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MERCURY IN SNOW COVER AND RAINFALL IN FINLAND 1983–1984

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Mercury contents were measured in the snow cover and in 17 different rainfall events in Finland during the years 1983–1984. The mercury concentrations of snow ranged between < 10 and 50 ng l^{-1} in rural areas. In urban and industrial sites concentrations of up to 90 ng l^{-1} mercury were measured in snow. In rain water, mercury concentrations ranged between <10 and 30 ng l^{-1}. On the basis of the concentrations of mercury in snow cover the total deposition rate was estimated to be 3 $\mu g\ m^{-2}\ a^{-1}$ in Finland. The deposition and the concentration of mercury was higher in southern Finland than in the north and also in urban areas, indicating the influence of anthropogenic sources on airborne mercury.

Index words: Mercury, snow cover, rain water, deposition.

1. INTRODUCTION

The deleterious effects of mercury appear particularly in freshwater lakes and very high concentrations are measured in fish even in lakes with no known mercury pollution (e.g. Lithner 1978, Akielaszek and Haines 1981, Wren and MacCrimmon 1983, Björklund et al. 1984, Verta et al. 1986). In addition to weathering processes airborne mercury composes the main source of mercury in lakewater. Mercury precipitates directly into lakes or to the soil of lake drainage areas, from where it reaches the lake by the process of leaching.

The most important natural processes contributing to the flux of mercury to the atmosphere are windblown dust from dry continental areas and escape of gaseous mercury from water bodies, soils and vegetation (e.g. McCarthy et al. 1969, Lantzy and Mackenzie 1979). In addition, the mercury content of the atmosphere is increased by anthropogenic emissions, of which the most noteworthy are the burning of fossil fuels, mining and some other industrial processes (e.g. the chlor-alkali industry), and pesticides.

It has been estimated that approximately 20% of the mercury in the atmosphere is derived from anthropogenic emissions (e.g. Watson 1979, Brosset 1983a). Data from peat bogs (Pheiffer Madsen 1981) and sediment profiles (e.g. Ouellet and Jones 1983, Tolonen and Jaakkola 1983, Lindqvist et al. 1984, Rekolainen et al. 1986) indicate that mercury deposition has increased since the 19th century. The higher con-

centration of mercury in the atmosphere in the northern Hemisphere (Slemr et al. 1981) and in urban areas compared with rural areas (Ferrara et al. 1982) has been interpreted as a consequence of anthropogenic emissions.

Although a major part of the mercury emitted is transported over long distances from the source (e.g. Brosset 1983a), a distinct peak in deposition is frequently found in nearby areas (Lodenius and Laaksovirta 1979, Björklund and Norling 1980, Lodenius 1981, Lodenius and Herranen 1981, Lodenius and Tulisalo 1984).

In this study the content of mercury in snow and rain water was studied in Finland and mercury deposition estimates were made. The significance of local emissions and long-distance transportation on mercury deposition in Finland was also discussed.

2. EXPERIMENTAL METHODS

Snow samples were collected in May – April at 54 stations throughout Finland in 1983–1984. The sampling stations were situated in rural areas far from known mercury sources. Samples were taken from the snow profile into plexiglass tubes with an inner diameter of 6.3–11.0 cm. The snow was removed from the tubes into 5 l polyethene vessels. Both the sampling tube and the vessels were precleaned with concentrated nitric acid.

Snow samples collected in 1983 were melted, transferred to 500 ml Sovirel bottles and conserved (25 ml 5% $KMnO_4$ +15 ml conc. HNO_3) in district laboratories or in the central research laboratory. In 1984 snow samples were sent deep-frozen in polyethene vessels to the central research laboratory, where all samples were melted and conserved (5 ml 5% $KMnO_4$ +15 ml conc. HNO_3) and analysed at the same time. Before conservation the melted snow was filtered (pore size 1 mm) through an acid-washed polyethene funnel to remove hay and litter.

