most likely a portion of it is oxidized while in the flue gases. The oxidized forms of Hg can be retained in modern flue gas cleaning systems. Mercury retained in fly ash (as well as in bottom ash) is then disposed on land or finds its way in a form of direct releases to the aquatic environment. It is very difficult to quantify these processes.

TABLE I

Global inputs of anthropogenic Hg to the environment (in 10^3 t/y) based on estimates by Nriagu and Pacyna (1988)

Environmental compartment	Source category	Emission
AIR	Combustion of fuels	0.7- 3.8
	Industrial manufacturing	0.1- 0.2
	Refuse incineration	0.2- 2.1
	Sub total	1.0- 6.1
WATER	Direct releases	0.1- 6.7
	Dumping of sewage sludge	0.1- 0.3
	Atmospheric deposition	0.4- 1.8
	Sub total	0.6- 8.8
SOIL	Direct releases	1.0-10.7
	Atmospheric deposition	0.6- 4.3
	Sub-total	1.6-15.0

Nriagu and Pacyna (1988) estimated that Hg in coal fly ash and bottom ash contribute up to 40% of the direct releases of the element to the terrestrial environment. As much as 40% of Hg entering the utility boilers in the United Kingdom is retained in flue gas control equipment and appears as liquid and solid wastes (in Pacyna, 1994). However, one should observe that

the flue gas control equipment, particularly the one for removal of gaseous pollutants is available mostly in the developed countries.

Refuse incineration seems to be the second largest source of Hg emissions to the atmosphere. Emission generation process for Hg during the incineration of wastes is similar to that during combustion of fossil fuels. However, more Hg in the oxidized form is expected from incinerators due to the higher content of chlorine in the wastes compared to fossil fuels.

While in the atmosphere, Hg undergoes various physical and chemical transformations (e.g. a review by Munthe, 1993) and finally is removed from the air by wet and dry deposition. Nriagu and Pacyna (1988) estimated that deposition on terrestrial surfaces is more than twice the deposition on aquatic surfaces (Table I).

Natural Hg emissions result from various processes including the off-gassing of Hg laden rock, and volatilization of Hg from soils and vegetation, as well as from various water bodies. Mercury enters the atmosphere as a result of forest fires, volcanic activity, and other biomass burning. Elevated ambient temperatures tend to increase the rate of Hg loss from soils. Very few direct measurements of the flux of Hg, by species, have been made to date under a wide enough set of meteorological conditions to allow for extrapolation of the old data to the present day. On the basis of recent information on re-emission rate from water and soil it can be suggested that the natural emissions of Hg are of the same order as the emissions from anthropogenic sources.

It should be noted that emissions from natural sources are difficult to distinguish from secondary emissions and diffusive re-emissions from anthropogenic sources. These include re-emissions of previously deposited Hg, emissions resulting from discharge into water bodies and from contaminated soils. Hence, it is more appropriate to differentiate between pre-industrial and post-industrial diffuse sources (e.g. Lindqvist, 1991).

3. Sources of Atmospheric Mercury in the Arctic and sub-Arctic

Obviously, only a part of worldwide emission sources is responsible for the contamination of the Arctic environment. However, no quantitative assessment is available to conclude in details on the origin of Hg in the Arctic air. Therefore, information on the origin of other trace

elements and sulfur has been utilized in this paper to suggest the possible sources and source regions for mercury in the Arctic air. Of particular interest in this case are trace elements emitted from the combustion of fossil fuels and the incineration of wastes. They include antimony, arsenic, cadmium, zinc, selenium, nickel, and vanadium. The differences in the fate and the environmental behavior of Hg and the above mentioned trace elements were taken into account.

