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Natural and anthropogenic atmospheric mercury in the European Arctic: a speciation study

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Abstract

It is agreed that gaseous elemental mercury (GEM) is converted to reactive gaseous mercury (RGM) during springtime Atmospheric Mercury Depletion Event (AMDE). RGM is associated with aerosols (PHg) provided that there are sufficient aerosols available for the conversion from RGM to PHg to occur. This study reports the longest time series of GEM, RGM and PHg concentrations from a European Arctic site. From 27 April 2007 until 31 December 2008 composite GEM, RGM and PHg measurements were conducted in Ny-Ålesund (78°54' N, 11°53' E). The average concentrations of the complete dataset were $1.62 \pm 0.3 \text{ ng m}^{-3}$, $8 \pm 13 \text{ pgm}^{-3}$ and $8 \pm 25 \text{ pgm}^{-3}$ for GEM, RGM and PHg, respectively. The study revealed a clear seasonal distribution of GEM, RGM and PHg previously undiscovered. For the complete dataset the atmospheric mercury distribution was 99% GEM, whereas RGM and PHg constituted <1%. Increased PHg concentration occurred exclusively from March through April, and constituted on average 75% of the reactive mercury species in the respective period. RGM was suggested as the precursor for the PHg existence, but long range transportation of PHg has to be taken into consideration. Surprisingly, RGM was not solely formed during the spring AMDE season. Environment Canada's Global/Regional Atmospheric Heavy Metal model (GRAHM) suggested that in situ oxidation of GEM by ozone may be producing the increased RGM concentrations from March through August. Most likely, in situ oxidation of GEM by BrO produced the observed RGM from March through August. The AMDEs occurred from late March until mid June and were thought to be of non-local origin, with GEM being transported to the study site by a wide variety of air masses. With some exceptions, no clear meteorological regime was associated with the GEM, RGM and PHg concentrations.

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1 Introduction

Since the discovery of the Atmospheric Mercury Depletion Event (AMDE) in 1995, significant efforts have been carried out to understand this circumpolar phenomenon (Ariya et al., 2004; Schroeder and Munthe, 1998; Skov et al., 2004). It is agreed that gaseous elemental mercury (GEM) is converted to reactive gaseous mercury (RGM) during an AMDE. RGM denotes water soluble, divalent mercury and constitute a minor part of the total atmospheric mercury (Lindberg and Stratton, 1998). RGM is subsequently either deposited or associated with aerosols (PHg) (Steffen et al., 2008). However, this argument holds provided that there are sufficient aerosols available for the conversion from RGM to PHg to occur.

The formation of RGM is likely caused by the oxidation of GEM by reactive halogens. BrO (Hedgecock and Pirrone, 2004), ClO (Donohoue et al., 2005) and IO (Calvert and Lindberg, 2004) are suggested as possible oxidants, where BrO is thought to be the predominant oxidant for GEM (Steffen, et al., 2008). The reactive halogens are assumed to be generated from refreezing sea ice forming on open waters and UV radiation (Steffen, et al., 2008).

Mercury is emitted to the atmosphere by both natural and anthropogenic processes. The lifetime of GEM on the order of 1 year allow for advection to the Arctic. RGM and PHg have atmospheric residence time on the order of days which restrict advection by winds (Schroeder and Munthe, 1998).

Studies reporting composite GEM, RGM and PHg measurements from Polar sites are conducted mainly in spring and restricted to the studies at Barrow (71°19' N, 156°37' E), March–June 2001 (Lindberg, 2002), Alert (82°58' N, 62°38' E), spring 2002 (Steffen et al., 2003b), Ny-Ålesund (78°54' N, 11°53' E), April–May 2000 (Berg et al., 2003b), Ny-Ålesund, May 2002 (Berg et al., 2003a), Ny-Ålesund, April–May 2003 (Aspmo et al., 2005; Gauchard et al., 2005; Sprovieri et al., 2005) and Alert, January–July 2005 (Cobbett et al., 2007). Aspmo et al. (2006) and Sommar et al. (2010) reported GEM, RGM and PHg concentrations over the North Atlantic Ocean, however

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the RGM and PHg concentrations remained low during summer. It is also inviting to mention that corresponding studies have been conducted at Station Nord, North-east Greenland (81°36' N, 16°40' E) (Skov, et al., 2004) and at Neumayer, Antarctica (70°39' S, 8°15' W) (Ebinghaus et al., 2002b).

