Tellus (2004), 56B, 213–222
Printed in UK. All rights reserved

TELLUS

Annual time-series of total filterable atmospheric mercury concentrations in the Arctic

By JULIA Y. LU[1]* and WILLIAM H. SCHROEDER[2], [1]*Department of Chemistry and Biology, Ryerson University, 350 Victoria Street, Toronto, Ontario, Canada M5B 2K3;* [2]*Environment Canada: MSC, 4905 Dufferin Street, Toronto, Ontario, Canada M3H 5T4*

(Manuscript received 28 July 2003; in final form 19 February 2004)

ABSTRACT

Total atmospheric mercury (TAM) is operationally defined as the sum of gaseous elemental mercury (GEM), gaseous oxidized inorganic mercury (GOIM), which is also called reactive gaseous mercury (RGM), and total filterable mercury (TFM). An annual weekly-integrated time-series of TFM concentrations in ambient air at Alert, Nunavut, Canada from March 1998 to March 1999 is reported in this paper. The observed TFM concentrations ranged from 0 to 0.37 ng m^{-3} with the highest value occurring in the spring after polar sunrise and the lowest in summer. The average values for the four seasons are 0.086 (March–May), 0.00085 (June–August), 0.0015 (September–November) and 0.013 ng m^{-3} (December 1998–February 1999) respectively. This pattern is anti-correlated to that of GEM concentration observed at the same location. The GOIM concentration, calculated using the equation [GOIM]=[TAM]−[GEM]−[TFM], is also anti-correlated with GEM concentration. The annual time-series of the TFM operational blank values shows the same temporal pattern as the TFM concentration, with values ranging from 0.00 to 0.011 ng m^{-3}. These results confirm that, in the polar regions, GEM oxidation and atmospheric deposition of mercury occur mainly in the 3-month period after polar sunrise. Incisive laboratory and field experiments are needed to either confirm or rule out the springtime GEM oxidation/conversion mechanisms proposed in this paper.

1. Introduction

Mercury and its methylated derivates are among the persistent, bioaccumulating and toxic substances present in the environment. Elemental mercury (Hg^0) is of great concern because it can be transported through the atmosphere far away from its emission sources and can be transformed, chemically and biologically, in the natural environment into much more toxic and bioavailable organomercury species (Ikingura and Akagi, 1999; Branfireun et al. 1999). Toxic methylmercury species can be readily accumulated in the food chain (Bowles et al. 2001) and ultimately reach humans through consumption of fish, shellfish or wild game (AMAP Assessment Report, 1998), hence accumulating in target organs and body tissues. More importantly, organomercury species can cause cell and neurological damage (Leong et al. 2001). Recent studies on lake and marine sediments have indicated a marked increase in Hg deposition to the Arctic ecosystem since the beginning of the Industrial Revolution (Hermanson, 1998; Lockhart et al. 1998; Asmund and Nielsen 2000), leading to elevated mercury levels that may affect the health of people who live in the Arctic regions (Bjerregaard and Hansen, 2000).

Both experimental data and model calculations show that the atmosphere is a major pathway for the introduction of Hg (along with other persistent toxic chemicals) into the Arctic environment (Jensen et al. 1997; Asmund and Nielsen, 2000; Cheng and Schroeder, 2000; Lu et al. 2001; Lin et al. 2001). In the atmosphere mercury can exist in the gas and/or the particulate (atmospheric aerosol) phase. For the gas phase, determination of mercury is at present operationally divided into two fractions: gaseous elemental mercury (GEM) and gaseous oxidized inorganic mercury (GOIM), also called reactive gaseous mercury (RGM), which consists mainly of one or more oxidized inorganic forms of mercury in the vapour phase. Mercury associated with airborne particles, such as dust, soot, sea-salt aerosols, ice crystals, pollution-derived solid/liquid aerosols, etc., can be in elemental (Hg^0) and/or oxidized forms, but the individual chemical species of mercury have so far not been differentiated analytically. Since the discovery of atmospheric mercury vapour depletion events (MDEs) and the strong correlation between surface-level GEM and tropospheric (marine boundary layer) ozone concentrations in the Arctic after polar sunrise (Schroeder et al. 1998), there has been a dramatic increase in research on the atmospheric behaviour of mercury in the Arctic

---

*Corresponding author.
e-mail: julialu@ryerson.ca

and in Antarctica. Mercury depletion events have now also been reported at other locations in the Arctic (Berg et al. 2003; Lindberg et al. 2001) and in the Antarctic (Ebinghaus et al. 2002), where simultaneous surface-ozone and GEM depletion events occur. We have presented evidence that the depletion of GEM is a result of photochemically initiated oxidation (by extremely reactive halogen-containing free radical intermediates intimately involved in the autocatalytic destruction of surface-level ozone) of GEM and that a substantial fraction of the oxidized forms of mercury can be recovered as particle-bound (filterable) mercury species (Lu et al. 2001). It was also expected, and has now been confirmed (Lindberg et al. 2002), that some of the Hg(II) species can exist in the gas phase. This paper presents the first annual time-series of "total filterable" mercury measurements at Alert and discusses possible mechanisms of transformation, transport/advection and removal (by dry deposition and other scavenging processes) of atmospheric mercury in polar environments.

2. Experimental

2.1. Total filterable mercury (TFM) determinations

The technique we previously developed (Lu et al. 1998) was used for sampling and analysis of total filterable mercury at Alert for a period of 1 year (March 1998–March 1999). This technique can be briefly described as follows: a miniaturized device is used for both collection and analysis (via pyrolysis) of mercury associated with airborne particulate matter. Filter discs, 6 mm in diameter, were cut from microquartz fibre filters (47 mm diameter, pore size <10 μm, Gelman) and used as collection media. All the sampling/analytical devices were thoroughly cleaned by heating them repeatedly at 450 °C for 5 min, with ultra-high-purity (UHP) argon flowing through at a rate of 200 ml min^{-1}, until a constant background signal (measured using atomic fluorescence spectrometry) was obtained. During sampling, as shown in Fig. 1, air was pulled through the device using a dual-stage pump (model DAA-V110-GB, GAST, Benton Harbor, MI) at rates ranging from 3.0 to 5.0 l min^{-1}. The sampling flow rate was measured and the air volume integrated by a mass flow meter with an attached totalizer (Hastings, Hampton, VA). The samples were collected in duplicate. After collection, the sampling devices, along with the collected airborne particulate matter, were shipped back to our laboratory and analyzed following the procedure described in Lu et al. (1998).

A dynamic mode field blank (see Lu et al. 1998) was collected for each batch of samples. A filter-pack assembly (47 mm diameter) with two Teflon membrane filters (pore size <0.2 μm), as shown in Fig. 1, was used to remove airborne particulate matter in the air stream before it entered the blank trap.