A quantitative assessment of the contribution of carbonaceous fuel combustion emissions to the Norwegian Arctic haze layers has been made using the receptor modeling approach (Maenhaut *et al.*, 1989), and two different dispersion modeling approaches (Pacyna *et al.*, 1985; Akredolu *et al.*, 1994). The assessment showed that up to 50% of various air pollutants, measured in the Arctic during winter and summer, were from fuel burning. Sulfur could not be quantitatively assessed owing to uncertainties in the magnitude of natural marine biogenic emissions and their fluxes to the Arctic. There is an indication, however, that over 50% of the non-sea salt sulfate present in the Norwegian Arctic is from fuel burning (Maenhaut *et al.*, 1989), and a similarly high percentage has been estimated for Barrow, Alaska, as well (Li and Winchester, 1989). Taking into account the above discussion on the presence of combustion-related air pollutants in the Arctic one can conclude that also Hg can be transported to this region.

An important question is where are the source regions which may contribute to the contamination of the Arctic and sub-Arctic air by Hg. No such source-receptor relationship studies have been carried out for the element. However, these relationships were studied for other trace elements emitted during the combustion of fossil fuels, as well as for sulfur (e.g. Maenhaut *et al.*, 1989). The results of these studies can be summarized as follows. Emissions from sources in Eurasia contribute more than half of the air pollution measured in the Arctic. The major source regions include the Urals, the Kola Peninsula, the Norilsk area, and the industrial regions in central Europe. Emissions from the Urals and the Norilsk area may be more important for the contamination of air over Alaska and the Canadian Arctic, whereas sources in the Kola Peninsula contribute more to the Norwegian Arctic. The contributions of the European and North American emissions to Arctic air pollution seem to be lower than the contribution from the Russian sources. The European and North American emission are,

however, major contributors to the contamination of the sub-Arctic regions, such as northern Scandinavia (the European emissions) and northern part of Canada (the North American emissions).

Obviously, there are also local sources within the Arctic and sub-Arctic region. Combustion of fossil fuels to produce electricity and heat is one of the major source categories present in the region, particularly for atmospheric Hg. However, the impact of these sources on the contamination of the Arctic air is on local scale only (e.g. Ottar *et al.*, 1986).

4. Emissions of Mercury in Europe, North America, and Northern Asia

A summary of atmospheric emissions of Hg from anthropogenic sources in Europe, including the European part of Russia, the United States, and Canada is presented in Table II. The most recent European emission inventory for the element uses the 1987 statistical data (Axenfeld *et al.*, 1991). The inventory was prepared on the basis of emission factors and the results of these estimates are shown in Table II. These regional estimates were then compared with national emission data available from some countries. Both national and regional emission inventories indicate that the combustion of fuels, particularly coal, emits more than half of the atmospheric Hg in Europe (Pacyna, 1994). In some countries where combustion of coal is the predominant method to produce heat and electricity, the contribution of Hg emission from fuel combustion to the total national emission of the element is even bigger. A recent emission report from Poland concludes that fuel combustion generates more than 75% of the atmospheric Hg in the country (Hlawiczka, 1994).

Emissions from waste incineration in Table II are clearly underestimated. They represent emission data collected from 8 European countries only.

A national emission inventory of Hg and Hg compounds in 1990 has recently been prepared in the United States (US EPA, 1993). Emissions from combustion of fossil fuels contribute almost half of the US total emissions of Hg, followed by emissions from incineration of wastes. The estimates made in Canada are quite old and new data for 1993 levels are expected by the end of 1994 (UN ECE, 1994).

TABLE II

Atmospheric emissions of Hg from anthropogenic sources in Europe, the United States and Canada (in t/y)

Source category	Europe, 1987 (Axenfeld <i>et al.</i> , 1991)	The United States, 1990 (US EPA, 1993)	Canada, 1982 (Jacques, 1987)
1. Fossil fuel combustion	405	141	8
2. Industrial processes	275	32	16
3. Waste incineration	35	118	2
4. Other sources	11	19	5
TOTAL	726	310	31

Information on spatial distribution of Hg emissions is available for Europe and North America. European emissions are distributed within the UN ECE European Monitoring and Evaluation Programme (EMEP) grid system of 150 km by 150 km. This distribution is presented in Figure 1. Major emission sources are located in central and eastern Europe. Episodes of pollution transport from these regions to the Norwegian Arctic have been documented (e.g. Ottar *et al.*, 1986). In comparison, emissions in Scandinavia are low and the Hg concentrations measured in the Scandinavian environment are to a great extent due to the long-range transport from other parts of Europe. High emissions of Hg in the United Kingdom should also be noted.