5 The relative distribution of RGM and PHg is thought to indicate the age of an air parcel, and consequently the origin of an AMDE (Aspmo, et al., 2005; Gauchard, et al., 2005; Lindberg, 2002; Sprovieri, et al., 2005; Steffen et al., 2003a). At Alert the predominant specie is PHg, whereas RGM dominates at Barrow (Cobbett, et al., 2007; Kirk, 2006; Lindberg, 2002; Steffen et al., 2005; Steffen et al., 2003c). Gauchard et
10 al. (2005) and Sprovieri et al. (2005) indicated no predominance of either RGM or PHg in Ny-Ålesund. Berg et al. (2003b) and Sommar et al. (2007) reported equal or larger PHg concentrations compared to RGM in Ny-Ålesund, suggesting AMDEs of non-local origin and deposition of mercury to snow and ice surfaces outside the measurement site.

15 The present study, which discusses the longest time series of atmospheric GEM, RGM and PHg available from Ny-Ålesund, reveals a clear seasonal distribution of GEM, RGM and PHg previously undiscovered at Polar sites. Additionally we investigate the behaviour of the mercury species during AMDEs and discuss the main variables associated with the GEM, RGM and PHg concentrations. The results presented in the
20 study are supplemented by natural and anthropogenic emissions of GEM, RGM and PHg seen by the Environment Canada's Global/Regional Atmospheric Heavy Metal model (GRAHM).

2 Experimental

2.1 Study site

25 Ny-Ålesund is a research settlement located next to Kongsfjorden on the west coast of Spitsbergen (Fig. 1b). The settlement is surrounded by steep mountains ranging in

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altitude from 500 to 1000 m to the south and east of Kongsfjorden and open sea to the north-west.

Composite RGM and PHg determinations were performed at the Zeppelin air monitoring station (henceforth named Zeppelin) from 27 April 2007. Zeppelin is located 2 km from the settlement at an elevation of 474 m above sea level. GEM has been sampled at a five minutes time resolution at Zeppelin by the Norwegian Institute for Air Research (NILU) since 2000 (Berg et al., 2003b).

2.2 Composite GEM, RGM and PHg measurements

A Tekran 1130 denuder module and a Tekran 1135 particulate module were attached to the front end of a Tekran 2537A analyzer (cold vapor atomic fluorescence spectrometry, CV-AFS) and provided continuous concentrations of RGM and PHg. The Tekran 1130, 1135 and 2537A are henceforth named the speciation system. The speciation system was programmed to collect one-hour composite RGM and PHg samples according to the method 35-2L5 (for details see Tekran's user manual). A detailed description of this speciation system is given in Landis et al. (2002) and Cobbett et al. (2007). A second Tekran 2537A was used to determine GEM at a five-minute resolution. The system is described elsewhere (Aspmo et al., 2005; Ebinghaus et al., 1999; Landis et al., 2002). Further details concerning the instrument at Zeppelin is given in Berg et al. (2003b).

GEM, RGM and PHg concentrations were reported in ng m^{-3} and pgm^{-3} , respectively. The final concentrations of RGM and PHg were calculated as the sum of the heating cycles.

An AMDE is operationally defined in this paper as periods where the concentration of $\text{GEM} < 1 \text{ ng m}^{-3}$. Observation times are in Coordinated Universal Time (UTC).

2.3 Ancillary data

Back trajectory data derived by the Norwegian Institute for Air Research (NILU) used meteorological data which originated from the European Centre for Medium Range

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Weather Forecasts (ECMWF). The trajectories describe the potential source areas within a 7- day transport time to Ny-Ålesund. Since Zeppelin is located at 474 m, air parcels arriving at 500 m height were used.

Monthly average BrO vertical densities were retrieved from the Sciamachy instrument (University of Bremen) (Richter et al., 1998), and facilitated tracking of the development and transport of BrO plumes in the Arctic boundary layer.

Global radiation, wind direction, air temperature and relative humidity reported from Ny-Ålesund were provided by the Alfred Wegener Institute (AWI). The meteorological data represent hourly averages. Meteorological values reported at Zeppelin itself were not used in order to promote consistency with Steen et al. (2009) and Durnford et al. (2010). Hourly average ozone concentrations from Zeppelin were provided by NILU. These ozone concentrations were recorded by UV absorption spectrometry (API 400A).