Calibration of the analytical system was achieved by injecting a known volume of air saturated with Hg^0 vapour at a predetermined temperature. The field blank values were subtracted from the analytical results ("raw data") obtained for the field samples.

2.2. Gaseous elemental mercury (GEM)

GEM concentrations were determined using a mercury vapour analyser (model 2537A, Tekran, Toronto) following the standard operating procedure (Steffen and Schroeder, 1999). During the sampling step, air was pulled through a Teflon membrane filter (47 mm in diameter, pore size <0.2 μm) at a rate of 1.5 l min^{-1} to remove the particulate matter. GEM present in the air steam is trapped on a gold cartridge through amalgamation. The amalgamated mercury is thermally desorbed during the subsequent analytical step and detected by cold vapour atomic fluorescence spectroscopy (CVAFS). The Tekran$^{\text{TM}}$ model 2537A is equipped with dual gold cartridges (arranged in parallel) to allow continuous sampling and analysis of GEM in air at ng m^{-3} levels. GEM

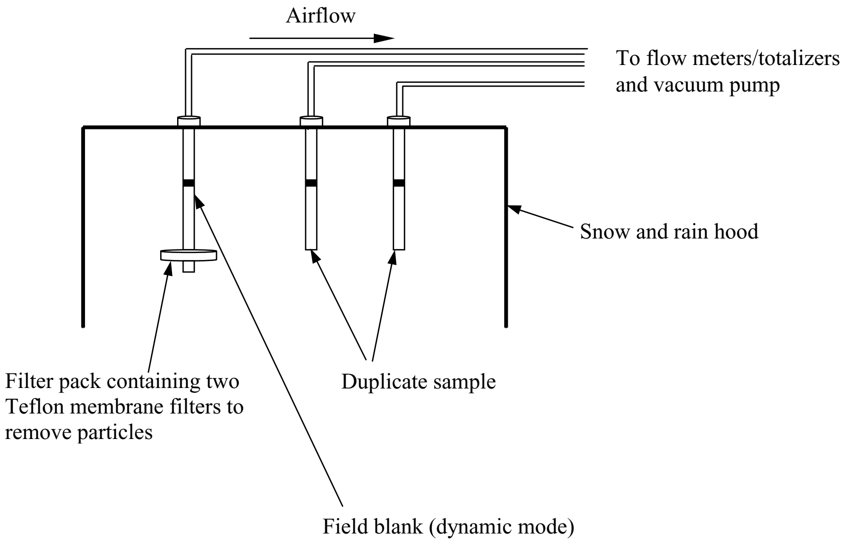

Figure 1. Experimental set-up for sample collection.

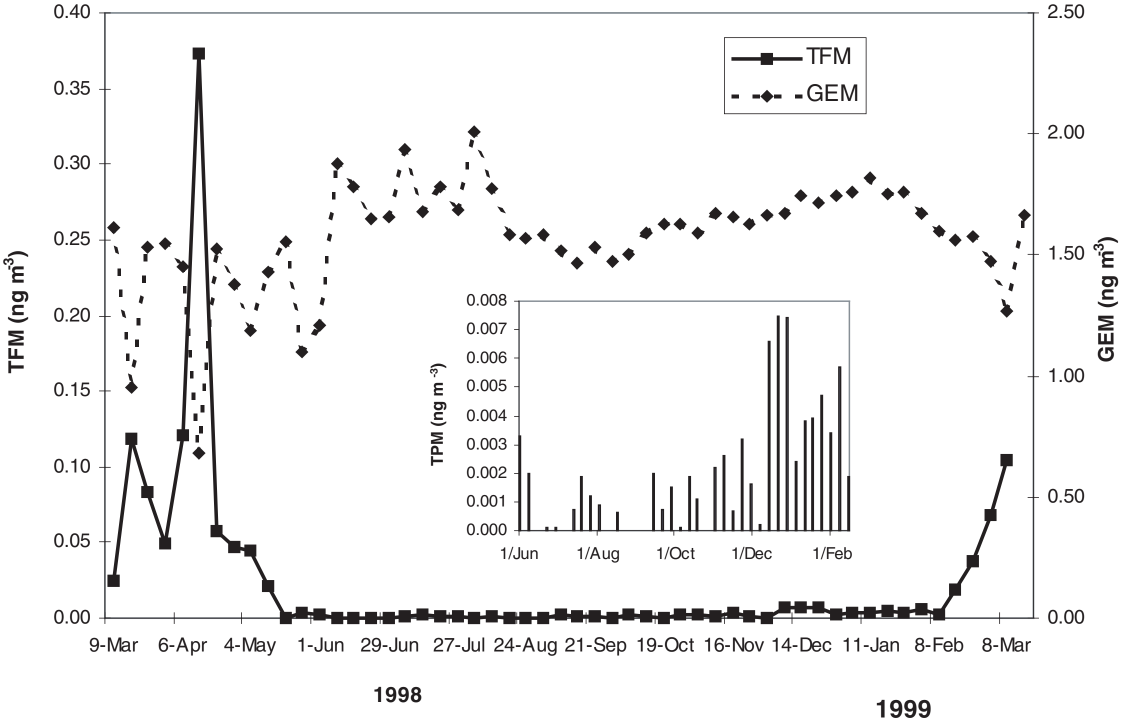

Figure 2. Annual time-series of total filterable mercury (TFM) concentrations and gaseous elemental mercury concentrations (weekly averages) at Alert, Nunavut, Canada, March 1998–March 1999.

concentrations are integrated over half an hour, but the results are presented as hourly averages to reduce the potential systematic error (due to possible cartridge bias).

2.3. *Total atmospheric mercury (TAM)*

The purposes of measuring TAM are (1) to study the atmospheric mercury balance during MDEs in the Arctic and (2) to calculate, by the difference between TAM and GEM, the sum of mercury associated with airborne particles and oxidized inorganic mercury in the gas phase (GOIM). A pyrolysis unit, taking in unfiltered air and heating it to 900 °C, converted all oxidized mercury species into GEM (Schroeder and Jackson, 1985; Wang et al. 1995), which was detected and quantified using a Tekran 2537A mercury vapour analyser. The mercury vapour analyser downstream of the pyrolysis unit, therefore, measured TAM concentrations.

