



Atmospheric mercury distribution in Northern Europe and in the Mediterranean region

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Abstract

Mercury species in air have been measured at five sites in Northwest Europe and at five coastal sites in the Mediterranean region during measurements at four seasons. Observed concentrations of total gaseous mercury (TGM), total particulate mercury (TPM) and reactive gaseous mercury (RGM) were generally slightly higher in the Mediterranean region than in Northwest Europe. Incoming clean Atlantic air seems to be enriched in TGM in comparison to air in Scandinavia. Trajectory analysis of events where high concentrations of TPM simultaneously were observed at sites in North Europe indicate source areas in Central Europe and provide evidence of transport of mercury on particles on a regional scale. © 2001 Elsevier Science Ltd. All rights reserved.

Keywords: Mercury field measurement; Total gaseous mercury; Total particulate mercury; Reactive gaseous mercury; Atmosphere

1. Introduction

Mercury is an atmospheric pollutant with a complex biogeochemical cycle. The atmospheric cycling includes chemical oxidation/reduction in both gaseous and aqueous phases, deposition and re-emission from natural surfaces in addition to emissions from both natural and anthropogenic sources (Schroeder and Munthe, 1998). In the atmosphere, mercury exists in

three different forms: elemental mercury vapour (Hg^0), gaseous divalent compounds (Hg(II)) and associated with particulate matter (Hg(p)). The three different species have different atmospheric behaviour and lifetimes. Hg^0 is stable with a lifetime in the range 0.5–2 yr and is thus capable of distribution on a global scale. Hg(II) and Hg(p) are more readily deposited on local to regional scales via wet or dry processes.

Long-range transport has been shown to be an important source of mercury in many remote regions (Brosset, 1987; Iverfeldt, 1991; Petersen et al., 1995). These findings were the basis for discussions of

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international agreements on mercury emission control which led to the signing of a heavy metals protocol for mercury (as well as cadmium and lead) within the framework of the UN-ECE Convention on Long-Range Transboundary Transport of Air Pollutants (UN-ECE, 1998).

In order to derive cost-efficient control strategies for mercury, it is necessary to have quantitative information on emissions, air transport and deposition of mercury. An important part of this information is measurement data of atmospheric mercury species which can be used for source-receptor calculations using regional atmospheric models. Mercury is a part of the EMEP monitoring network in Europe but has until this year been a voluntary contribution. For this reason, information on atmospheric distribution and concentrations of different mercury species is not readily available.

This paper describes the outcome of co-ordinated measurements of atmospheric mercury performed within two research projects funded by the Fourth Framework Programme, Environment and Climate, of the European Commission; Mercury Over Europe (MOE) and Mediterranean Atmospheric Mercury Cycle System (MAMCS). Both projects are focussed on atmospheric cycling of mercury in Europe and contain measurements and modelling activities. The measurement activities in MOE are focussed on the northern part of Europe whereas MAMCS is focussed on the Mediterranean region.

2. Experimental

Four MOE/MAMCS co-ordinated measurement campaigns of 14 days duration were performed. The time schedule of each campaign is shown in Table 1. An additional fifth campaign was made within the MOE project. The locations of the measurement sites are shown in Fig. 1. Three airborne mercury species, TGM, TPM and RGM were simultaneously measured at all sites. Total mercury and methylmercury in precipitation was also measured. Some meteorological parameters such as wind speed and wind direction were measured at most sites during the campaigns. All methods employed