Snow samples were also collected from several locations in southern Finland during a snowfall on 22.3.1983 and at ten stations along a line running 0–80 km to the northeast of Helsinki on 17.1.1984.

The collection of 17 different rainfall events was carried out in urban and rural districts in southern, western and northern Finland. The collection apparatus consisted of a 26 cm diameter glass funnel which lead directly into a 500 ml acid-washed Sovirel bottle. To prevent mercury adsorption from air on the glass material of the bottles they were filled with deionized water after washing. This water was poured off immediately before sampling.

The conserved snow and rainwater samples were oxidized to Hg^{2+} in a 95°C water bath (two hours). Potassium peroxodisulphate and sulfuric acid were added to effect the oxidation. Samples were analyzed with the cold-vapour AAS-technique in which Hg^{2+} was reduced to Hg^{o} with stannous chloride. Blank bottles containing deionized water were handled in an identical manner.

3. RESULTS

3.1 Mercury in snow cover

The concentrations of mercury found in the snow cover in Finland in March 1983–1984 are shown in Figs. 1 and 2. The concentrations ranged between < 10 and 50 ng l^{-1} in 1983 and < 10 and 20 ng l^{-1} in 1984. The level of mercury was higher in 1983 than in 1984 especially in eastern Finland. In 1983 56 % and in 1984

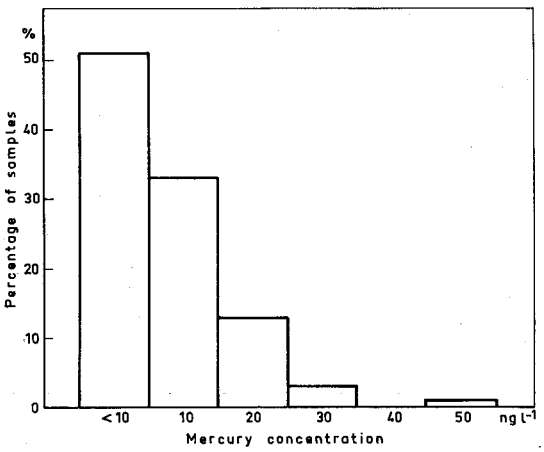

Fig. 2. The frequency of mercury concentrations in the snow cover 1983–1984.