3. Results

Figure 2 shows the year-long (March 1998–March 1999) weekly time series of TFM and GEM at Alert. The concentrations of TFM were blank corrected. The figure shows that: (1) TFM and GEM concentrations are inversely correlated; (2) TFM concentrations were highest in spring during GEM depletion events and lowest in summer. The annual average value of TFM is 25.3 pg m^{-3} and the average values for the four seasons are: 86 (March–May), ranging from 21–370 pg m^{-3}; 0.85 (June–August), ranging from 0.02–1.9 pg m^{-3}; 1.5 (September–November), ranging from 0.2–3.2 pg m^{-3}; and 13 pg m^{-3} (December–February, 1999) ranging from 1.9–19 pg m^{-3}. Our values for the period of June 1988 to February 1999 (see Fig. 2) agree well with those observed by Berg et al. (2001) at Ny-Ålesund, Svalbard. But from March to May, especially during major ozone and GEM depletion events, our TFM concentrations are appreciably higher. Figure 3 shows the annual time-series of TFM blank values derived from the dynamic mode of operation (Lu et al. 1998). The series displays a similar pattern as the TFM concentrations, with high blank values occurring during April and May. The blank values ranged from 0.1–15 pg m^{-3}, with an average of 2 pg m^{-3}, and the highest values occurred in the spring during the main period of GEM depletion events. The elevated blank values found in the spring could conceivably be a result of adsorption by the quartz fibre membrane filter disc of gaseous oxidized inorganic mercury species (i.e. GOIM), either (1) desorbed from the aerosols retained by the Teflon filter located upstream of the blank filter disc and/or (2) present in the ambient air as a result of photochemically initiated environmental oxidation processes linked to the springtime polar tropospheric mercury oxidation/depletion phenomenon (Schroeder et al. 1998).

The daily time-series of TFM and GEM (Fig. 4) shows more detailed information about the relationship between TFM and

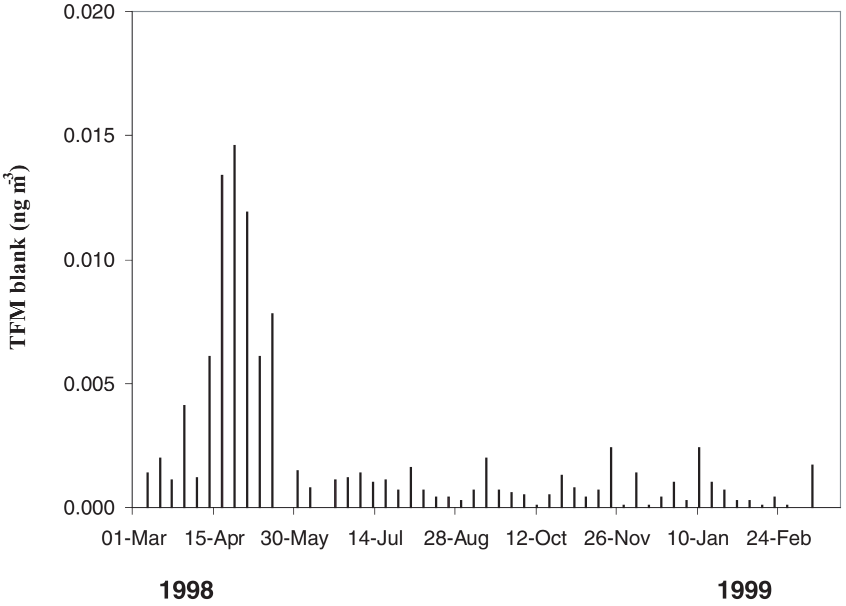

Figure 3. Observed total filterable mercury (TFM) blank values (weekly averages) at Alert, Nunavut, Canada, March 1998–March 1999.

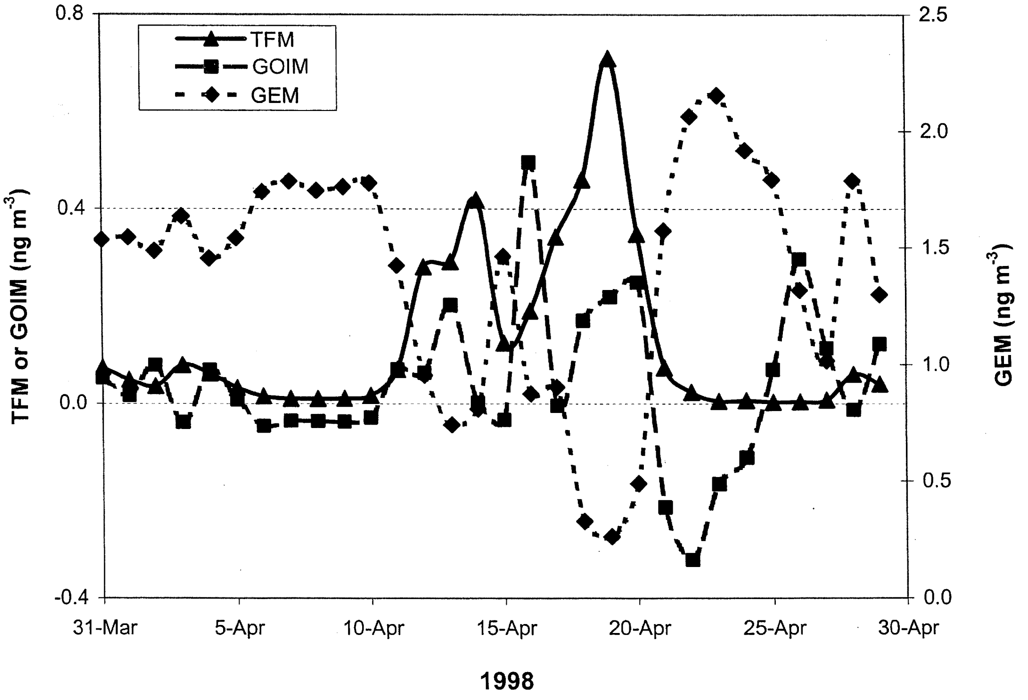

Figure 4. Time-series of total filterable mercury (TFM) concentrations, gaseous oxidized inorganic mercury (GOIM) concentrations and gaseous elemental mercury (GEM) concentrations (daily averages) at Alert, Nunavut, Canada, April 1998.

GEM in the springtime during several Hg^0 vapour depletion episodes. In one case (18 April 1998), the concentration of TFM reached a value of 0.65 ng m^{-3}, which accounts for 71% of the total airborne mercury in the surface-level air at the sampling site. The results displayed in Fig. 4 strongly suggest that GEM (Hg^0) was transformed into one or more oxidized inorganic Hg species. Since these inorganic mercury species are less volatile than the precursor Hg^0, as can be seen from Table 1, they are likely to be found in the particulate phase (i.e. associated with atmospheric aerosols/airborne particulate matter). The increased TFM concentrations, however, cannot account for the entire depleted GEM. On average, the TFM concentrations are about 37% of the typical springtime GEM value of 1.74 ng m^{-3} during those times when there are no MDEs.