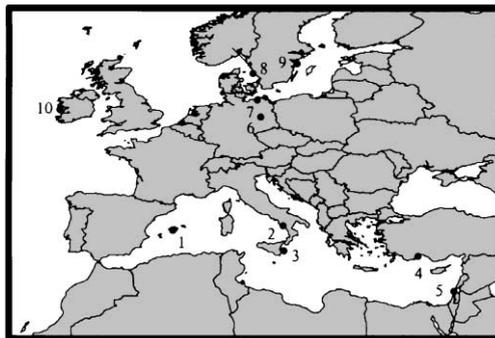


Fig. 1. The measurement sites: 1. Mallorca ($39^{\circ}40'30''\text{N}$, $2^{\circ}41'36''\text{E}$); 2. Calabria ($39^{\circ}25'\text{N}$, $16^{\circ}00'\text{E}$); 3. Sicily ($36^{\circ}40'\text{N}$, $15^{\circ}10'\text{E}$); 4. Turkey ($36^{\circ}28'12''\text{N}$, $30^{\circ}20'24''\text{E}$); 5. Israel ($32^{\circ}40'\text{N}$, $34^{\circ}56'\text{E}$); 6. Germany ($53^{\circ}08'34''\text{N}$, $13^{\circ}02'00''\text{E}$); 7. Germany ($54^{\circ}26'14''\text{N}$, $12^{\circ}43'30''\text{E}$); 8. Sweden ($57^{\circ}24'48''\text{N}$, $11^{\circ}56'06''\text{E}$); 9. Sweden ($58^{\circ}48'00''\text{N}$, $17^{\circ}22'54''\text{E}$); 10. Ireland ($53^{\circ}20'\text{N}$, $9^{\circ}54'\text{W}$).

for measurements of TGM, TPM and RGM were evaluated in a field intercomparison prior to the measurement campaigns. Results from the intercomparison are presented in (Munthe et al., 2001).

2.1. Sampling and analysis of total gaseous mercury (TGM)

The methods employed to measure this species varied among the sites. A Gardis automatic TGM analyser was used at site 3. Tekran automatic TGM analysers were used on sites 2, 6, 7 and 10. At sites 1, 4, and 5 a manual method was used. At the Swedish sites (sites 8 and 9) both the manual TGM method and automatic Tekran instruments were used. The results from the intercomparison (Munthe et al., 2001), showed that TGM measured with the different techniques are in excellent agreement, provided that careful site selection is made and stringent operational procedures are followed. The TGM measurement methods used in this work are briefly explained below.

The Tekran gas phase mercury analysers (Model 2537A) is an automatic instrument. The pre-filtered sample air stream is passed through gold cartridges where the mercury is collected. The mercury is then thermally desorbed in an argon gas stream and detected with an integrated atomic fluorescence spectrophotometer (AFS) (TEKRAN, 1998). The instrument utilises two gold cartridges in parallel, with an alternating operation modes (sampling and desorbing/analysing) on a predefined time base of 10 min. A sampling flow rate of 1.51 min^{-1} was used. Under these conditions, a detection limit of roughly 0.15 ng m^{-3} was achieved. A 47 mm diameter Teflon pre-filter protects the sampling cartridges against contamination by particulate matter.

Table 1
MAMCS-MOE multi-sites measurement campaigns

Campaign	Start date	Stop date
MAMCS-1/MOE-1	23 November, 1998	6 December, 1998
MAMCS-2/MOE-2	15 February, 1999	1 March, 1999
MAMCS-3/MOE-3	3 May, 1999	17 May, 1999
MAMCS-4/MOE-4	19 July, 1999	2 August, 1999
MOE-5	1 November, 1999	15 November, 1999

The accuracy and precision of this instrument has recently been assessed in measurements intercomparisons performed at various locations (Schroeder et al., 1995a, b; Ebinghaus et al., 1999).

The Gardis analyser was used for TGM measurements at site 3. The Gardis instrument is based on gold amalgamation and atomic absorption spectrometry (AAS) detection (Urba et al., 1995). The Gardis instrument operates with ambient air as carrier gas and does not require argon or helium for detection. The sampling is run at about 1 l min^{-1} with sampling times of 10 min. Under these conditions, a detection limit of about 0.1 ng m^{-3} is achieved. A 25 mm diameter PTFE membrane is used to protect the analyser gas inlet from contamination by aerosol particles. This instrument has been utilised in previous intercomparison exercises (Urba et al., 1999; Ebinghaus et al., 1999).