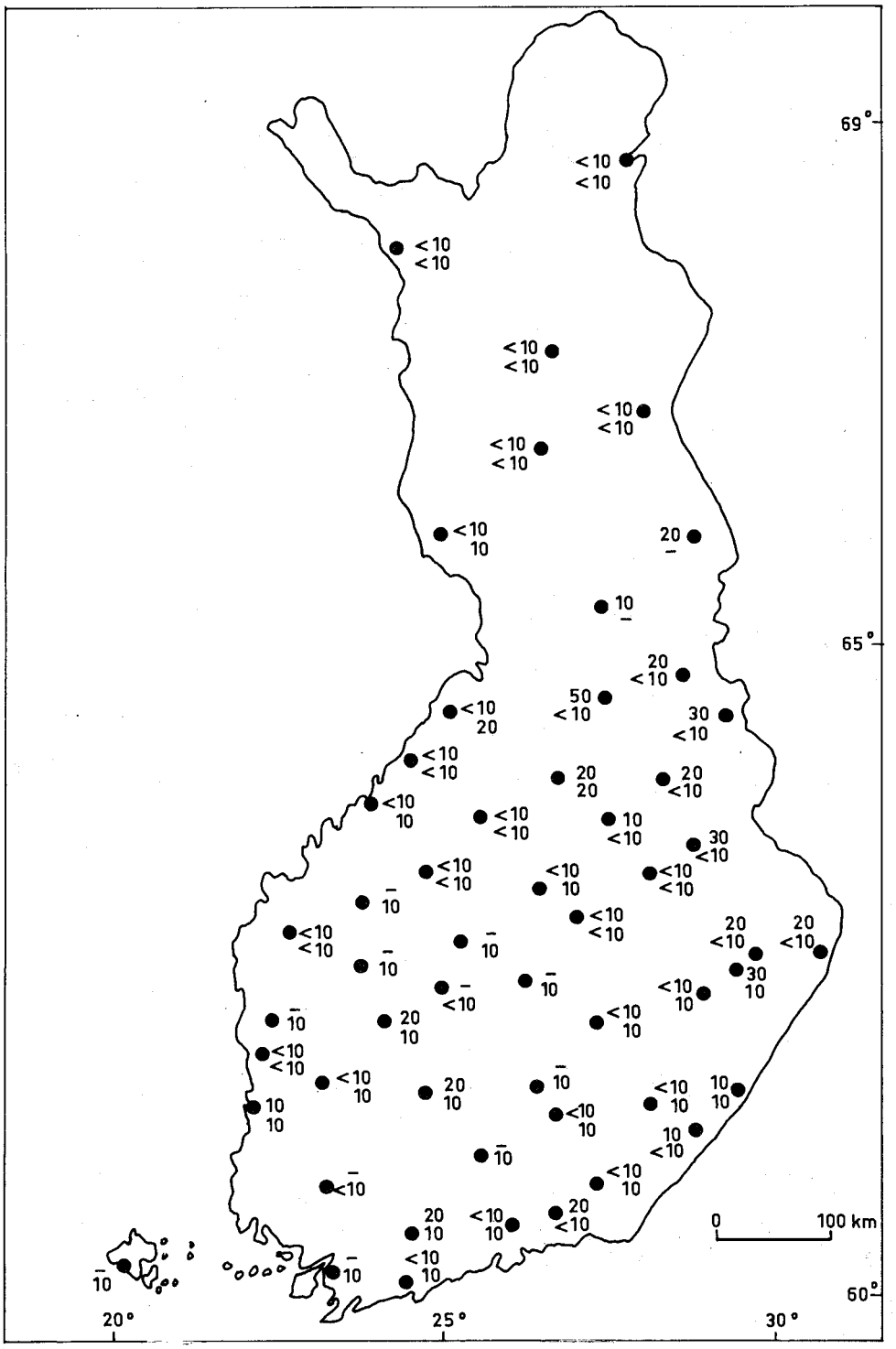

Fig. 1. Content of total mercury (ng l^{-1}) in the snow cover in Finland in 1983 (upper figures) and 1984 (lower figures).

46 % of the accepted measurements, were below the detection limit (10 ng l^{-1}). Eleven results were rejected because of contamination in 1983 and two in 1984. The decrease of the number of rejected results was a consequence of the more careful and analogous sampling method. The water content of the snow cover ranged between 10 and 200 mm in 1983 and 80 and 240 mm in 1984.

The content of mercury in the snow fall of 22.3.1983 is presented in Fig. 3. Five samples were collected in urban areas and one in a rural area. The concentrations ranged between < 10 and 90 ng l^{-1} and were higher in the southernmost and urban areas.

The concentration of mercury in snow along a line running 0–80 km to the northeast of Helsinki ranged between < 10 and 70 ng l^{-1} and was at its highest in the central areas of Helsinki city (Fig. 4). At the moment of sampling the thickness of the snow cover varied greatly (10–60 cm). In urban (coastal) areas the snow was old and partly melted and had refrozen at the time of sampling.

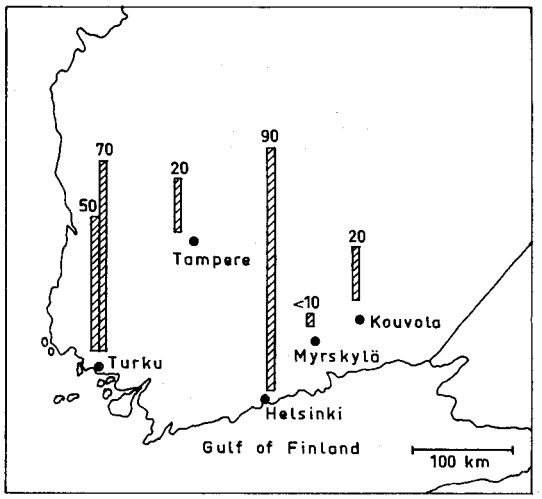

Fig. 3. Content of total Hg (ng l^{-1}) in the snow fall of 22.3.1983. The Myrskylä sampling site is rural, others are situated in urban areas.