2.4 Quality control

Both Tekran 2537A instruments were auto-calibrated every 25 h using the instruments internal permeation source. Additionally, the accuracy of the GEM measurements was assured by manual injection of a known amount of GEM six times during the current study (Tekran 2505 instrument, mercury vapor primary calibration unit). The permeation rate was changed when the measured GEM was larger than $\pm 5\%$ of the expected concentration.

The measurement error of the Tekran 2537A instrument is estimated to be $\pm 2\%$ resulting in an average instrumental detection limit of 0.06 ng m^{-3} (Poissant et al., 2005). The sample inlet for GEM measurements was located three meters above the surface snow, two meters to the east of the station building. Further details are given in Berg et al. (2003b). Currently, no standardized calibration method exists to determine RGM and PHg concentrations. Since elevated concentrations of RGM and PHg were observed during non-AMDEs (Fig. 2), MDL was calculated as three times the standard deviation of the two last zero air measurements in a speciation cycle (for details see the

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Tekran's user manual). An MDL of 7 pg m^{-3} was calculated for the RGM and PHg measurements. The sample inlet for RGM and PHg measurements was located to the east, one meter from the building wall and two meters from the sample inlet for the determination of GEM. The denuder and regenerable particulate filter (RPF) were replaced once a month. The denuders were blanked in situ. The soda lime was replaced every week. The instrumental setup was consistent with the Tekran's user manual. RGM and PHg concentrations were automatically corrected for flow rate, scaling by a user entered constant (i.e. 8.33) and blank (i.e. mean of the two last zero air concentrations prior to the desorption).

Only Teflon lines and fittings were used. GEM, RGM and PHg concentrations less than the MDL were set to MDL/2 unless specified otherwise.

3 Results and discussions

3.1 Overall atmospheric mercury speciation

GEM, RGM and PHg concentrations from 27 April 2007 through 31 December 2008 are presented in Fig. 2. The average GEM concentration of the complete dataset \pm one standard deviation (SD) was 1.62 ± 0.3 . The concentrations corresponded to the global background concentration of GEM in the Northern Hemisphere ($1.5\text{--}1.7 \text{ ng m}^{-3}$) (Ebinghaus et al., 2002a; Slemr et al., 2003; Steffen, et al., 2008; Temme et al., 2004) as well as to the mean surface-level GEM concentration ($1.56 \pm 0.2 \text{ ng m}^{-3}$) simulated at Ny-Ålesund for the observed dataset's time period by Environment Canada's Global/Regional Atmospheric Heavy Metals model (GRAHM; for details concerning the GRAHM see Durnford et al., 2010). The average concentrations (complete dataset) \pm one SD were $8 \pm 13 \text{ pgm}^{-3}$ and $8 \pm 25 \text{ pgm}^{-3}$ for RGM and PHg, respectively. The observed mean concentration of PHg was in good agreement with the surface-level mean concentration of $7 \pm 12 \text{ pgm}^{-3}$ simulated by GRAHM for the observed dataset's time period at Ny-Ålesund. However, the simulated mean concentration of RGM at

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23±46 pgm⁻³ was higher than observed. It is possible that the observed RGM mean concentration is somewhat too low. Aspmo et al. (2005) reported that concentrations of oxidized mercury cannot yet be quantified accurately, while Lyman et al. (2010) indicated that KCl coated denuders for collection of RGM may not be as robust as previously thought. It is also difficult to compare simulated concentrations with point-source observations since a simulated value represents the average concentration for an entire 1°×1° grid cell while an observation represents a single location. The observed concentrations were consistent with the RGM and PHg concentrations reported by Aspmo et al. (2005) from Ny-Ålesund using a similar speciation system (maximum 160±57 pgm⁻³ and 63±34 pgm⁻³ for RGM and PHg, respectively, during AMDEs). The larger standard deviations calculated for RGM and PHg, compared to GEM, reflects faster reactivity and lower atmospheric residence time (Junge, 1972; Poissant, et al., 2005).