The manual gold trap method is based on gold trap amalgamation and subsequent analysis using AFS (Brosset, 1987; Bloom and Fitzgerald, 1988). Samples were collected on 14 cm long traps consisting of a 6 mm quartz tube with gold-coated 1 mm quartz glass grains. The airflow was kept at $0.3\text{--}0.4 \text{ l min}^{-1}$. The samples were brought to a mercury analysis laboratory near the site or shipped to IVL's laboratory for analysis. To keep blanks to a minimum the TGM traps were cleaned by heating in a purified gas stream at the sampling site, prior to sampling. A 21 h average TGM sample was collected each day starting 12:00 (local time) and ending 9:00 in the morning the following day. The manual TGM sampling technique allowed a detection limit of 0.01 ng m^{-3} for a 21 h sample.

2.2. Sampling and analysis of total particulate mercury (TPM)

A quartz glass filter trap for sampling of particulate mercury was developed, based on the AES mini trap developed by Lu et al. (1998). A quartz micro-fibre filter (Munktell MK 360), of 7 mm diameter is housed in a quartz glass tube of 140 mm length, see Fig. 2. The micro-fibre filter is supported by a pure Ni-screen support as is illustrated in Fig. 2. This device serves as both particulate trap and pyrolyzer for airborne particulate species. Air is drawn through the quartz tube at a flow rate of $4\text{--}6 \text{ l min}^{-1}$. After sampling, the Hg content is analysed via pyrolysis where the trap is heated to 900°C in a stream of argon. In this step, the Hg is reduced to Hg^0 and subsequently transferred to the gas phase and detected by conventional amalgamation—thermal desorption—AFS detection. This technique does not require any sample preparation (e.g. extraction/digestion), or manual handling of the sample that may create a risk for contamination. To bring down blanks to a minimum the TPM traps were cleaned by pyrolysis at the sampling site prior to sampling. A 21 h

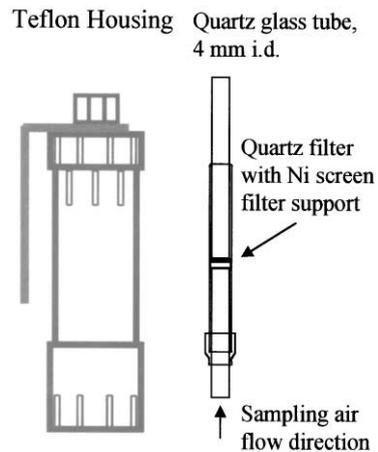


Fig. 2. Particulate trap with protective housing.

average TPM sample was collected each day starting 12:00 (local time) and ending 9:00 in the morning the following day. The TPM sampling technique allowed a detection limit of as low as 1.0 pg m^{-3} with 21 h sampling time.

2.3. Sampling and analysis of reactive gaseous mercury (RGM)

The RGM content of air is most likely mainly HgCl_2 . The term reactive mercury refers to divalent mercury species that easily undergoes reduction to elemental mercury in the analysis step. The RGM content of ambient air was measured using a mist chamber (MC), similar to that developed for RGM (Stratton and Lindberg, 1995). The procedure is as follows. Air is drawn through a Pyrex glass chamber of 100 ml total volume containing 40 ml diluted HCl solution. Part of the MC-solution is dispersed as a fine aerosol, by a nebulizer inside the glass chamber. A hydrophobic filter at the top of the MC separates the droplets from the air and allows the liquid to drain back into the chamber. Mercury(II) species adsorbed in the MC-solution are analysed after reduction to elemental mercury by SnCl_2 and AFS detection. The air sampling flow rate was $10\text{--}15 \text{ l min}^{-1}$. The detection limit obtained for a 6 h average sample was 1 pg m^{-3} . A 6–8 h average sample was collected every second Day.

2.4. Trajectory calculations

The NOAA Hybrid single-particle lagrangian integrated trajectory (HYSPLIT) model was used to calculate backward trajectories for the evaluation of results from MOE campaigns 2–5. The calculations were made interactively using the FNL meteorological

datasets which is also provided on the web (<http://www.arl.noaa.gov/ready/hysplit4.html>). Backward trajectories valid for site 8 during the MOE campaign 1 were obtained from EMEP/MSC-W, Norwegian Meteorological Institute, Norway, where calculations were made using the NWP model. The stability of the back trajectories were verified by expanding the starting area or by calculating at different starting heights.

