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Nationwide survey of airborne mercury in Finland

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Continuous measurements of total gaseous mercury (TGM) at an urban background station in Helsinki, Finland, were performed in 2006–2007. Additionally, a one-month campaign to measure TGM continuously from a moving car was organized in 2007, when several cities and industrial areas around Finland were surveyed. In Helsinki, a one-year average of 1.54 ± 0.20 ng m^{-3} was measured, which is about the global average for this persistent pollutant. The highest concentrations, up to 2500 ng m^{-3}, were measured during firing practice that took place next to the station. Seasonal and diurnal variation was studied, and trajectory maps were constructed to analyze mercury source regions. In the mobile measurement campaign, concentrations varying between 1.0 and 13.8 ng m^{-3} were measured. The highest concentrations (above 10 ng m^{-3}) were recorded close to former chlor-alkali plants that used mercury in their electrolytic production process.

Introduction

Mercury is a naturally-occurring element that is ubiquitous around the globe. It is a long-lived pollutant that can bio-accumulate in ecosystems and have adverse effects on human health, especially on children and the developing foetus. In the atmosphere, mercury is mostly present in its elemental form, Hg^0. Fish consumption is the primary source of mercury for many populations. Due to bacterial activity in waterbodies, inorganic mercury can be transformed into highly toxic methyl mercury, and end up in the fish humans consume. Thus, food safety authorities such as the US Food and Drug Administration and the Finnish Food Safety Authority (Evira) issue consumer advisories about mercury in fish and shellfish. Recently, mercury has gained much international attention due to the launch of new legislation (European Parliament, Council 2004), concerns in the Arctic (AMAP 2011) and, most recently, the UNEP Global Legally Binding Treaty on Mercury that is open for signature by governments in October 2013.

At the global scale, there are several anthropogenic sources of mercury, mainly coal combustion, small-scale gold mining, manufacturing of non-ferrous metals, cement production, waste disposal and caustic soda production (Pirrone *et al*. 2010). In Europe, major contributors are the combustion of coal in power plants and residential heat furnaces (~50%), the production of caustic soda using the Hg cell process (17%) and cement production (13%) (Pacyna *et al*. 2006b). Mercury can also be emitted from natural sources such as waterbodies, soil and vegetation. Re-emission of earlier-deposited mercury affects the mercury budget greatly, although it is extremely

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difficult to quantify (Pacyna *et al*. 2006a). It has been estimated that natural sources plus re-emission account for two-thirds of total Hg emissions, while anthropogenic sources explain the remaining one-third (Pirrone *et al*. 2010). In Finland, combustion in the energy, transfer and manufacturing industries and production processes were the main Hg emission sources in the 1990s (Melanen *et al*. 1999, Mukherjee *et al*. 2000). Nowadays according to the national pollutant emission database, a single steel-making plant, numerous power plants and the national chemical industry are the main Hg emitters. As estimated by Travnikov *et al*. (2012), domestic (*vs*. foreign) Hg emission sources contribute about one-third of the mercury anthropogenic deposition in Finland.

In urban environments, TGM concentrations were measured at different locations in e.g. Canada, China, Korea, Mexico, Sweden, Taiwan and the USA (Feng *et al*. 2004, Poissant *et al*. 2005, Stamenkovic *et al*. 2007, Kim *et al*. 2009, Li *et al*. 2009, Peterson *et al*. 2009, Rutter *et al*. 2009, Liu *et al*. 2010, Huang *et al*. 2012, Zhu *et al*. 2012, Jen *et al*. 2013). Measurements close to anthropogenic point sources were carried out to a lesser degree, but studies close to chlor-alkali plants, in particular, were made in Belgium, France, Sweden and the USA (Dommergue *et al*. 2002, Wängberg 2003, 2005, Landis *et al*. 2004, De Temmerman *et al*. 2007). However, data from urban and industrial environments in Europe are scarce; moreover, published data on Finnish background or urban mercury concentrations in the atmosphere are almost nonexistent.

This paper describes the TGM levels the Finnish public is exposed to in its daily life. Measurements were conducted for one year at an urban background station and also in several cities and industrial areas around the country using a mobile measurement method. These data sets for TGM in Finland are by far the largest so far published. Mobile air quality measurements had been conducted earlier. In Finland, studies with a "Sniffer" Mobile Laboratory Vehicle were carried out several times, but these campaigns did not measure mercury (Pirjola *et al*. 2004, 2009, 2012). To our knowledge, this is the first time TGM was measured continuously from a moving car.

Experimental sites and methods

Measurement sites

Total gaseous mercury in the ground-level air was measured between September 2006 and August 2007 at the Isosaari weather station (60°06´16´´N, 25°04´05´´E; WMO number 02988). The station is located on the Baltic Sea island of Isosaari about 8 km south-east from the shore of the Helsinki city centre (Fig. 1). The island (76 hectares) is covered mainly by coniferous forests. However, the station is located on a rocky cape with hardly any vegetation. Isosaari is governed by the Finnish Defence Forces and is closed to the public. The operation of the garrison, with occasional firing exercises, is practically the only human activity on the island. The site is categorised as an urban background station for Helsinki. The weather station was also operated by the military during the measurement period.