Figures 2 and 3 clearly indicate a seasonal distribution of the GEM, RGM and PHg concentrations. Overall, the concentration of GEM is fairly constant throughout the year apart from the abrupt concentration drops attributed to AMDEs. As proposed by Steen et al. (2009) GEM emission from surface snow and efflux from surface waters were presumed to encourage the springtime “hump” seen in Fig. 2 and expressed by the whiskers in March through May (Fig. 3). A similar process was also recorded following spring melt at Alert (Schroeder et al., 1998) and Barrow (Lindberg et al., 2002). Despite the wrong unit (ngm⁻² s⁻¹ instead of ng m⁻² h⁻¹) (Steen, et al., 2009), Cobbett et al. (2007) observed a GEM flux above bare soil. The air – soil efflux of GEM in Ny-Ålesund will be investigated (Steen, In preparation). Efflux from surface waters and arrival of air parcels with increased GEM concentration explain the fluctuating GEM concentrations from June through August.

During polar night the GEM concentration remains at background concentration, likely as a result of stable atmospheric conditions and consequently less vertical mixing.

During polar night (October through January) the RGM concentration was <MDL.

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In March the RGM concentration was $11 \pm 15 \text{ pgm}^{-3}$. Note that data from February is missing. The RGM concentration from April through August corresponded to the concentration during the insolation transition period ($13 \pm 16 \text{ pgm}^{-3}$). Lindberg (2002) observed that RGM formation ceased from the onset of snow melt at Barrow, whereas Cobbett et al. (2007) observed RGM until June at Alert. The present study is, however, the only study to date that reports increased concentrations of RGM from early March throughout August ($11 \pm 16 \text{ pgm}^{-3}$). This finding suggests that RGM is not solely formed during AMDEs. The maximum concentration of RGM (145 pgm^{-3}) was observed during 24-hours daylight at 27 July 2007, suggesting that radiation is of importance for the existence of RGM. As discussed in more detail below RGM correlated with global radiation emphasising the importance of radiation upon RGM existence.

The increased PHg concentrations occurred almost exclusively in March and April ($42 \pm 65 \text{ pgm}^{-3}$). PHg concentrations $< \text{MDL}$ during summer, in the presence of increased RGM concentrations, could be attributed to short-lived RGM species which are easily reduced to GEM. The decreased PHg concentrations could also simply be explained by the fact that there are fewer aerosols present in the atmosphere at this time of the year (Ström et al., 2003). The lack of oxidation of GEM and local formation of RGM and PHg due complete darkness, and the lack of advection from sunlit source regions to the south, thus seem to be the most credible explanations for the sudden decrease in RGM and PHg concentrations in the fall/winter.

For the complete dataset the atmospheric distribution was 99% and $< 1\%$ for GEM and the reactive mercury species (RGM and PHg), respectively. The monthly (March through August) atmospheric distribution of the reactive mercury species is presented in Fig. 4. Calculations included observations from a given month from both years in the complete dataset. Concentrations $< \text{MDL}$ were set to $\text{MDL}/2$. PHg dominates from March through April (on average 75% of the reactive mercury species). A clear shift to a predominance of RGM occurred from May through August. For the first time increased RGM concentrations were observed during non-AMDEs which states that RGM is not solely a springtime phenomenon. From May through August the atmospheric distri-

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5 bution of the reactive mercury species was on average 72% and 28% for RGM and PHg, respectively. This states that a seasonal variation in the atmospheric distribution of RGM and PHg is evident. High RGM concentrations (Table 1) observed from March through May suggests that RGM is the main precursor for PHg existence, but long range transportation of PHg have to be taken into consideration.

3.2 Atmospheric mercury speciation during the spring AMDE season

AMDEs were observed from late March until mid June. The strongest AMDEs (GEM<MDL) were observed from late March until late April (Figs. 2 and 3). The period from March through April showed the highest monthly BrO vertical densities above Svalbard, indicating that reactive bromine species are contributors in the oxidation of GEM during polar spring (Lu et al., 1998; Sommar, et al., 2007).