3. Results and discussion

3.1. Comparison between Mediterranean region and Northwest Europe

Average TGM, TPM and RGM from the MOE and MAMCS campaigns, representing average values from Northwest Europe and the Mediterranean area at different seasons, are shown in Figs. 3–5. The average TGM concentrations varied between 1.6 and 2.4 ng m^{-3} with no significant seasonal variations. The relative uniform distribution found is reasonable since TGM predominately depends on the relatively stable global/hemispheric background concentration and only occasionally shows higher values, due to influence from major sources. Except for the first campaign, the

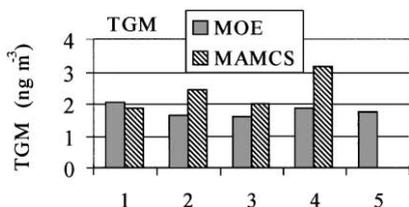


Fig. 3. Average TGM, TPM and RGM values obtained at campaign MOE 1–5 and MAMCS 1–4. The TGM value from the MAMCS campaign 4 should be regarded with some caution since it is based on measurements from two sites only.

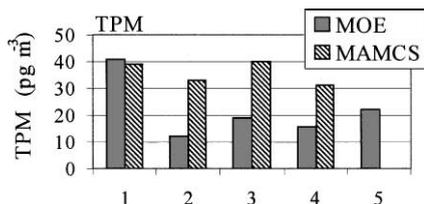


Fig. 4. Average TGM, TPM and RGM values obtained at campaign MOE 1–5 and MAMCS 1–4. The TGM value from the MAMCS campaign 4 should be regarded with some caution since it is based on measurements from two sites only.

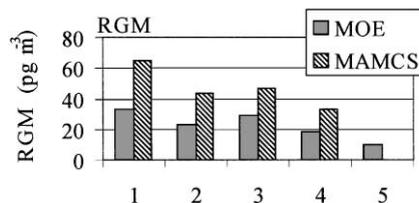


Fig. 5. Average TGM, TPM and RGM values obtained at campaign MOE 1–5 and MAMCS 1–4. The TGM value from the MAMCS campaign 4 should be regarded with some caution since it is based on measurements from two sites only.

data indicates that the TGM is slightly but significantly higher in the Mediterranean area than in North Europe.

Several reasons for this observation are possible. In the Mediterranean area, mercury enriched minerals are abundant. The world most abundant deposits are located in Almadén (Spain), Idria (Slovenia) and Monte Amiata (Italy) which have been exploited since ancient times for gold and silver extractions in Europe and North America (Nriagu 1989; Pirrone et al., 1996, 1998).

These mercuriferous areas may give rise to significant natural emissions both from diffuse sources and volcanoes (Ferrara et al., 2000a, b; Pirrone et al., 2001). Another possible explanation is enhanced emissions of mercury from the sea surface. Re-emissions from water surfaces are partly governed by sunlight and temperature and the warmer climate in the Mediterranean basin would thus enhance the fluxes from the water to the atmosphere.

Concerning particulate mercury, there seems to be a more pronounced difference between the two regions. Again values from the first campaign contradict this general trend. However, unusually high TGM and TPM concentrations were found at the MOE sites during the first campaign. From an air mass trajectory study this episode was confirmed to be due to transport of polluted air from Central Europe, (see below). The Mediterranean RGM concentrations also are higher than in Northern Europe. Thus, indicating a similar trend as for TPM and TGM. The reason for higher average TPM and RGM concentrations in the Mediterranean region may be due to higher emission rates and/or more active atmospheric transformation processes. Photochemical processes in the marine boundary layer may lead to enhanced oxidation of elemental mercury vapour which would lead to increased concentrations of RGM and possibly TPM, via gas-particle interactions. Another aspect influencing the atmospheric content of TPM and RGM is precipitation. The lower values in Northern Europe may be due to washout being a more efficient removal process in the north in comparison to that in the south.