Table 1. Vapour pressure of mercury compounds (Schroeder and Munthe, 1998; Sommar, 2001)

Hg compound	Vapour pressure (Pa)	
	20 °C	25 °C
Hg^0	0.180	0.27
$HgCl_2$	8.99×10^{-3}	0.017
HgO		9.20×10^{-12}
$(CH_3)_2$ Hg		8.3×10^3
CH_3HgCl		1.76
CH_3HgOH		0.9

Figure 5 shows the hourly mean results obtained from the pyrolysis unit, presented as the difference between TAM and GEM. The same pattern as that of GEM and TFM (shown in Fig. 4) is observed, confirming GEM transformation, i.e. oxidation of Hg^0 to Hg(II). For the major depletion event (which occurred on 18 April), the maximum difference between the TAM and GEM concentrations was 1.04 ng m^{-3}, which is substantially higher than that of TFM (0.65 ng m^{-3}). On average, the values of the difference between TAM and GEM (the daily values were calculated from the half-hourly measurements) are at least twice the experimental TFM values (see Fig. 6) determined at Alert in the spring of 1998. The higher values of the former parameter (namely, the difference between TAM and GEM concentrations) relative to the latter (TFM) reflect the two different measurement methodologies, i.e. *in situ* pyrolysis versus filtration. In fact, the difference between TAM and GEM should be equal to the sum of TFM and GOIM in the ambient air. The time-series of GOIM concentrations, calculated using the equation [GOIM]=[TAM]−[GEM]−[TFM], at Alert during April 1998 is also shown in Fig. 4. In essence, GOIM determined in this fashion should be comparable with the results of measurements of reactive gaseous mercury (RGM) using denuder samplers (Lindberg et al. 2002). An inverse correlation between GEM and GOIM is also observed (just as for GEM and TFM). The highest GOIM concentration (0.50 ng m^{-3}) was observed on 16 April, following the first major spring 1998 MDE (from 11 to 15 April) and on the leading edge of the second major mercury vapour depletion episode commencing on 15 April 1998. The *negative* values of GOIM are due to GEM concentrations being higher than those of TAM following the deepest depletion event (which occurred on 18 April). The temporarily higher GEM values may be a result of conversion/decomposition of Hg(II) species associated with the particulate matter that was accumulated on the Teflon filter at the inlet of the mercury vapour analyser (which is inside a temperature-controlled laboratory) during the depletion event. The (smaller) negative values prior to 10 April are perhaps due to measurement uncertainties inherent in the individual species determinations.

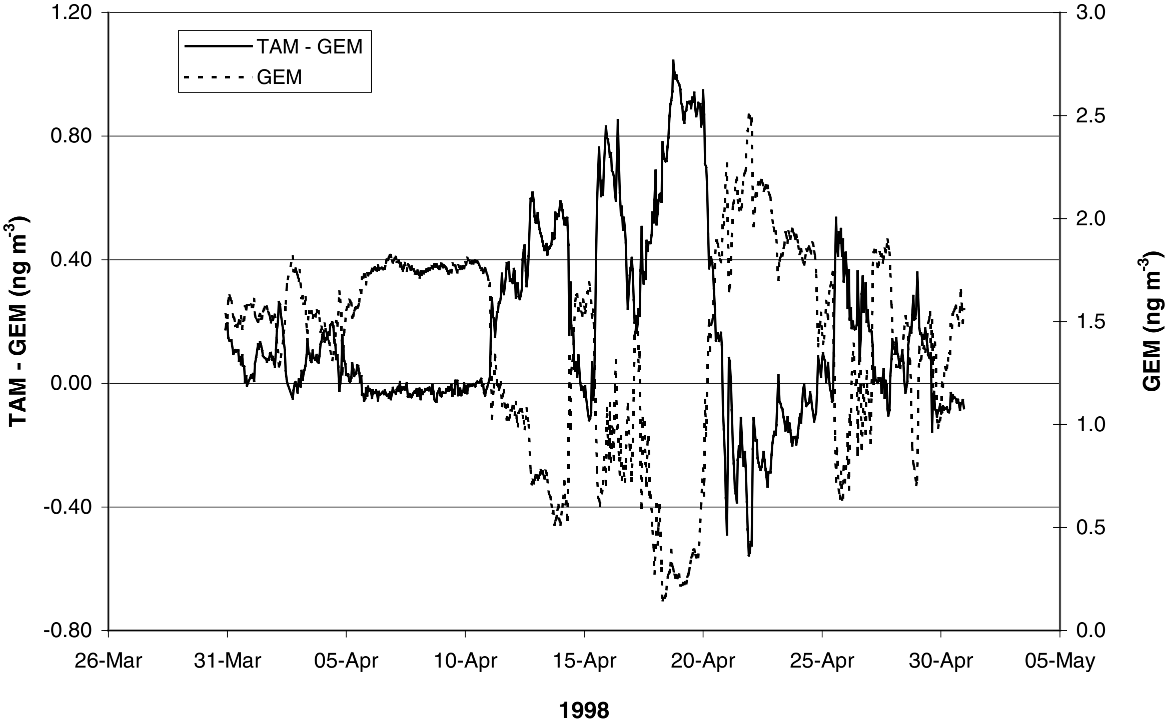

Figure 5. Time-series of total atmospheric mercury (TAM) concentrations and gaseous elemental mercury (GEM) concentrations (hourly averages) at Alert, Nunavut, Canada, April 1998.

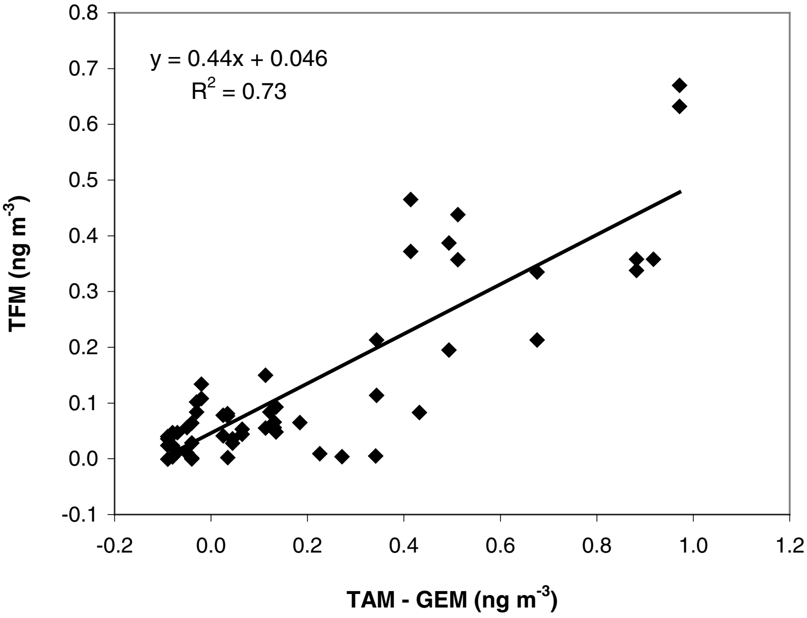

Figure 6. Correlation between total filterable mercury (TFM) concentrations and [TAM]−[GEM] at Alert, Nunavut, Canada, April 1998.