3.2 Concentrations of mercury in rainwater

The concentrations of mercury in samples collected from rainfall events are given in Table 1. The concentrations in rainwater ranged between < 10 and 30 ng l^{-1}. A total of 17 samples were analyzed, of which eight contained mercury at levels below the detection limit (10 ng l^{-1}).

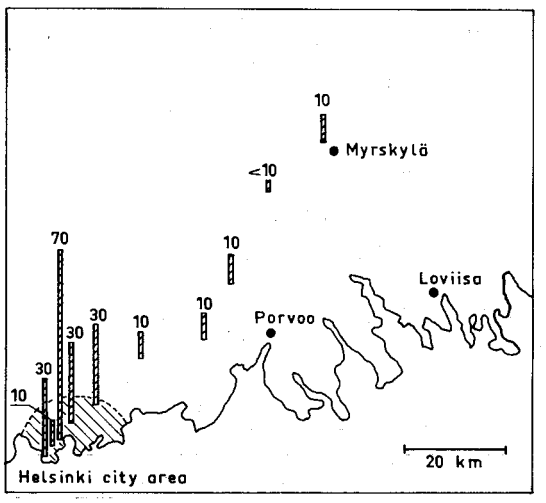

Fig. 4. Content of total Hg (ng l^{-1}) in the snow cover north-east of the City of Helsinki, 17.1.1984.

Table 1. The concentration of mercury in rainwater in 1983–1984.

Study area	Date	Hg (ng l^{-1})
Southern Finland, urban	21.10.1983	20
	1.11.1984	10
	15.6.1984	< 10
	1.7.1984	10
	2.7.1984	30
Southern Finland, rural	9.5.1984	10
Western Finland, rural	22.9.1983	10
	23.9.1983	< 10
	19.10.1983	< 10
	20.10.1983	< 10
Northern Finland, urban	9.9.1983	< 10
	6.10.1983	< 10
	7.10.1983	< 10
	10.10.1983	20
	19.6.1984	10
Northern Finland, rural	19.9.1983	< 10
	16.7.1983	20

4. DISCUSSION

4.1 Sampling and analytical methods

The risk of contamination is the most important difficulty in mercury sampling. The intercalibration tests carried out in district laboratories demonstrated that the possible sources of contamination are carelessly washed sampling equipment, reagents used in conservation, deionized water used in blank samples and also the laboratory air, which may contain mercury from reagents and analytical apparatus used in other analyses. To avoid contaminations a more careful and analogous sampling method was used in 1984 and the number of the apparently contaminated samples decreased.

Depending on the sampling site, snow contains various amounts of hay and litter (e.g. needles), which causes inaccuracy in the results. When melting snow samples it was also observed that soot and ash particles in snow adsorbed to the sides of the vessels and could not be removed with concentrated acid. As a consequence, the true mercury content in snow may be higher than that measured since soot and ash particles have been found to contain mercury (Brosset 1981, 1982, 1983a,b, Ferrara et al. 1982).

The wide range (approximately 10—100 ng l^{-1}) in mercury concentrations in blank samples disturbed the interpretation of results in 1983. To avoid this in 1984, the snow samples were melted and conserved in the central research laboratory. The mercury contents of blank samples were tested several times with varying additions of reagents (Table 2.).

Since no mercury was found in deionized water it could be assumed that the mercury in blank samples originated from reagents. The most probable source of contamination, namely the solution of $KMnO_4$, did not however contain mercury, as can be seen from the results of the test performed in January. This implies that the probable source of mercury in the blank samples was other reagents.