Information on the location of major point sources of Hg emissions is available for the United States. In addition, the state-by state data are also estimated (US EPA, 1993). The largest emissions are estimated for the states around the Great Lakes, mostly due to coal combustion in the region. Atmospheric transport of pollution from this region northwards, with additional inputs from sources in Ontario has been a subject of preliminary studies (Clark, 1992). Combustion of fossil fuels in Alaska contributes about 1% of the Hg emissions from this source category in the country. A dozen of municipal waste incinerators and three sewage sludge incinerators in Alaska do not generate any significant emissions of atmospheric Hg.

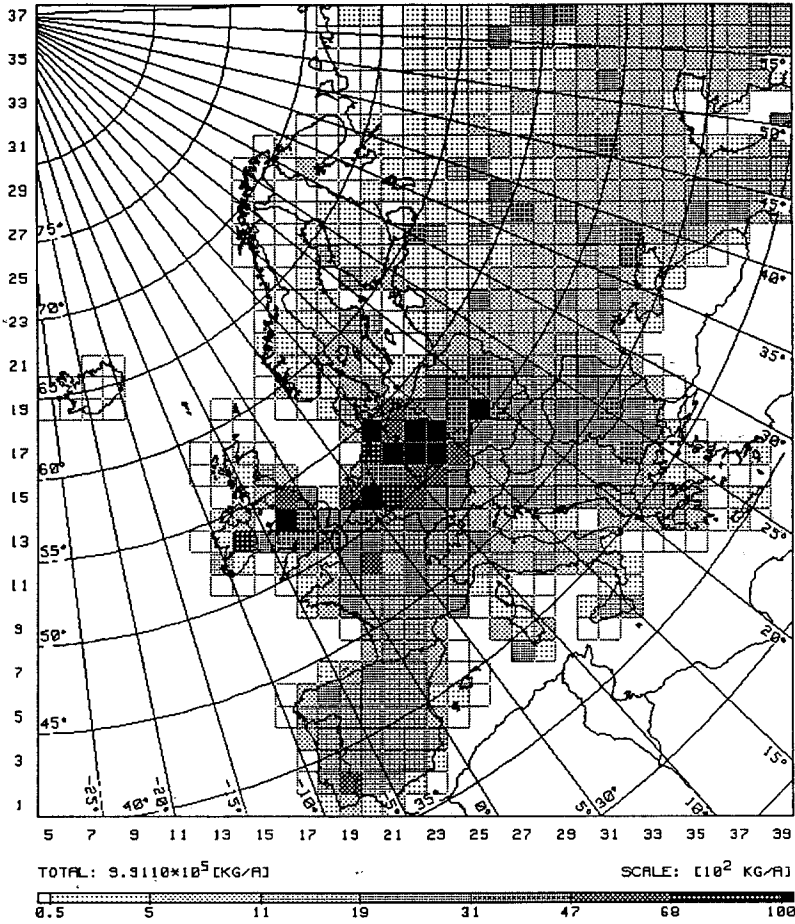


Fig. 1. Spatial distribution of total (anthropogenic and natural) emissions of mercury in Europe in 1987 within the EMEP grid system of 150 km by 150 km.

The Canadian emission data are available for all provinces, separately (Jacques, 1987). The largest emissions, about 10 t in 1982, were estimated for Manitoba, followed by North West Territories, Quebec, and Ontario. Huge emissions of about 3500 t/yr were calculated for natural sources in the country (in Voldner and Smith, 1989). These natural emissions are by far overestimated due to the overestimation of emission factors used in these calculations.