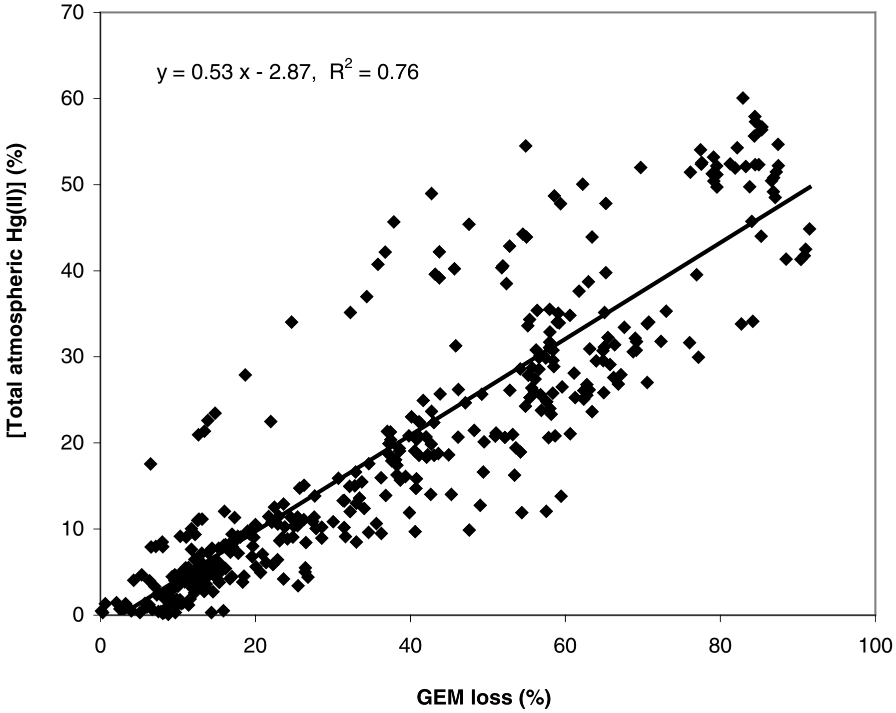

Figure 7. Normalized total atmospheric concentrations of Hg(II) as a function of GEM loss at Alert, Nunavut, Canada, April 1998. [Total atmospheric Hg(II)] (%) = ([TAM]−[GEM]) × 100/[average spring GEM]$_{\mathrm{noMDEs}}$ and GEM loss = ([average spring GEM]$_{\mathrm{noMDEs}}$−[GEM]) × 100/[average spring GEM]$_{\mathrm{noMDEs}}$.

The results presented here clearly demonstrate that chemical transformation, i.e. oxidation, of GEM leads to the previously reported Arctic and Antarctic elemental mercury vapour depletion events (MDEs) and that the oxidation/transformation reaction products of mercury exist in both the gas and the particulate phases, i.e. as both particulate-phase mercury and GOIM (or RGM). Nevertheless, the amount of mercury recovered, in terms of [TAM]−[GEM]=[TFM]+ [GOIM], cannot entirely account for all the GEM losses as shown in Fig. 7: only about 50% of the depleted ("missing") GEM (starting from an average value of about 1.75 ng m^{-3} in ambient air at Alert during times in spring 1998 when there are no MDEs) was recovered as TFM and GOIM. The missing concentration of GEM is very likely to be the result of dry (and/or wet) deposition of the reaction products—the particulate-phase mercury and/or inorganic oxidized gaseous Hg(II) species—during transport/advection of the air masses from the source/depletion region (believed to be predominantly over the Arctic Ocean, by analogy with tropospheric ozone depletion episodes, see Hopper et al. 1994) *en route* to the observation site at Alert (6 km inland, south of the Arctic Ocean). This hypothesis is supported by the fact that the concentrations of mercury in surface snow samples collected on the frozen Arctic Ocean were indeed significantly elevated following mercury depletion events (Lu et al. 2001).

4. Discussion

4.1. Mercury transformation/oxidation mechanisms/chemistry

Based on experimental evidence available so far, we propose the mechanisms/processes shown in Fig. 8 as an explanation for the Arctic (and Antarctic) springtime gaseous atmospheric mercury depletion events which occur annually during and after polar sunrise. In the Arctic (and Antarctic) during and after polar sunrise, photochemically induced chemical reactions taking place at the snow/ice surface produce (from oceanic halide salts/aerosols) various highly reactive chemical species (e.g. Br·, BrO·, ClO·, HO·, RO·) and other strong oxidants, e.g. BrCl (Barrie and Platt, 1997; Dickerson et al. 1999; Foster et al. 2001). The salt water/sea salt aerosol-derived halogen atoms and halogen oxide species, especially Br· and BrO·, play a key role in the post-polar-sunrise springtime episodic destruction of atmospheric boundary-layer (i.e. tropospheric) ozone (Barrie et al. 1988; Langendorfer et al. 1999; McElroy et al. 1999). With respect to the polar springtime (post-polar-sunrise) depletion of atmospheric mercury vapour, we think that BrO·/ClO· and/or BrCl could be of special interest and importance among the potential oxidants of Hg^0 (Schroeder et al. 1999; Lu et al. 2001). It is plausible that BrO· radicals attack Hg^0 to form $HgO_{(s)}$ and/or HgX_2 or $HgXY_{(g,s)}$ because:

(1) We have obtained evidence that a strong correlation exists between surface-level ozone and GEM concentrations in the Arctic in the spring (Schroeder et al. 1998). For example, during the period from 1 March to 31 May of 1995–1997 $R^2 = 0.76$, 0.68 and 0.77 respectively.