10 The concentrations of RGM and PHg accounted for on average about 10% of the depleted GEM during the AMDEs. The remainder of the converted GEM was likely deposited to nearby snow and ice surfaces (Aspmo, et al., 2005; Lindberg, 2002; Steffen et al., 2002). The predominance of PHg with respect to RGM and no clear meteorological regime associated with the AMDEs (discussed in more detail later) would all suggest that the AMDEs are of non-local origin. There are several possible explanations for the advection of air masses already depleted in GEM. Ny-Ålesund is located at the ridge of the Gulf Stream delivering warm water masses to the Barents Sea, which may affect the formation mechanism for RGM and PHg. Reduction in ice cover, due to the warmer water masses, would also affect the location and strength of mercury deposition due to AMDEs. It seems likely that there is an optimum combination of open water, first-year ice and snow for AMDEs to occur. Too much open water would restrict the occurrence of AMDEs (Macdonald et al., 2008).

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3.3 Local formation and anthropogenic source areas of RGM and PHg

The AMDEs of non-local origin and the short atmospheric residence time of RGM are all indications suggesting that the observed RGM and PHg have been formed locally. Assuming that local oxidation of GEM by BrO occurs in the spring through the summer followed by increased concentration of RGM (personal communication, K. Gårdfeldt, 2010), the oxidation (i.e. depletion of GEM) could be masked by the arrival of air masses already depleted in GEM and the strong reemission from surface snow during the spring AMDE season. In summer, it is possible that the in situ oxidation of GEM by ozone is responsible for the observed elevated concentrations of RGM; the GRAHM, which uses ozone oxidation, is able to reproduce the elevated concentrations. The attendant decreases in GEM concentration are likely masked by either a strong reemission of GEM from surface waters and/or the arrival of air masses with increased GEM concentrations. The attendant depletion of ozone is likely masked by the summertime arrival from the south of air masses with enhanced ozone concentrations; only in spring do ozone concentrations decline significantly. However, there is a significant debate as to whether the O_3 -GEM reaction rate in the atmosphere is fast enough for the oxidation of GEM by ozone to be prevalent (Peterson et al., 2009). Thus, the continuation throughout summer of the springtime in situ oxidation of GEM by BrO may be responsible for the observed summertime elevated concentrations of RGM.

Interestingly, increased concentrations of RGM and PHg were neither found by Sommar et al. (2010) nor by Aspö et al. (2006) in the summer over the North Atlantic Ocean.

Although a local RGM formation seems likely, the presence of possible anthropogenic source areas to the East are supported by the RGM and PHg emissions presented in Fig. 5. This figure presents mercury emitted during 2008 as seen GRAHM (Dastoor et al., 2008). Anthropogenic emission fields were produced by Pacyna et al. (2006) and are valid in 2005. Figure 5 demonstrates that northern Europe and Asia

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are by far the most important Northern Hemispheric high-latitude sources of RGM and PHg. GEM emitted in the regions of Europe and Asia shown in Fig. 5 is also produced primarily by anthropogenic emissions. GEM emissions over polar waters represent the rapid reemission of mercury deposited onto the cryosphere. This process is particularly active during the spring AMDE season.

3.4 Correlation analysis

A correlation analysis was performed to assess the main meteorological variables (air temperature, wind direction, relative humidity, ozone and global radiation) associated with low and high GEM, RGM and PHg concentrations (Fig. 6), (Table 1). All calculations used the 25th and 75th percentiles of the complete dataset as thresholds for low and high concentrations, with the requirement that the monthly average correlation coefficients (R) are calculated only when at least ten pairs of valid data points are available for the calculation. Only concentrations \geq MDL were used unless stated otherwise. In addition, a correlation analysis was performed for AMDEs. In order to include the GEM concentrations $<$ MDL observed during the strongest AMDEs, all concentrations were included in the correlation calculations, with concentrations $<$ MDL set to MDL/2. All variables were interpolated to the species' times. Calculations included observations from a given month from both years in the complete dataset.

Meteorological variables with $R > 0.5$ and $R < -0.5$ form the basis for this discussion. Variables not strongly correlated reflect the absence of a specific meteorological regime controlling the concentrations of the given species (shaded area in Fig. 6).

Air temperature and the low GEM concentrations are correlated in January. The correlation for ozone and the low GEM concentrations in spring reflects the occurrence of AMDEs and corresponds to the study by Eneroth et al. (2007).