A nation-wide survey of the total gaseous mercury in Finland was accomplished with a one-month mobile measurement campaign (henceforth referred to as the MMC) in 2007. All major population centres and a large number of industrial sites were surveyed. The industrial sites visited included e.g. former chlor-alkali plants, power plants and pulp and paper mills.

Sampling

Sampling was conducted using a Tekran 2537A mercury analyser. This instrument collects samples in turns into two gold cartridges at 5-minute intervals, and analyses them continuously. While one cartridge is collecting a sample, the other is being desorbed and the mercury in the sample is analysed by atomic fluorescence spectrometry (AFS). The analyser collects only total gaseous mercury, not particles: these are removed from the sample flow with a PTFE filter. The method has a detection limit of 0.1 ng m^{-3}, repeatability of 2% and a measurement uncertainty of 10%. The analyser was calibrated daily with its internal permeation source at the Isosaari site. During the MMC, the analyser was calibrated at the start of each measurement day and, if needed, later

during the day. Even in the car, the calibrations were stable with a relative standard deviation of 5%. During the MMC, only two routine calibrations were omitted from the whole data set but later on the same day they were successfully carried out. A detailed description of the instrument can be found in Kyllönen *et al*. (2012). This instrument model was used successfully at a number of locations around the world, and has been found to give results comparable to those from other methods (Ebinghaus *et al*. 1999).

During the MMC, the Tekran mercury analyser described above was installed in a car. This measuring method does not necessarily give information about the typical TGM concentration level in the studied area, since the data are based on 5-min samples provided by Tekran. It rather gives valuable information about possible source areas around the country in a reasonably short time, and gives an indication of the Hg pollution level in various parts of Finland. When a possible source was identified, measurements were conducted in the nearby area for an extended time. Electrical power for the equipment was obtained from the car battery via a 12VDC/230VAC inverter. The sampling line inlet was above the car roof ahead of the position of the car's exhaust pipe. Thus ingestion of exhaust fumes into the analyser was prevented when the car was moving.

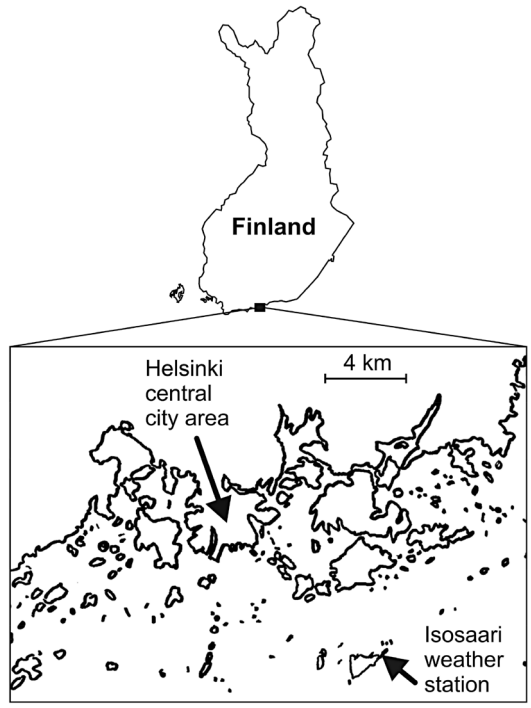

Fig. 1. Location of the measurement site at Isosaari.

Concentration fields

Trajectories, i.e. the paths of air parcels arriving at Helsinki, were utilized in an analysis of the long-range transport of mercury. They were calculated five days backwards using a three-dimensional kinematic FLEXTRA trajectory model (e.g. Stohl *et al*. 1995, Stohl and Seibert 1998), using numerical meteorological data from the European Centre for Medium-Range Weather Forecasts (ECWMF) MARS database. Trajectories were calculated at three-hour intervals, their arrival level at Helsinki being 950 hPa. Trajectories were applied to construct maps of mercury source regions. In this method, the concentration observations are distributed along the corresponding trajectory paths (Stohl 1996). In the first step, the measured TGM concentrations are distributed evenly along the path and averaged at every grid point crossed by several paths and thus multiple concentration values. Concentrations are scaled and redistributed again along the trajectory path following the methods developed by Stohl (1996). Redistribution continues until the change in the resulting concentration field is negligible. Finally, a chart is obtained where high-concentration regions refer to multiple occasions of air masses with elevated concentrations passing that region before arriving at Helsinki. Trajectory altitudes < 1000 m were included in order to allow only transport inside the boundary layer and continuous contact with the surface sources.