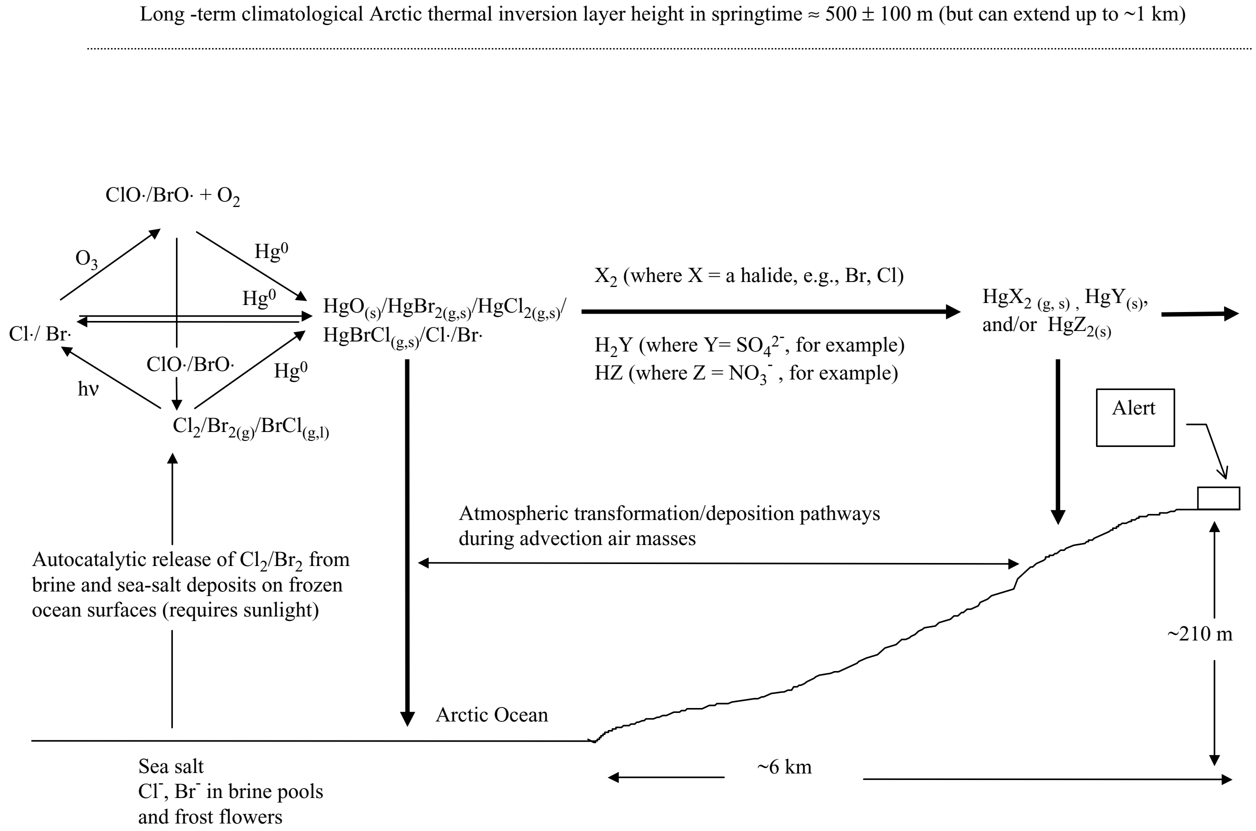

Figure 8. Schematic diagram incorporating possible mechanisms of gaseous elemental mercury (GEM) oxidation and deposition in the polar regions after polar sunrise.

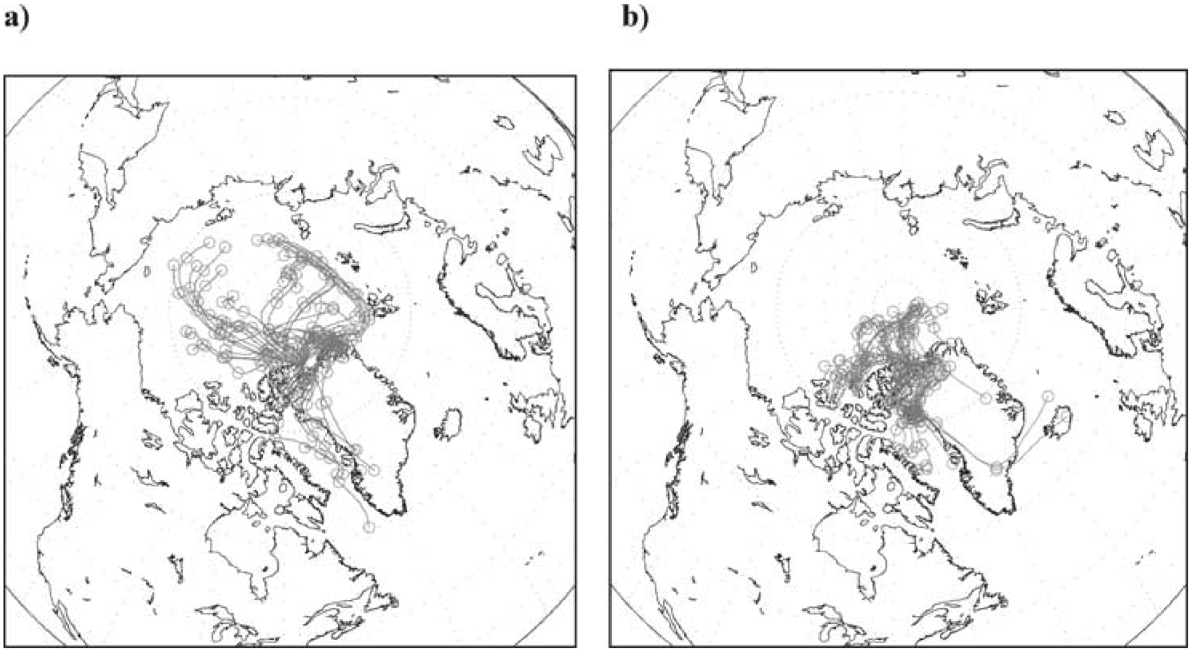

Figure 9. Back-trajectories over the previous 5 days of air parcels reaching Alert, between 31 March and 11 May 1998, at the 925 mbar level, with the gaseous elemental mercury (GEM) concentrations: (a) <1.0 ng m^{-3} and (b) >1.0 ng m^{-3}.

(2) The 5-day air parcel back-trajectories sorted by GEM concentration (see Fig. 9 (a) and (b)) show that air masses containing low (i.e. depleted) concentrations of GEM (<1.0 ng m^{-3}) were mainly transported to Alert from over the Arctic Ocean, suggesting that the GEM depletion phenomenon occurs regionally, taking place over the same area/terrain as the ozone depletion events.

(3) BrO· is a key intermediate chemical entity produced in the Arctic tropospheric ozone destruction cycle (McConnell et al. 1992; Hausmann and Platt, 1994; Richter et al. 1998; Wagner and Platt, 1998) at concentrations (several tens of parts per trillion (ppt)) significantly higher than those of GEM (0.1–0.2 ppt).

(4) The reaction of Hg^0 with BrO· (g) would (i) produce HgO (i.e. mercuric oxide) which has a very low vapour pressure (as shown in Table 1) and should exist mostly, if not entirely, in the solid state in the Arctic environment, at least during the winter and spring; and (ii) produce $HgBr_2$, which may be largely in the vapour phase, but could also be associated with airborne particulate matter (a variety of atmospheric aerosols) under typical springtime Arctic environmental conditions (air temperatures of ~ -15 to ~ -35°C), at least at Alert, in April and May.

Existence of the BrCl species (Foster et al. 2001) after polar sunrise forms another possible mechanism of Hg^0 oxidation, because:

(1) BrCl is a well-known strong oxidation reagent and has been routinely used to convert $Hg^0$to Hg(II).

(2) Its atmospheric concentrations (up to 35 ppt) at Alert, Nunavut, Canada (Foster et al. 2001) are much higher than those of GEM (0.1–0.2 ppt).