The reproducibility of the blank samples was however very good (Table 2.) and the mercury concentrations were calculated as differences between the sample and the blank. When estimating mercury deposition from the snow results in 1984 the mercury content was calculated with an accuracy of 1 ng l^{-1}. Because the content in blank sample was usually somewhat greater than the calculated difference, the results of mercury deposition reported at this accuracy were somewhat uncertain.

4.2. Mercury concentrations in snow and rainwater

The mercury content in rainwater (< 10—30 ng l^{-1}) was of the same magnitude as found elsewhere in recent studies. The lowest mercury contents measured have been about 1 ng l^{-1} (Matsunaga and Goto 1976), the normal range being 5—100 ng l^{-1} (e.g. Fogg and Fitzgerald 1979, Tomlinson et al. 1980, Brosset 1981, 1983a,b, Ferrara et al. 1982). The measured content in snow (< 10—90 ng l^{-1}) also agrees quite well with the Hg content in snow in Sweden (1—100 ng l^{-1}, Brosset 1982, 1983a) and in Hokkaido, Japan (about 1 ng l^{-1}, Matsunaga and Goto 1976).

Previously, very high concentrations of mercury have been measured in Finland, e.g. 37 000 ng l^{-1} (Soveri 1979), < 200—1300 ng l^{-1} (Anon. 1979), 200—1800 ng l^{-1} (Soveri 1981) and 60 —540 ng l^{-1} (Paasivirta et al. 1984). It is obvious that there must have been some contamination of the samples with concentrations above 100—1000 ng l^{-1}. The risk of contamination and analytical difficulties have also been reported

Table 2. The content of mercury (ng l^{-1}) in replicate blank samples with various additions of reagents.

25 ml 5% $KMnO_4$ + 15 ml conc. HNO_3 January 1984	5 ml 5% $KMnO_4$ + 15 ml conc. HNO_3 January 1984	5 ml 5% $KMnO_4$ + 15 ml conc. HNO_3 March 1984
17	17	23
19	17	24
18	18	23
18	18	25
18	18	23

elsewhere, e.g. in studies of mercury in ice profiles in Greenland (Appelquist et al. 1978) and in international intercalibrations (Olafsson 1982).

In spite of the aforementioned greater risk of contamination in snow samples in 1983, it is evident that mercury content in the snow cover was higher in 1983 than in 1984, especially in eastern Finland.

In both years the mercury content of snow was somewhat higher in southern Finland, where the concentration commonly exceeded the detection limit of the analytical method (10 ng l^{-1}). In northern Finland only a few results over this limit were found. Therefore at least some of the mercury in snow is of anthropogenic origin, from southern Finland or from western and central Europe. This conclusion is supported by the work of Brosset (1981, 1982, 1983a,b), who found that about 20% of the mercury in the atmosphere is dependent on wind direction. Brosset reported higher mercury contents in the atmosphere over southern Sweden during periods of prevalent southerly winds.

Because of the physical and chemical properties of mercury its residence time in the atmosphere is rather long and thus the major part of the mercury in the atmosphere may arise from distant sources (Lindqvist et al. 1984). However, many observations have been made of higher mercury contents in the vicinity of local sources (e.g. Lodenius and Laaksovirta 1979, Björklund and Norling 1980, Lodenius 1981, Lodenius and Herranen 1981, Lodenius and Tulisalo 1984).

In southern Finland the mercury concentration in rainwater was higher in urban than in rural areas, whereas no clear differences were found between southern and northern Finland (Table 3.). However, the number of rainwater measurements was too small for more extensive conclusions. Higher mercury concentrations were also recorded in urban areas in the snowfall of 22.3.1983 and in a line running northeast from Helsinki. These data indicate that mercury emissions in urban areas are washed down rather effectively. This may be a consequence of the fact that mercury in waste incineration and coal burning emissions has been found to be mostly in a water soluble form (Brosset 1983a).

Table 3. The frequences of mercury concentrations in rainwater in northern and southern Finland.