Less information is available on emissions of Hg in Asia, particularly in the northern part of the continent. Major emission source regions in the former Soviet Union and the emission quantities for several heavy metals were reviewed by Pacyna (1984). These data were used to assess the Hg emissions in this region. The results are presented in Table III. Again, the combustion of fossil fuels is the major source of atmospheric Hg emissions. The emissions from sources in the Ural dominate the emissions from other sources mentioned in Table III. The sources in the Ural include the production of non-ferrous and ferrous metals, chemical industry, and building material industry in addition to the combustion of fossil fuels (e.g. Pacyna, 1984). There are also other large source regions in the Asian part of the former Soviet Union, generating Hg and other trace element emissions to the air. They include the Kuznetsk area, the Fergana area, Caucasus, and the Baikal area. The location of these source regions in the southern part of the former Soviet Union raises a question whether sources a few thousand kilometers away can contribute to the Hg levels in the Arctic. Such very long range transport was suggested for some pesticides, e.g. lindane, used extensively in Asia and measured in the Arctic air (e.g. Pacyna and Oehme, 1988), and some crustal elements originating during dust storms in the Asian deserts and measured at elevated altitudes in the Arctic air (e.g. Pacyna and Ottar, 1989).

TABLE III

Mercury emissions from anthropogenic sources in the former Soviet Union at the beginning of the 1980's (in t/y)

Region	Combustion of fossil fuels	Industrial sources	Total
1. The Kola Peninsula	14	6	20
2. The Pechora Basin	6	2	8
3. The Norilsk Area	14	6	20
4. The Urals	110	44	154
5. The Yakutsk Area	22	2	24

The total emissions of Hg in Eurasia and North America are about 1300 t/y. No estimates were performed to assess what portion of these emissions enter the Arctic region and is

deposited there. Such calculations were made for other trace elements. An estimated 11 to 14% of antimony, arsenic, cadmium, lead, zinc, and vanadium, predicted to be entering the Arctic by the model used by Akeredolu *et al.* (1994) are deposited there. As much as 12% of the anthropogenic sulfur reaching this region from mid-latitudes is deposited in the Arctic (Jickells *et al.*, 1988). It was also estimated that up to 6% of the total emissions of arsenic, cadmium, lead, zinc, vanadium, and antimony in all of Eurasia is deposited in the Arctic (in UN ECE, 1994). About 6% of sulfur emissions in Eurasia is also deposited in the Arctic (Barrie *et al.*, 1989). If the same proportion is true for Hg, up to 60 t of the element emitted in Eurasia can be deposited from the air in the Arctic region annually. Additional quantities of the element would originate from anthropogenic and natural sources of Hg in North America.

5. Trends of Mercury Emissions

The decreasing trend of Hg levels in the atmospheric deposition in Scandinavia has been observed during the last few years (Munthe *et al.*, 1994). This trend was related to possible decline of Hg emissions, particularly in Central and Eastern Europe. As much as 30% decrease of these emissions can be expected. The decline of the economical growth in this part of Europe at the beginning of the 1990's, related to the transition of the centrally planned economies in these countries to the market oriented ones was suggested as the major reason for the possible changes of Hg emissions. No doubt, the lower consumption of fuels in Eastern Europe during the recent years has caused the decrease of Hg emissions.

Reasons for likely decrease of Hg emissions to the air in Western Europe and North America are different and relate mostly to the installation of control equipment removing sulfur and nitrogen compounds from exhaust gases in various industries. This equipment removes also other gaseous pollutants, including Hg.

An interesting question is whether the above mentioned decline of Hg emissions into the air is a permanent process or whether an emission increase can be expected in the near future. Munthe *et al.* (1994) conclude that the industrial decline in Eastern Europe and consequently lower electricity and heat demands is only a temporary process. Therefore, emissions of Hg are expected to raise again in the near future.

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