The much less volatile reaction product(s), such as a mercury halide, an oxide, or a sulfate or nitrate compound of mercury produced by oxidation of Hg^0 vapour can be expected, on the basis of their vapour pressures (see Table 1), to preferentially exist or become associated with atmospheric aerosol, rather than remaining in the gas phase under typical (still very cold) springtime environmental conditions in the Arctic, thus explaining the dramatically elevated concentrations of TFM observed frequently at Alert during the 3-month period following polar sunrise (Lu et al. 2001). This hypothesis is supported by the increase in the particle concentrations during the mercury depletion events (Gauchard et al. 2003). It is also possible, but less likely, that the halogen atoms (Cl·, Br·, and/or I·) attack Hg^0 vapour, forming HgX_2 (since this would require either a concerted "three-body" collision or a two-step sequential reaction).

4.2. *Distribution of oxidized inorganic mercury species between gas and particulate phases*

The results obtained in this study show (Fig. 4) that, in general, the concentrations of TFM are higher than those of RGM at Alert. This relative distribution has in the mean time been confirmed at the same site using both a Tekran-1130 and -1135 automatic mercury speciation unit and manual measurements (Steffen et al. 2003), even though the particulate-phase mercury measured by these methods represents only a fraction (aerosol particles <2.5 μm in diameter) of the total filterable mercury (Foltesco and Zahn, 1995), as all the particles larger than 2.5 μm were excluded by an impactor located at the air inlet upstream of the KCl-coated denuder. In addition, RGM in these measurements was removed using a KCl-coated denuder prior to particulate mercury collection to eliminate possible condensation of RGM species on the particles retained by the filter. A similar distribution between particulate-phase mercury and RGM has been observed by Poissant and Pilote (2003) at Kuujjuarapik, Quebec, Canada, also using a Tekran-1130 and -1135 mercury speciation unit. The results obtained using the same type of mercury speciation unit (Tekran-1120 and -1135) at Point Barrow, Alaska, however, show that the RGM is the predominant species (i.e. PM/RGM <1) in the atmosphere during the mercury depletion events (Lindberg et al. 2002). The ratio of RGM/PM may depend upon the "chemical age" of the particular air mass being sampled, and perhaps also on different meteorological/climatological conditions prevailing at the various polar sites. When using a manual method (Lu et al. 1998) for total filterable mercury and a KCl-coated denuder method for RGM measurements, Wängberg et al. (2003) at Ny-Ålesund and Temme et al. (2003) in the Antarctic observed that the concentrations of RGM were higher than those of TFM. The above studies demonstrate that even if the RGM concentration was more than 30 times higher than that of particulate mercury in the air stream, absorption/condensation of RGM on the particles retained by the filter during sampling is not significant when using the manual sampling method (Lu et al. 1998).

5. Conclusion

Oxidation/conversion of GEM (Hg^0) in the Arctic occurs in the spring after polar sunrise, leading to formation of Hg(II) species that exist in both gas and particulate phases. Halogen species (e.g. ClO·/BrO·, Br· and BrCl) produced through photochemically induced heterogeneous reactions are believed to be responsible for oxidizing Hg^0. Elevated concentrations of mercury associated with particulate matter are real and are not a result of sampling artefact/bias. Research is needed to develop standard sources to calibrate the methods currently used to differentiate mercury species and to identify the mercury species that are named as RGM. Definitive, targeted laboratory research under conditions that mimic polar springtime, and simultaneous measurements with high time resolution of all the mercury species in the Arctic atmosphere/environment are needed to either confirm or rule out the chemical mechanisms proposed in this paper.

6. Acknowledgments

We are grateful to the Meteorological Service of Canada (MSC) and the Department of National Defence (DND) personnel at Canadian Forces Station (CFS) Alert for the operational and logistical supports and to A. Steffen (MSC) for assistance with this project.

References

AMAP (Arctic Monitoring and Assessment Programme) 1998. *AMAP Assessment Report: Arctic Pollution Issues*, Ch. 7, Oslo, Norway.

Asmund, G. and Nielsen, S. P. 2000. Mercury in dated Greenland marine sediments. *Sci. Total Environ.* **245**, 61–72.

Barrie, L. and Platt, U. 1997. Arctic troposphere chemistry: an overview. *Tellus* **49B**, 450–454.

Barrie, L. A., Bottenheim, J. W., Schnell, R. C., Crutzen, P. J. and Rasmussen, R. A. 1988. Ozone destruction and photochemical reactions ar polar sunrise in the lower Arctic troposphere. *Nature* **334**, 138–141.

Berg, T., Bartnicki, J., Munthe, J., Lattila, H., Hrehoruk, J., et al. 2001. Atmospheric mercury species in the European Arctic: Measurements and modeling. *Atmos. Environ.* **35**, 2569–2582.

Berg, T., Sommar, J., Wangberg, I., Gardfeldt, K., Munthe, J., et al. 2003. Arctic mercury depletion events at two elevations as observed at the Zeppelin Station and Dirigible Italia, Ny-Alesund, spring 2002. *J. Physique IV* **107**, 151–154.

Bjerregaard, P. and Hansen, J. C. 2000. Organochlorines and heavy metals in pregnant women from the Disko Bay area in Greenland. *Sci. Total Environ.* **245**, 195–202.

Branfireun, B. A., Roulet, N. T., Kelly, C. A. and Rudd, J. W. M. 1999. *In situ* sulphate stimulation of mercury methylation in a boreal peatland: toward a link between acid rain and methylmercury contamination in remote environment. *Glob. Biogeochem. Cy.* **13**, 743–750.

Bowles, K. C., Apte, S. C., Maher, W. A., Kawei, M. and Smith, R. 2001. Bioaccumulation and biomagnification of mercury in Lake Murray, Papua New Guinea. *Can. J. Fish. Aquatic Sci.* **58**, 888–897.

Cheng, M. D. and Schroeder, W. H. 2000. Potential atmospheric transport pathways for mercury measured in the Canadian high arctic. *J. Atmos. Chem.* **35**, 101–107.

Dickerson, R. R., Rhoads, K. P., Carsey, T. P., Oltmans, S. J., Burrows, J. P., et al. 1999. Ozone in the remote marine boundary layer: a possible role for halogens. *J. Geophys. Res.* **104**, 21 385–21 395.

Ebinghaus, R., Kock, H. H., Temme, C., Einax, J. W., Lowe, A. G., et al. 2002. Antarctic springtime depletion of atmospheric mercury. *Environ. Sci. Technol.* **36**, 1238–1244.

Foltesco, V. L. and Zahn, A. 1995. Aerosols used as tracers for stratosphere-troposphere exchange in the Arctic. *Atmos. Environ.* **29**, 1777–1784.