Air temperature, relative humidity, ozone and global radiation tend to be correlated with high GEM concentrations during polar night. The correlation of the high GEM concentrations with ozone reflects GEM and ozone at background concentrations. High RGM concentrations anti correlated with ozone in April. The importance of ozone cor-

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responds to the oxidation of GEM and formation of RGM during the spring AMDE season. It is also inviting to mention that high RGM concentrations are most pronounced in April. The high RGM concentrations correlate with global radiation in May, which suggests a diurnal cycle of photochemistry (Peterson, et al., 2009): interestingly, a mid-day peak in RGM concentrations was not observed. The importance of global radiation is, however, consistent with the importance of radiation for the oxidation of GEM to occur (Steffen et al., 2008). Although no such clear correlation was found in this study, Peterson et al. (2009) proposed that more incident light, warmer air temperatures and lower relative humidity would all promote formation of atmospheric oxidants available for oxidation of GEM and formation of RGM in summer.

Ozone and air temperature anti correlate with the low PHg concentrations in October, suggesting that the highest of the low PHg concentrations are found in cold air masses low in ozone, whereas the lowest of the low PHg concentrations are found in warmer air masses higher in ozone. This could be explained by increased condensation of vapour phase into particulate phase at lower air temperatures (Kim et al., 2009).

The low (i.e. strongest) AMDEs, which constituted 18% of the AMDEs, are anti correlated with meridional wind in March. The topography in Ny-Ålesund causes funnelling effects (Solberg et al., 1996), and contradictory to the anti correlation with meridional wind, the 7-day back trajectories arriving in Ny-Ålesund during the strongest AMDEs indicate that the air parcels originated from the Arctic Ocean (Fig. 7). The anti correlation for the strongest AMDEs with relative humidity in March suggests that only a minimum atmospheric moisture contents is required for depletion of GEM to occur. Similar results were found by Cobbett et al. (2007) at Alert.

The majority of fairly low correlation coefficients calculated for AMDEs indicate that the AMDEs in this study are mainly of non local origin, with GEM being transported to Zeppelin by a wide variety of different air masses.

4 Conclusions

The study revealed a clear seasonal distribution previously undiscovered at Polar sites. PHg was exclusively observed from March through April. From May through August, increased RGM concentration was observed. Surprisingly, RGM was not solely formed during AMDEs. It was speculated whether local oxidation of GEM by BrO was responsible for the formation of RGM. The AMDEs were mainly of non local origin with GEM being transported by a wide variety of different air masses. With some exceptions, no clear meteorological regime was found for the GEM, RGM and PHg concentrations. It seems possible that atmospheric mercury acts differently across Polar Regions.

This study gives new valuable insight into the atmospheric mercury speciation, of outmost importance for mercury as a global pollutant. In order to further improve the knowledge this study proposes three future research directions:

- Long term atmospheric mercury speciation measurements: Previously work is generally restricted to periods of weeks to months, and this study is a clear evidence that long term measurements (> year) will uncover new aspects concerning the temporal distribution of atmospheric mercury speciation. Comparison of the atmospheric mercury speciation at several polar sites is also desirable.
- Evaluation of the seasonal distribution of RGM and PHg: Previously work is mainly conducted during spring due to the occurrence of AMDEs. However, this work revealed a clear seasonal distribution which stresses the importance year-round measurements. Summer measurements at other polar measurement sites could aid in the third suggested research direction.
- Formation mechanism for RGM: RGM was observed at increased concentrations during spring and summer in the duration of this study. BrO and O₃, despite the slow GEM-O₃-reaction rate, were suggested as oxidants for GEM. Still the formation mechanism remains less clear.

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Table 1. Thresholds for high and low concentrations used in the correlation analysis.

Species	Low (\leq)	High (\geq)	Concentrations
GEM (ng m^{-3})	1.55	1.75	≥ 0.36
RGM (pg m^{-3})	10.37	26.05	≥ 7
PHg (pg m^{-3})	8.55	45.02	≥ 7
AMDE (GEM, ng m^{-3})	0.54	0.85	$\geq 0, < 1$

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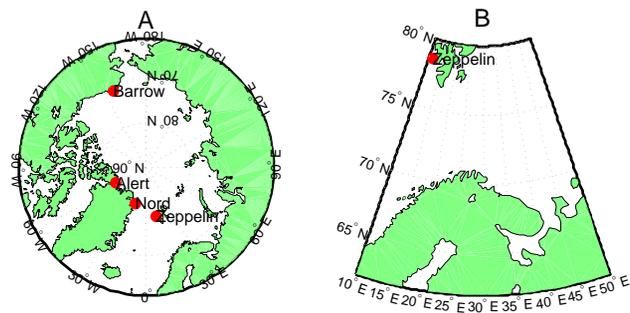


Fig. 1. (A): Map indicating the location of Polar study sites for atmospheric GEM, RGM and PHg measurements. (B): Map indicating the location of Ny-Ålesund/Zepelin, Svalbard.