3.2. Distribution of mercury species within Northwest Europe

The MOE stations 6 (Neuglobsow), 7 (Zingst), 8 (Rörvik) and 9 (Aspvreten) form a transect from south to north, as can be seen in Fig. 1. Previous measurement campaigns at these stations (Schmolke et al., 1999) have clearly indicated the presence of a south–north gradient for TGM with the highest values in the south. The present TGM data confirm this pattern. Average TGM from site 6–10 including values from all five MOE campaigns are presented in Fig. 6. The descending TGM trend as presented in Fig. 6 is also representative for the results from the five individual campaigns, suggesting that this trend is typical for all seasons.

The average TGM measured at Mace Head, Ireland (site 10) is also included in Fig. 6. Site 10 has been used as a reference for the input of Atlantic air to Northern Europe. The prevailing wind sector at Mace Head is 180° – 300° , representing clean Atlantic air. Only TGM data associated with the clean wind sector have been used. Hence, TGM values from the Mace Head station are that of the Atlantic background. Interestingly, the average TGM value from Mace Head is about 0.2 – 0.3 ng m^{-3} higher than those at the Swedish sites 8 and 9, but lower than at the German sites 6 and 7. The German sites are close to sources in Central Europe which explains the higher values found there (Schmolke et al., 1999). The lower values in Sweden are more difficult to explain. Transport from North America across the Atlantic may well occur, but such large scale processes should not affect only the Mace Head station, but all European sites. No local anthropogenic sources exist near Mace Head. Enhanced reemissions from the coastal sea may partly explain the observed differences. Other feasible explanations are enhanced removal processes like dry deposition to vegetation or atmospheric conversion to oxidised species when the air masses enter the European atmosphere.

As is discussed above the highest TGM values were obtained at the measurement sites in Germany. The same is true regarding TPM. Single 21 h average values

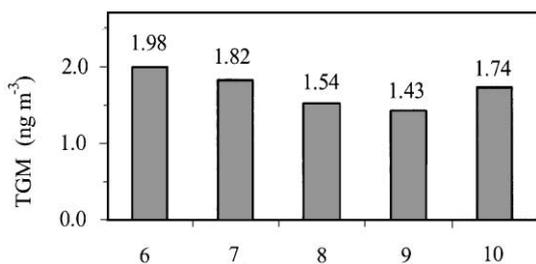


Fig. 6. Average TGM from the MOE measurements sites 6–10. The averages are based on the data from all 5 MOE campaigns.

as high as 150 – 200 pg m^{-3} were observed at the Neuglobsow (site 6) during an episode that occurred in the beginning of the first MOE campaign, November 1998. TPM values at Zingst (site 7) may also reach very high concentrations but are about 20%–30% lower on an average basis. Typical TPM values at the German stations (sites 6 and 7) are from 15 – 50 pg m^{-3} . Whereas most TPM values at the Swedish stations (sites 8 and 9) are below 10 pg m^{-3} . The lowest TPM values were found at Mace Head, where the TPM concentration often is lower than 5 pg m^{-3} .

A wind trajectory analysis showed that elevated TPM values, which occasionally are observed at the Swedish sites, are caused by transport from Central Europe. One event, during November 1999, with elevated TPM values at Neuglobsow, Zingst and Rörvik clearly demonstrates this phenomenon. The TPM data from this period is shown in Fig. 7. As shown in figure, the TPM concentration at the German sites 6 and 7 increases dramatically from relatively low values of about 20 pg m^{-3} , on the 2nd of November, to 74 and 110 pg m^{-3} , respectively, during a three days long period. After four days the concentrations have decreased to an intermediate level that lasted for one more day until it eventually goes further down. The same pattern is seen in the Rörvik data, but with about 24 h delay. The TPM values from Aspvreten also seem to be part in this trend although the effect is weaker. Back trajectories calculated for the Rörvik site show that the wind is coming from the Atlantic during the 2nd and 3rd of November. Then the wind path changes quite dramatically to southeast, and simultaneously the TPM at Rörvik increases from 6 to 31 pg m^{-3} . The wind direction remains from the southeast until the 9th of November when it changes back to westerly winds again. Simultaneously the TPM at Rörvik drops from 23 to 6 pg m^{-3} . One wind trajectory, calculated for Rörvik