Foster, K. L., Plastridge, R. A., Bottenheim, J. W., Shepson, P. B., Finlayson-Pitts, B. J., et al. 2001. The role of Br_2 and BrCl in surface ozone destruction at polar sunrise. *Science* **291**, 471–474.

Gauchard, P. A., Guehenneux, G., Ferrari, C. P., Poissant, L., Dommergue, A., et al. 2003. Evidences of modifications in the size distribution of particles during a mercury depletion event at Kuujjuarapik/Whapmagoostui, Quebec (Canada). *J. Physique IV* **107**, 521–524.

Hausmann, M. and Platt, U. 1994. Spectroscopic measurement of bromine oxide and ozone in the high Arctic during Polar Sunrise Experiment 1992. *J. Geophys. Res.* **99**, 25 399–25 413.

Hermanson, M. H. 1998. Anthropogenic mercury deposition to Arctic lake sediments. *Water, Air, Soil Pollut.* **101**, 309–321.

Hopper, J. F., Peters, B., Yokouchi, Y., Niki, H., Yobson, B. T., et al. 1994. Chemical and meteorological observations at ice camp SWAN during Polar Sunrise Experiment 1992. *J. Geophys. Res.* **99**(D12), 25 489–25 498.

Ikingura, J. R. and Akagi, H. 1999. Methylmercury production and distribution in aquatic systems. *Sci. Total Environ.* **234**, 109–118.

Jensen, J., Adare, K. and Shearer, R.(eds.) 1997. In: *Canadian Arctic Contaminants Assessment Report*, Ch. 2, Department of Indian Affairs and Northern Development, Ottawa, Canada.

Langendorfer, U., Lehrer, E., Wagenbach, D. and Platt, U. 1999. Observation of filterable bromine variabilities during Arctic tropospheric ozone depletion events in high (1 hour) time resolution. *J. Atmos. Chem.* **34**, 39–54.

Leong, C. C. W., Syed, N. I. and Lorscheider, F. L. 2001. Retrograde degeneration of neurite membrane structural integrity of nerve growth cones following *in vitro* exposure to mercury. *NeuroReport* **12**, 733–737

Lin, C. J., Cheng, M. D. and Schroeder, W. H. 2001. Transport patterns and potential sources of total gaseous mercury measured in Canadian high Arctic in 1995. *Atmos. Environ.* **35**, 1141–1154.

Lindberg, S. E., Brooks, S., Lin, C. J., Scott, K., Meyers, T., et al. 2001. Formation of reactive gaseous mercury in the Arctic: evidence of oxidation of Hg° to gas-phase Hg-II compounds after Arctic sunrise. *Water, Air, Soil Pollut.: Focus* **1**, 295–302.

Lindberg, S. E., Brooks, S., Lin, C. J., Scott, K. J., Landis, M. S., et al. 2002. Dynamic oxidation of gaseous mercury in the Arctic troposphere at polar sunrise. *Environ. Sci. Technol.* **36**, 1245–1256.

Lockhart, W. L., Wilkinson, P., Billeck, B. N., Danell, R. A., Hunt, R. V., et al. 1998. Fluxes of Hg to lake sediments in central & northern Canada inferred from dated sediment cores. *Biogeochemistry* **40**, 163–173.

Lu, J. Y., Schroeder, W. H., Berg, T., Munthe, J., Schneeberger, D. et al. 1998. A novel device for sampling and determination of total particulate mercury in ambient air. *Anal. Chem.* **70**, 2403–2408.

Lu, J. Y., Schroeder, W. H., Barrie, L. A., Steffen, A., Welch, E. H., et al. 2001. Magnification of atmospheric mercury deposition to polar regions in springtime: the link to tropospheric ozone depletion chemistry. *Geophys. Res. Lett.* **28**, 3219–3222.

McConnell, J. C., Henderson, G. S., Barrie, L., Bottenheim, J., Niki, H., et al. 1992. Photochemical bromine production implicated in Arctic boundary-layer ozone depletion. *Nature* **355**, 150–152.

McElroy, C. T., McLinden, C. A. and McConnell, J. C. 1999. Evidence for bromine monoxide in the free troposphere during the Arctic polar sunrise. *Nature* **397**, 338–341.

Poissant, L. and Pilote, M. 2003. Time series analysis of atmospheric mercury in Kuujjuarapik/Whapmagoostui (Quebec). *J. Physique IV* **107**, 1079–1082.

Richter, A., Wittrock, F., Eisinger, M. and Burrows, J. P. 1998. GOME observations of tropospheric BrO in northern hemispheric spring and summer 1997. *Geophys. Res. Lett.* **25**, 2683–2686.

Schroeder, W. H. and Munthe, J. 1998. Atmospheric mercury–an overview. Atmos. *Environ.* **32**, 809–822.

Schroeder, W. H. and Jackson, R. A. 1985. An instrumental analytical technique for speciation of atmospheric mercury. *Int. J. Environ. Anal. Chem.* **22**, 1–18.

Schroeder, W. H., Anlauf, K. G., Barrie, L. A., Lu, J. Y., Steffen, A., et al. 1998. Arctic springtime depletion of mercury. *Nature* **394**, 331–332.

Schroeder, W. H., Anlauf, K. G., Barrie, L. A., Steffen, A. and Lu, J. 1999. Depletion of mercury vapor in the Arctic troposphere after polar sunrise. In: *Proceedings of EUROTRAC Symposium '98* (eds M. P. Borrell and P. Borrell). WIT Press, Southampton, p. 358.

Sommar, J. S. 2001. The atmospheric chemistry of mercury—kinetics, mechanisms and speciation. *Thesis*, Göteborg University, Department of Chemistry, p. 53.

Steffen, A. and Schroeder, W. H. 1999. *Standard Operating Procedures Manual for Total Gaseous Mercury Measurements*. Canadian Atmospheric Mercury Measurement Network, Meteorological Service of Canada, Environment Canada, Toronto.

Steffen, A., Schroeder, W. H., Edwards, G. and Banic, C. 2003. Mercury throughout polar sunrise 2002. *J. Physique IV* **107**, 1267–1270.

Temme, C., Einax, J. W., Ebinghaus, R. and Schroeder, W. H. 2003. Measurements of atmospheric mercury species at a coastal site in the Antarctic and over the South Atlantic Ocean during polar summer. *Environ. Sci. Technol.* **37**, 22–31.

Wagner, T. and Platt, U. 1998. Satellite mapping of enhanced BrO concentrations in the troposphere. *Nature* **395**, 486–490.

Wang, J., Xiao, Z. and Lindqvist, O. 1995. On-line measurement of mercury in simulated flue gas. *Water, Air, Soil Pollut.* **80**, 1217–1226.

Wängberg, I., Sommar, J., Berg, T., Gardfelt, K. and Munthe, J. 2003. Interpretation of mercury depletion events observed at Ny-Ålesund, Svalbard during spring 2002. *J. Physique IV* **107**, 1353–1356.