Results and discussion

One-year study of TGM in Helsinki

The hourly TGM concentrations in the ambient air at the urban background station in Helsinki (Fig. 2) remained mostly close to the global background value of 1.7 ng m^{-3} for the north-

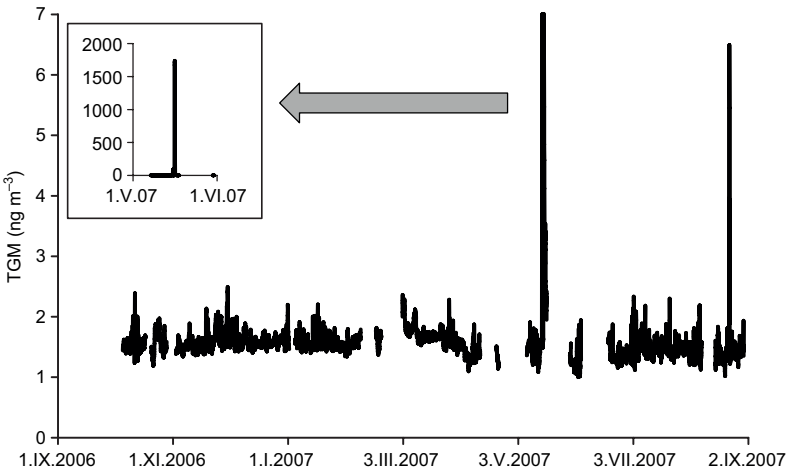

Fig. 2. Hourly TGM concentrations in Helsinki. Two peaks exceeding the scale, and presented in the smaller figure, represent data during the firing practice.

ern hemisphere (Slemr *et al*. 2003). During the campaign, the average hourly TGM concentration was 1.54 ± 0.20 ng m^{-3}. Closeness to both local pollution sources (the city of Helsinki) and neighbouring countries with high mercury emissions are reflected in the frequent peaks in the data. Hourly concentrations above 3 ng m^{-3} were seen only on three days during the project.

In August, a pollution plume arrived from the east, resulting in short-term concentrations of up to 14.9 ng m^{-3} and an hourly concentration of up to 6.5 ng m^{-3}. According to NOAA HYSPLIT backward trajectories, the air masses arrived from Russia and northeast Estonia, and reached mainland Helsinki just before reaching the station. In Estonia, a power generation complex with the world's largest oil-shale-fired thermal power plants is located in the northeastern part of the country. Oil shale burning is known to emit significant amounts of Hg into the atmosphere (Aunela-Tapola *et al*. 1998). In the TGM data of the Finnish EMEP station at Virolahti, we noticed that when elevated TGM concentrations are observed, the typical source area according to the NOAA HYSPLIT back-trajectories seems to be in northern Estonia (data not published). It remains unclear whether the high TGM values at Isosaari in August resulted from a pollution plume from local sources or from further afield, possibly Estonia.

In May, concentrations above 100 ng m^{-3} were detected during a two-day period. At first, they were thought to be due to an instrumental failure, since such concentrations are not likely to be measured at background stations (Munthe *et al*. 2003, Kim *et al*. 2005). Closer inspection of the activities at the site showed that, unbeknown to us, target practice took place next to the station at exactly the same time as the elevated concentrations were measured. This led to the conclusion that mercury might originate from the shooting activities. In the firing practice, five 12.7 mm anti-aircraft machine guns were operated. We believe that the TGM originated from mercury fulminate in the rounds shot during the training. Mercury fulminate is a primary explosive and has been used widely in the past. Today, mercury fulminate has been replaced in primers by more efficient chemical substances such as lead compounds, but in military training old rounds are still used. On the first day, the 5-min concentrations had risen to 280 ng m^{-3}, and then returned to and remained at the background level of 1.4–1.9 ng m^{-3} during the night, and then skyrocketed again in the morning shortly after the firing resumed. The highest concentrations occurred during the second day, with values above 1000 ng m^{-3} for several hours, maximum value being 2470 ng m^{-3}. After the second day of firing practice, the concentrations did not return to the background level until the sample line filter was replaced. We believe that the firing released huge amounts of particulate mercury in addition to TGM overloading the filter with particulate Hg (and possibly RGM trapped in the filter), which slowly

transformed from the particulate form into the gaseous state. Wallace (1998) showed that when mercury-containing ammunition is used, 86% of the mercury is released mainly via the muzzle, of which 17%–20% was particulate, which supports our conclusion. Although Wallace (1998) reported that only a small percentage of mercury was deposited on the gun operator, our results indicate a possible health concern for the conscripts and especially for the military staff who regularly attend firing practices. For example, in the study by Munthe *et al*. (2003) similar to ours Hg concentrations (1.4–26.9 μg m^{-3}) were measured in flue gas from five coal-fired (three hard coal and two brown coal) power plants. In a study by Frey and Hillamo (2011), TGM concentrations were measured in the raw flue gas of a coal-fired power plant in Helsinki and they were lower than those we measured during the firing practice maxima.

The average TGM concentrations remained stable throughout the year (Table 1), and their variability was small. In summer, when the consumption of energy is reduced resulting in less Hg emissions, a slightly smaller average concentration was recorded. The data collected during the firing practice were omitted from the spring average value, since the huge concentrations affected the average clearly (*see* Table 1).