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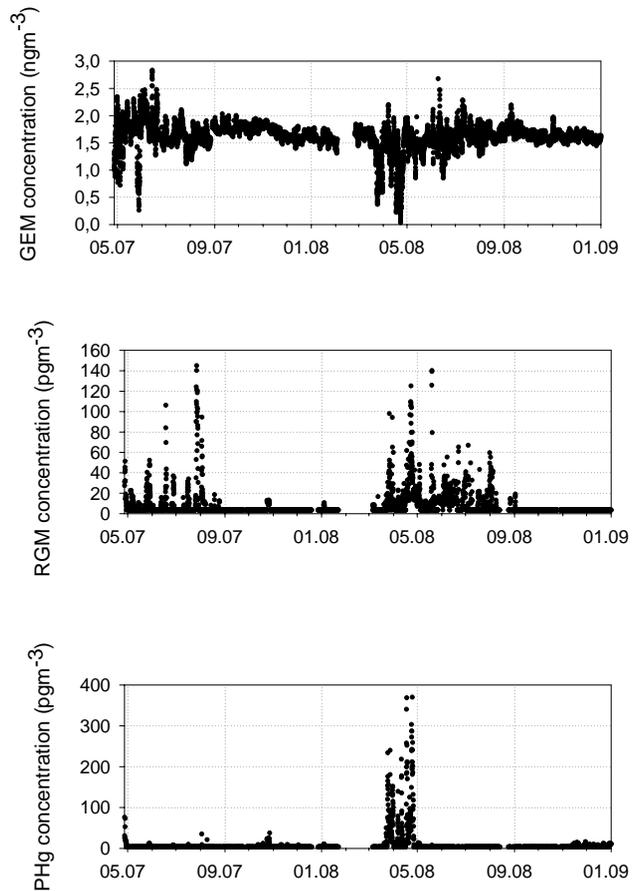


Fig. 2. Hourly average GEM concentrations and two hour average concentrations of RGM and PHg.

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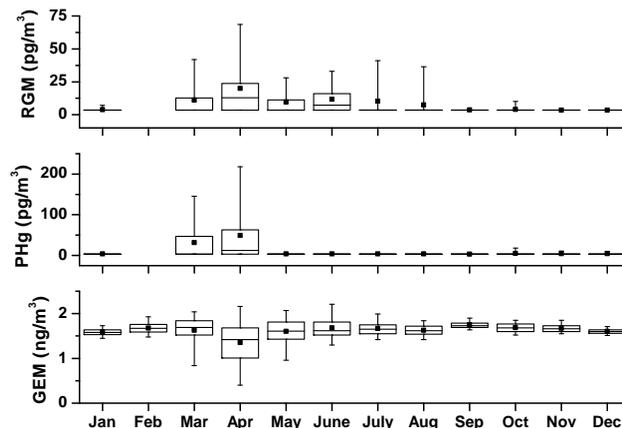


Fig. 3. Seasonal distribution of the GEM, RGM and PHg concentrations for 2007 and 2008. The box represents the monthly 25th and 75th percentiles of the complete dataset, the line represents the monthly median concentrations, and the black square represents the monthly average concentrations. The whiskers represent the monthly 10th and 90th percentiles of the complete dataset.

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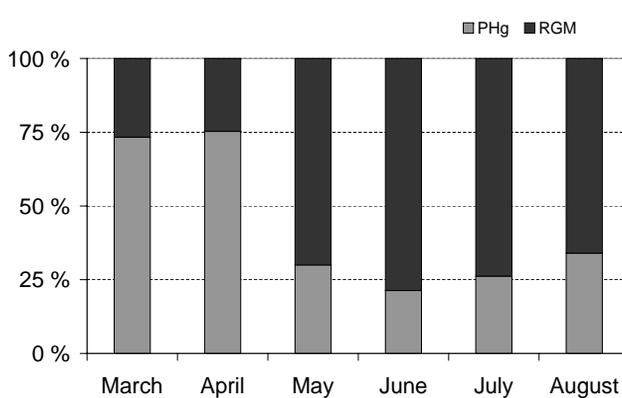


Fig. 4. The monthly distribution of RGM and PHg from March through August.