Study area	< 10 ng l^{-1} n	10–30 ng l^{-1} n
Southern Finland		
urban	1	4
rural	3	2
Northern Finland		
urban	3	2
rural	1	1

4.3 Mercury deposition

Data of mercury concentrations in snow and rainwater give information of mercury deposition. However, rainwater contents reveal only wet deposition, whereas snow may also contain mercury from other sources, e.g. dust precipitation. It is also uncertain whether the snow surface can be compared to other surfaces (e.g. water, soil, vegetation) as a recipient of total deposition (Lindqvist et al. 1984).

Mercury deposition was estimated using the snow results of 1984. As about half of the results were below the detection limit of 10 ng l^{-1} the calculated results with the accuracy of 1 ng l^{-1} were used. Using this method the average mercury concentration of snow was found to be 5 ng l^{-1}. If the mercury in snow is regarded as an estimate for mercury in rain water too, and the total precipitation in Finland is 550 mm a^{-1}, the total deposition rate of mercury could be calculated to be approximately 3 μg m^{-2} a^{-1}. In northern Finland the deposition was about half this amount and in southern Finland 3–6 μg m^{-2} a^{-1}. The water content of snow does not, however, correspond to the amount of accumulated snow, because a great part of the snow may evaporate during the winter, especially in southern Finland (Kuusisto 1984). It is not known whether mercury is concentrated in snow or volatilized with water.

In Sweden the total deposition rate of mercury has been estimated to be 5–50 μg m^{-2} a^{-1} (Lindqvist et al. 1984), which is of the same magnitude as in the present work. Deposition rates of 30–200 μg m^{-2} a^{-1} estimated from peat bog profiles (Pheiffer Madsen 1981) in Denmark were higher but were in Finland of the same order (8 μg m^{-2} a^{-1}) as in the present study (Pakarinen and Häsänen 1983). When using bog profiles it must be taken into account that plants absorb mercury actively from the air (Huckabee 1973).

As all of the soot was not included in the analysis, it is apparent that the mercury deposition was somewhat underestimated. It has been approximated that 20% of the mercury in the atmosphere is adsorbed on soot particles (Brosset 1982). The mercury adsorbed on the sides of the sampling vessels can therefore be estimated as representing only a minor part of the total deposited mercury. However, Brosset (1984) has reported considerably higher mercury concentrations in rainwater with coincident high soot contents in the atmosphere. It is likely that the mercury adsorbed on soot particles deposits more rapidly than do other mercuric components in the atmosphere.

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TIIVISTELMÄ

Ilman kautta kulkeutuva elohopea joutuu vesistöihin joko suorana laskeumana veteen tai valuma-alueen maaperään, mistä se saattaa huuhtoutua vesistöihin. Osa ilmakehässä olevasta elohopeasta on peräisin antropogeenisistä lähteistä.

Tämän tutkimuksen tarkoituksena oli selvittää lumen ja sadeveden elohopeapitoisuus, arvioida elohopealaskeumaa ja antropogeenisten lähteiden osuutta ilman kautta kulkeutuvassa elohopeassa.

Luminäytteet kerättiin 54 asemalla eri puolilta Suomea sekä vuonna 1983 että 1984. Lisäksi luminäytteitä kerättiin 22.3.1983 sattuneesta lumisateesta sekä n. 80 km pitkältä linjalta Helsingistä koilliseen vuonna 1984. Sadevesinäytteitä kerättiin yhteensä 17 kpl vuosina 1983–1984.

Lumen elohopeapitoisuus vaihteli taustaalueilla $< 10–50$ ng l^{-1}, kaupunkialueilla 10–90 ng l^{-1} ja sadeveden $< 10–30$ ng l^{-1}. Keskimääräiseksi elohopealaskeumaksi arvioitiin luminäytteiden perusteella 3 μg m^{-2} a^{-1}. Laskeuma oli suurempi Etelä-Suomessa ja kaupunkialueilla, mikä osoittaa antropogeenisistä lähteistä tulevan elohopean vaikutusta ilmalevintäiseen elohopeaan.

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