The diurnal variation of TGM was calculated as monthly means for each hour, and proved to be very small. During the cold season (Oct–Mar), there is practically no diurnal variation while in the warm season (Apr–Aug) slight differences were seen. At midnight and during the early morning hours, the concentrations were typically ~0.2 ng m^{-3} higher than in the afternoon. This tendency is the opposite to that found at a forested background site in Finland (Kyllönen *et al*. 2012). In Helsinki, the concentration pattern in the warm season was opposite to that of air temperature and wind speed. In the cold season, the temperature and wind speed patterns were not as evident. On calm summer nights, mercury emitted from the sea and the soil around the station accumulates in the stable surface air and is then mixed in the morning as the wind speed increases and solar radiation breaks the surface inversion. In the cold season, the ice and snow covers inhibit this effect. Additionally, thermal mixing increases the boundary layer depth during the daytime and consequently dilutes TGM concentrations (Lee *et al*. 1998). Similar behaviour was observed e.g. in a rural region in England (Lee *et al*. 1998) and at urban sites in China (Feng *et al*. 2004), Sweden (Li *et al*. 2008) and the USA (Stamenkovich *et al*. 2007).

A clear majority (97%) of the hourly TGM concentrations were in the range of 1–2 ng m^{-3}, while 3% of the data were between 2 and 3 ng m^{-3} (Fig. 3). Hourly concentrations greater than 3 ng m^{-3} were rare, and if the firing practice data were omitted, they were almost nonexistent.

The TGM data were divided into wind sectors (Fig. 4) with the exception of the data collected during the firing practice. The lowest concentrations were found, quite unexpectedly, when the wind blew from the east, i.e., from Russia. Several pollutants, e.g., SO_2, NO_x and PAHs, typically arrive from this direction, St. Petersburg being one of the source areas (Vestenius *et al*. 2011). However, according to ESPREME (http://espreme.ier.uni-stuttgart.de), the St. Petersburg area is a significant source for mercury emissions. As stated earlier in this chapter, a pollution

Table 1. Seasonal variation of TGM concentrations (mean ± SD) and meteorological data measured in Helsinki from Oct 2006 to Aug 2007. Autumn = Oct–Nov, winter = Dec–Feb, spring = Mar–May, summer = Jun–Aug.

	Autumn	Winter	Spring	Summer
TGM (ng m^{-3})	1.57 ± 0.17	1.59 ± 0.12	1.59 ± 0.21 (6.1 ± 81.6)[a]	1.45 ± 0.29
Temperature (°C)	5.9 ± 4.3	–0.7 ± 6.5	4.9 ± 3.8	16.3 ± 2.7
Humidity (%)	87 ± 9	85 ± 8	79 ± 15	79 ± 12
Wind speed (m s^{-1})	7.8 ± 3.6	8.1 ± 3.4	6.1 ± 2.9	5.5 ± 2.8
Precipitation amount (mm)	216	149	84	172

[a] Firing practice data included.

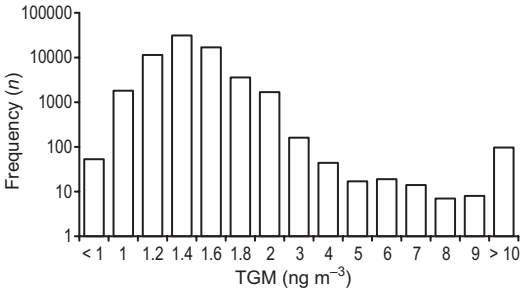

Fig. 3. Frequency pattern of 5-min TGM concentrations in Helsinki during the study period. Concentrations of 1–2 ng m^{-3} are at 0.2 ng m^{-3} intervals (e.g. a bar of 1.2 represents data in the range of 1.20–1.39 ng m^{-3}) while higher concentrations are at 1 ng m^{-3} intervals.

plume with concentrations above 10 ng m^{-3} was recorded during prevailing easterly winds. One should note that only 6.9% of the wind factors at the site are related to this low-concentration factor (90°–130°), while typical wind factors are from the west and southwest (45%, 210°–290°). Still, both the median and the 5 and 95 percentiles remained mostly low for the ESE sector.

The highest concentrations occurred with winds in the 300°–40° sectors (N factor) and the 160°–240° sector (S factor). The N factor is accounted for by domestic and especially, by close emission sources. Two coal-fired power plants and a natural gas power station are located in Helsinki, and these produce annually approximately 1000 MW of electricity and 1500 MW of municipal heating. The S factor contains polluted air masses from the highly-industrialised Baltic and European areas.

To study the source areas for high TGM concentrations in more detail, a TGM concentration field for Isosaari was calculated (Fig. 5). According to the trajectory analysis, the strongest source areas for Helsinki were located in the densely-populated and industrialized areas of central Europe, and also to some extent in the southern part of eastern Europe. Additionally, there seemed to be a source area in Russia. However, these Russian grid points with high concentration values were located on the periphery of the map and the exact location was thus uncertain. This source might explain the high 95th percentile for the 80° wind sector (Fig. 4). Low concentrations over the sea and Fenno-Scandinavia resulted from the lack of significant sources in these areas. The TGM concentration field (Fig. 5) resembled the spatial distribution map of mercury emissions over the EMEP domain in 2010 as modelled by EMEP (Travnikov *et al*. 2012), thus indicating that the hot spot sources detected by trajectory analysis are in line with the anthropogenic emissions.