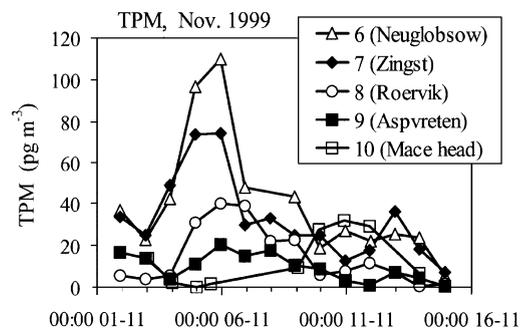


Fig. 7. Time series with 21 h average TPM obtained during the fifth MOE campaign. The measurements started 12:00 and ended 09:00 the following day. TPM data are plotted against mean time. Some of the measurements made at Mace Head were not strictly following this schedule.

at the day with the maximum TPM concentration is shown in Fig. 8. As is shown, the time needed for transport of air masses between Rörvik and Northeast Europe, a distance of 500 km, is about 24 h. This transport time fits exactly with the time delay in the TPM data as mentioned above. A trajectory analysis covering all campaigns clearly show that elevated TPM values at the Swedish sites, which occurred during two of the campaigns (November 1998 and 1999), actually are due to transport from Central Europe. The principal TPM source during this events is probably coal combustion or other major point sources in Central Europe.

As is seen in Fig. 7 TPM values at Mace Head are low during the beginning of the campaign and increases in the end. Back trajectories show that the airmasses reaching Mace Head during the first period with low TPM are originate in the Arctic with some influence from Northwest Europe. The higher values in the end of the campaign seem to be due to wind from the inland of Ireland. The evaluation of the Mace Head TPM data show that the content of TPM in airmasses coming from the North Atlantic Ocean are generally very low. Occasions with somewhat elevated TPM values seem to be connected to air coming from the inland of Ireland or from England to Scotland and the European continent.

Effects of long distance transport were also observed for TGM. During the measurements November 1999 elevated TGM values were observed simultaneously to the peak in TPM at sites 6, 7, and 8. No long distance

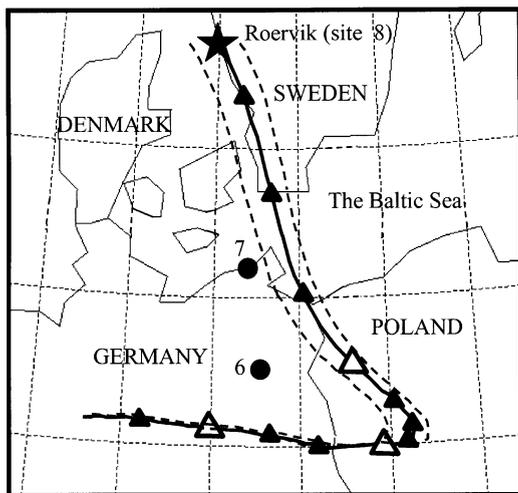


Fig. 8. 82 h back trajectories starting at Roervik 22:00, 5 November 1999. The starting height was 20 m. Filled triangles mark 6 h time intervals and the open triangles indicate 24 h intervals. Dotted lines are back trajectories calculated with starting points 37 km west and 37 km east of the Rörvik site.

correlation in the RGM data has been observed, however. This is probably due to a more efficient depletion of RGM due to dry deposition or wash out by rain (Stratton and Lindberg, 1995). RGM is also likely to be scavenged by dry aerosols.

4. Conclusions

For the first time airborne mercury species have simultaneously been measured at 10 sites in the Mediterranean area and in Northwest Europe. The result indicates that average concentrations of airborne mercury species are higher in the Mediterranean area than in Northwest Europe. The concentration of elemental mercury in airmasses reaching Mace Head from the west are often higher in comparison to values obtained in Scandinavia. As can be expected, the content of particulate mercury in incoming air from the west is very low. The distribution of TPM within Northern Europe is influenced by meteorological conditions such as wind-flow. During certain wind conditions, particulate mercury may be transported from the source areas in Central Europe to Rörvik and Aspveten in Sweden some 500–800 km northward.

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