Since mercury as an airborne pollutant has a rather long lifetime of 0.5–2 years (Schroeder and Munthe 1998), the Hg emissions from neighbouring countries affect the ambient air concentration levels in Helsinki in addition to domestic Hg emissions. According to ESPREME (http://espreme.ier.uni-stuttgart.de), the total annual emissions in 2000 of Hg in Finland, Estonia, Latvia, Lithuania, Poland and Russia were 0.60, 0.55, 0.15, 0.25, 25.6 and 66.1 t, respectively.

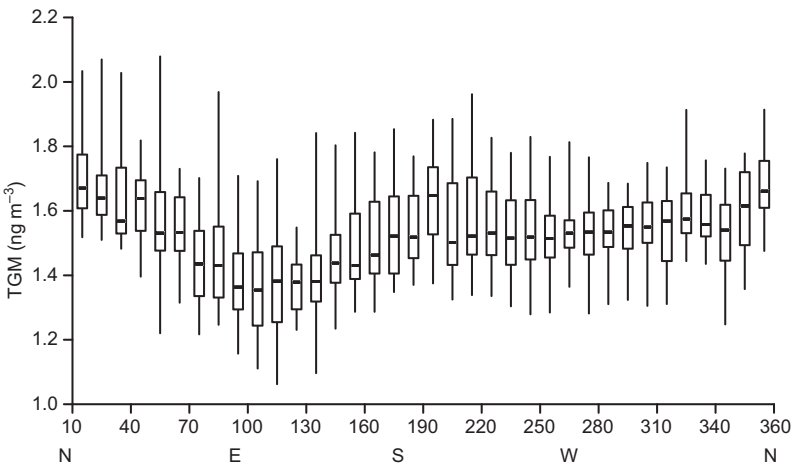

Fig. 4. TGM distribution in different wind sectors. The box represents the 25th and 75th percentiles, the line gives the median concentrations, and whiskers the 5th and 95th percentiles.

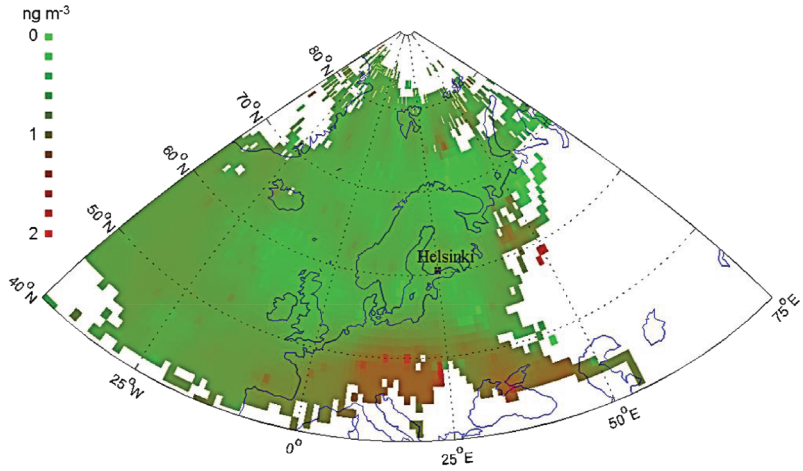

Fig. 5. TGM concentration field for Helsinki calculated from the hourly TGM concentrations.

Nationwide survey

In the mobile measurement campaign, all the major population centres and a large number of industrial sites were surveyed by online measurements from a moving car. Furthermore, most of the municipalities in the country were visited with the exception of the province of Lapland. The industrial sites visited included e.g. former chlor-alkali plants, power plants, chemical industry and pulp and paper mills.

The measurement days were mostly sunny or cloudy, while rain occurred a few times. During rain showers measurements were stopped to prevent water from entering the instrument through the horizontally-placed sampling tube on the roof of the car. In fog, measurements were conducted, but concentrations were typically lower than just before or after it. We believe that this phenomenon is caused by gaseous mercury binding to the tiny water droplets, as clouds act as a reaction vessel for aqueous chemistry, influencing the rates at which atmospheric Hg is incorporated into raindrops (Malcolm *et al*. 2003) and Hg is accumulated and concentrated in fog banks (Ritchie *et al*. 2006).

The majority of the measured concentrations remained at a low level, with no clear local influence (Fig. 6A). During the campaign, 89% of the data were below the global average of 1.7 ng m^{-3} with a median of 1.56 ± 0.91 ng m^{-3} for the whole data set. The 5th and 95th percentiles of the whole data were 1.13 and 1.97 ng m^{-3}, respectively, indicating a quite stable TGM concentration throughout Finland. This is due to the stable behaviour of this element and lack of significant local sources. It was noted that during different measurement days, the concentration levels changed a bit as can be seen in the bluish and greenish routes on the map (*see* Fig. 6A). This occurrence was also evident in the data from the measurements at the same location on different days. Due to changing wind patterns, the effect of long-range transported pollution plumes varied. The changes were small though, indicating an effective mixing of this long-life pollutant. Also changing weather (i.e. fog) affected the concentration level, as noted earlier. One must also remember that the instrument has a measurement uncertainty of 10%, so that changes of about 0.1 ng m^{-3} can also be due to instrumental performance.

Only 2% of the data exceeded 3 ng m^{-3}. These events were short and never represented the concentration level in an entire town. These peaks were measured close to an obvious source and remained high only in certain measurement places. Typically, a few hundred metres away from the location of the high TGM concentration, it declined back to the background level. This measurement method may have neglected some hot-spot areas due to unfavorable wind conditions or difficulties in driving to the optimal measurement location.

High TGM concentrations (here > 3 ng m^{-3}) were measured in certain areas of Riihimäki,

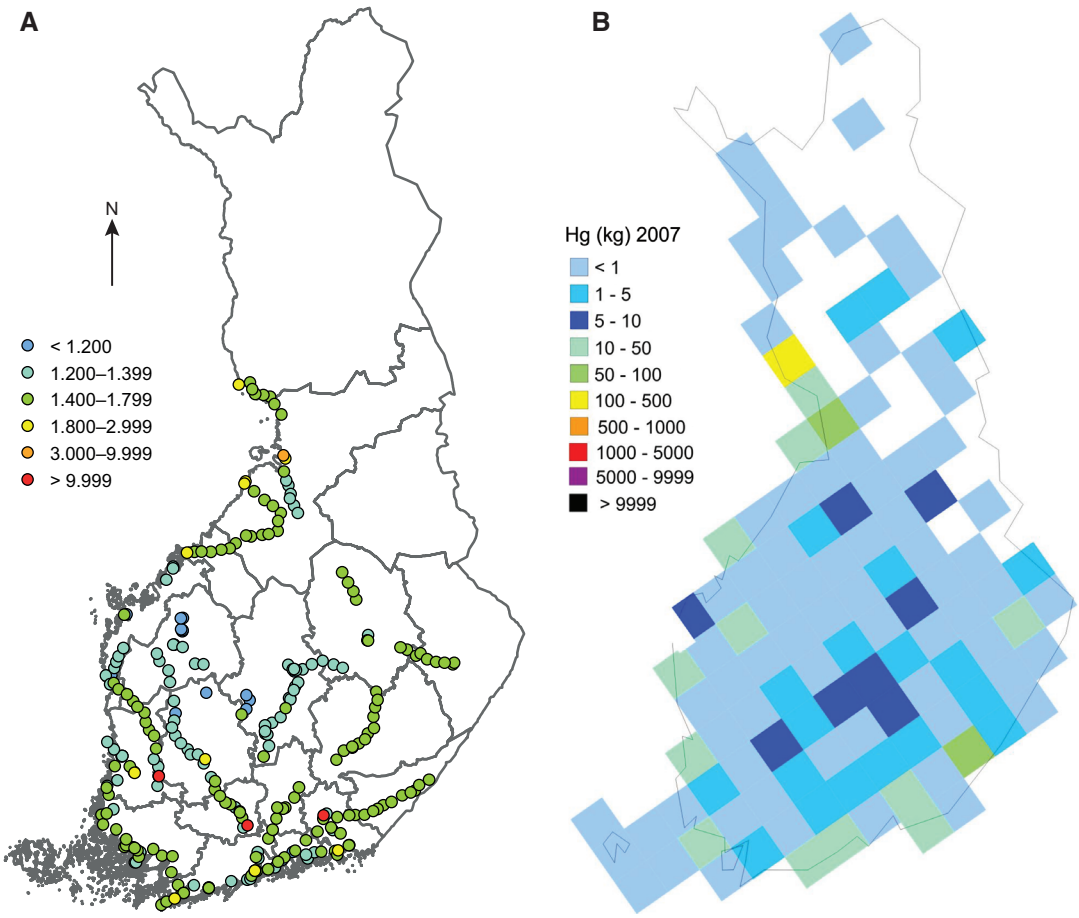

Fig. 6. (**A**) TGM concentrations (ng m^{-3}) measured during the mobile measurement campaign in Finland. Red dots are at Äetsä, Riihimäki and Kuusankoski (left to right) and the orange dot is at Oulu. (**B**) Spatial distribution of mercury emissions in Finland in 2007 (source Finnish Environment Institute 2013b; reproduced with permission from the copyright holder).

Äetsä, Kuusankoski and Oulu. With the exception of Riihimäki, the common factor for all these sites was proximity to a chemical industrial plant. In Riihimäki, concentrations in the range of 1.5–10.5 ng m^{-3} were measured in the vicinity of a toxic-waste disposal plant. We believe that the peak concentrations here may have resulted from toxic waste located temporarily in the outdoor area, since elevated levels were measured next to the building only and not in the area around it.

In Äetsä, concentrations reaching 14 ng m^{-3} were measured in a residential area next to the chemical plant and a former chlor-alkali plant. It remains undetermined whether the high concentrations were due to mercury emissions from the current chemical industrial processes, or a result of past chlor-alkali activities at the same site causing (a) strong re-emission of Hg from polluted ground in the area or (b) possible residual mercury in the stacks currently used for other industrial processes. In Kuusankoski, a maximum of 13.0 ng m^{-3} was similarly detected next to a chemical industrial plant and a former chlor-alkali plant. However, at this location the concentration levels varied widely in the vicinity of the plant. The environmental impact of these two former chlor-alkali plants has been noted earlier (Sarvala and Sarvala 2008, Verta *et al*. 2009). In the coastal surface and river sediments close to both locations, elevated levels of mercury were detected, and these results were considered to be from former chlor-alkali plants (Sarvala and Sarvala 2008, Verta *et al*. 2009) and in the case of

Kuusankoski also from the pulp and paper industry (Verta *et al*. 2009). Despite these findings, in the vicinity of another former chlor-alkali plant in Joutseno no elevated TGM levels were measured as compared with the background concentration of 1.5 ng m^{-3} in the area.

The only chlor-alkali plant remaining in Finland is located in an industrial area of Oulu. The area was not accessible by car; concentrations up to 4.6 ng m^{-3} were measured outside the industrial area. Much higher concentrations (around 50–250 ng m^{-3}) were measured in 2001 in the plumes of a chlor-alkali plant in Sweden, 70 m from the source (Wängberg *et al*. 2003). However, in a residential area 560 m away from the plant, concentrations of 1.4–40 ng m^{-3} were detected (mean = 3.5 ng m^{-3}) (Wängberg *et al*. 2005).

Elevated levels were also measured in Helsinki, Tampere, Harjavalta, Kokkola, Raahe and Tornio (up to 2.8, 2.0, 2.3, 2.2, 2.1 and 2.7 ng m^{-3}, respectively). These were all detected within industrial areas, or in one case in a residential area. In Helsinki, the areas around crematoria were carefully surveyed, but no increase in TGM concentrations were found during the campaign.

The Finnish Environment Institute maintains a national environmental monitoring database VAHTI containing the reports made periodically by individual facilities regarding their pollutant (e.g. Hg) emissions. The top five polluters include the steel and chemical industry, while most of the facilities reporting Hg emissions are power plants (data not shown). Globally, coal combustion is the main source of Hg emissions (Pirrone *et al*. 2010). We could not clearly connect power production with elevated concentration levels during our mobile measurement campaign. This is likely to be due to effective dilution resulting from the use of tall smokestacks in power plants.

Comparison with other studies

There are no published urban or industrial TGM data in Finland with which to compare our measurements. In June 1971, particle-bound mercury with an average concentration of 0.28 ng m^{-3} and maximum of 1.0 ng m^{-3} was measured in central Helsinki by the Department of Radiochemistry, University of Helsinki (Miettinen 1973, as cited in Mattsson and Jaakkola 1979). As mercury exists mostly in the gaseous elemental form Hg^0 (95%–99%), while mercury associated with particulate matter makes up only 0.2%–1.4% (Ebinghaus *et al*. 2008), we used a very rough estimating factor of 100 to calculate the TGM concentration in Helsinki in 1971. This would give an approx. mean value of 30 ng m^{-3} of TGM and a maximum of 100 ng m^{-3} of TGM in Helsinki during that year. These figures are about 20 times higher than the average TGM concentration and seven times higher than the maximum TGM concentration measured in Helsinki during our study. These high concentrations were due to the incineration of unsorted household waste and heating of buildings with coal (Mattsson and Jaakkola 1979). However, these values are uncertain, since the ratio between TGM and particle-bound mercury may then have been smaller, and the measurement method did not take into account the problem of mercury interactions with the particles collected on the filter.

Additionally, we made a comparison with recent TGM data from other countries (Table 2) to set the mercury situation in Helsinki into a global perspective. In Asia and Mexico, much higher TGM concentrations are found in urban areas as compared with the sites in Europe and North America. Our results from the one-year study in Helsinki and the mobile measurement campaign around the country, are located in the lower part of those given in the published studies (*see* Table 2).

Mercury emissions in Finland

According to the national mercury emissions reported by the Finnish Environment Institute (SYKE), during the last two decades Hg emissions did not change much. After 1990, the annual emission levels of Hg into the air have been below 1000 kg (Fig. 7, numerical values obtained from the Finnish Environment Institute 2013a). In recent years, emissions have remained rather steady, although unfortunately at the same level as in the early 1990s. The time series is not

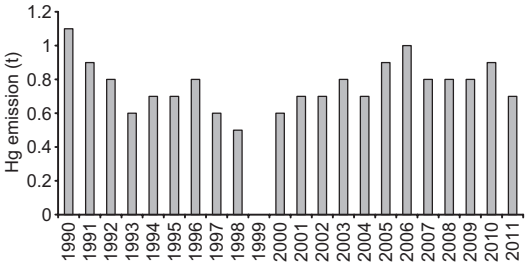

Fig. 7. Mercury emissions into the air in Finland in 1990–2011.

fully consistent due to the pending recalculation of the energy-sector emissions. According to the European Environment Agency (EEA), a clear decreasing trend in Hg emissions occurred in Europe in 1990–2010 (European Environment Agency 2014a). Additionally, EEA reports a 20% decrease in the Hg emissions in Finland during the same period (European Environment Agency 2014b). However, even so, this reduction is among the smallest in the EEA Member Countries. In the other Nordic countries, the reduction was substantial (60%–85%).

A map of Hg emissions in 2007 (Fig. 6B) published in Finnish Environment Institute (2013b) shows some resemblance to our map (Fig. 6A), although in certain emission areas we did not detect any increase in TGM concentrations. This is likely due to the limitations of our measurement method, addressed earlier in this article.

Table 2. Summary of TGM (or gaseous elemental mercury, GEM) measurements in urban areas and close to point sources.

Location	Site	Year	Period	Mean ± SD (min–max) TGM or GEM (ng m^{-3})	Reference
Helsinki, Finland	Urban	2006–2007	11 months	1.54 ± 0.20 (0.86–14.9)	Our study, part 1
Finland*	Industrial, urban, background	2007	1 month	(0.97–13.75)	Our study, part 2
Taiwan	Industrial/urban	2010	7 months	6.66 ± 1.42	Jen *et al.* (2013)
Nanjing, China	Urban	2011	1 yr	7.9 ± 7.0 (0.8–180)	Zhu *et al.* (2012)
Taiwan	Urban	2010–2011	1 yr	6.14 ± 3.91	Huang *et al.* (2012)
Detroit, USA	Urban/Industrial	2004	1 yr	2.5 ± 1.4 (0.36-25.6)	Liu *et al.* (2010)
Reno, USA	Urban	2004–2007	3 yr	1.6 ± 0.5 (0.5–6.4)	Peterson *et al.* (2009)
Seoul, Korea	Urban	2005–2006	1 yr	3.22 ± 2.10	Kim *et al.* (2009)
Mexico City	Urban	2006	1 month	7.2 ± 4.8	Rutter *et al.* (2009)
Gothenburg, Sweden	Urban	2005	1 month	1.96 ± 0.38	Li *et al.* (2008)
Belgium	Industrial	1999–2004	–	20 (max 150)	De Temmerman *et al.* (2007)
Reno, USA	Urban	2002–2005	3 yr	2.3 ± 0.6 (0.9–8.6)	Stamenkovic *et al.* (2007)
Quebec, Canada	Urban	2003	1 yr	1.65 ± 0.42	Poissant *et al.* (2005)
Bohus, Sweden	Industrial	2001–2003	10 weeks	55 (1.5–540)	Wängberg *et al.* (2005)
Bohus, Sweden	Industrial/urban	2001–2003	10 weeks	3.5 (1.4-40)	Wängberg *et al.* (2005)
Michigan, USA	Industrial (2)	2000	10 days	3.9 and 8.7 (1.9–77.6)	Landis *et al.* (2004)
Grenoble, France	Industrial/suburban	1999–2000	40 days	3.4 ± 3.6 (max 45.9)	Dommergue *et al.* (2002)

* Comprises several measuring locations around the country.

Conclusions

This study presents the total gaseous mercury (TGM) concentrations in the air in Finland measured during (1) a one-year measurement campaign at an urban background station in Helsinki, Finland, to measure TGM concentrations in the air and to study the behaviour of this pollutant; and (2) a mobile measurement campaign around Finland to study the regional variation of TGM in urban, industrial and background areas and to find possible Hg hot spots.

The hourly TGM values measured during the one-year urban campaign mostly remained close to the global background value, with an average of 1.54 ± 0.20 ng m^{-3}. Proximity to both local pollution sources (city of Helsinki) and neighbouring countries with high mercury emissions was reflected in the frequent peaks in the data. The seasonal variation of TGM concentrations was small, however slightly smaller than average concentrations were measured in summer due to less energy consumption. A diurnal variation was observed during the warm season (Apr–Aug) with a peak at night or during early morning hours. Values above 1000 ng m^{-3} were detected for several hours on one measurement day when, a firing practice took place next to the station.

During the mobile measurement campaign, the highest concentrations (10–15 ng m^{-3}) were measured in the immediate vicinity of certain chemical manufacturing plants formerly used in chlor-alkali industry, and a toxic waste disposal plant. In other industrial areas or residential areas close to industry, the TGM concentrations were less than 5 ng m^{-3}. In general, the measured concentrations were low, with a median of 1.43 ng m^{-3}, and elevated levels of TGM could not be connected to power plant emissions or crematoria.

The results from these campaigns indicate that the domestic anthropogenic emissions are only a minor source of mercury exposure to the general public in Finland.

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