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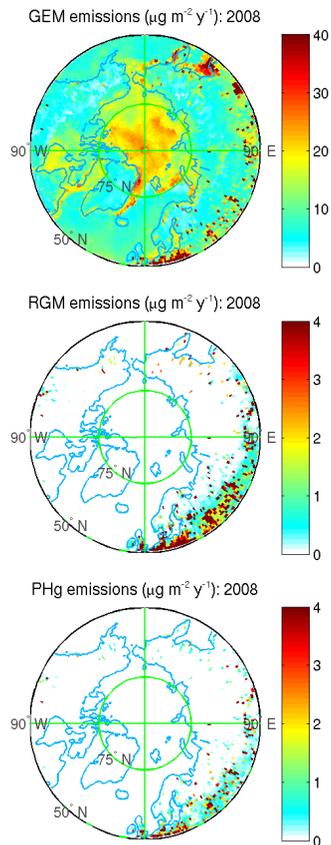


Fig. 5. Northern Hemisphere high-latitude mercury emissions for 2008, as seen by Environment Canada's Global/Regional Atmospheric Heavy Metals model.

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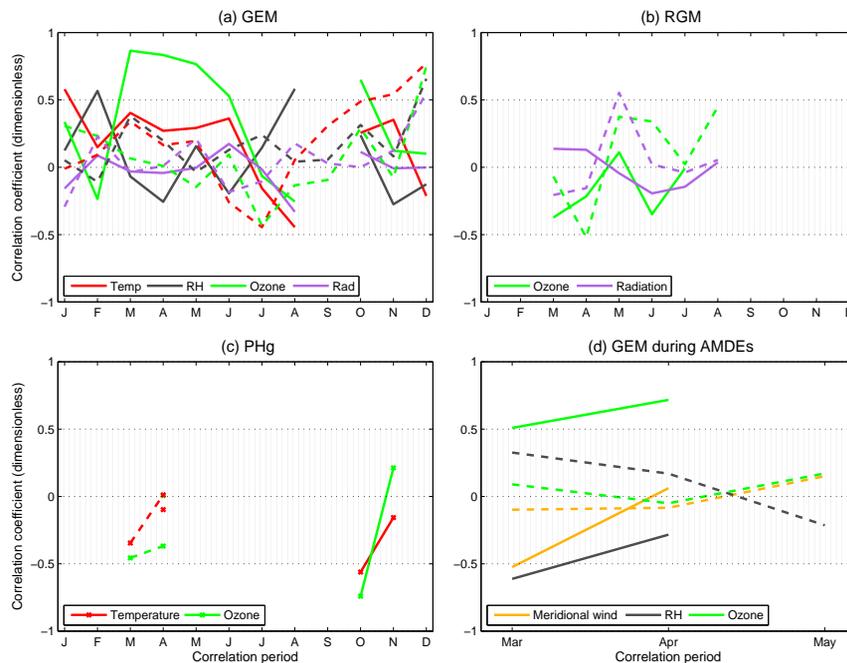


Fig. 6. Correlations of GEM, RGM and PHg concentrations with air temperature (Temp), relative humidity (RH), ozone, global radiation (Rad) and meridional wind. Solid lines represent correlation with low concentration (<25th percentile of the complete dataset) and dashed lines represent correlation with high concentration (>75th percentile of the complete dataset). Figure 6a shows strong positive correlation of ozone with low GEM concentrations from March through June. A strong correlation of air temperature with low GEM concentrations was observed in January whereas relative humidity correlates with low GEM concentrations in February and August. In November and December air temperature correlates with high GEM concentrations. In December relative humidity, ozone and global radiation are strongly correlated with high GEM concentrations. (b) shows a significant anti correlation of ozone with high RGM concentrations in April. High RGM concentrations correlate with global radiation in May. (c) shows a strong anti correlation of air temperature and ozone with low PHg concentrations in October. (d) shows a strong anti correlation of meridional wind and relative humidity with strong AMDEs in March. A strong correlation of ozone with strong AMDEs was observed from March through April.

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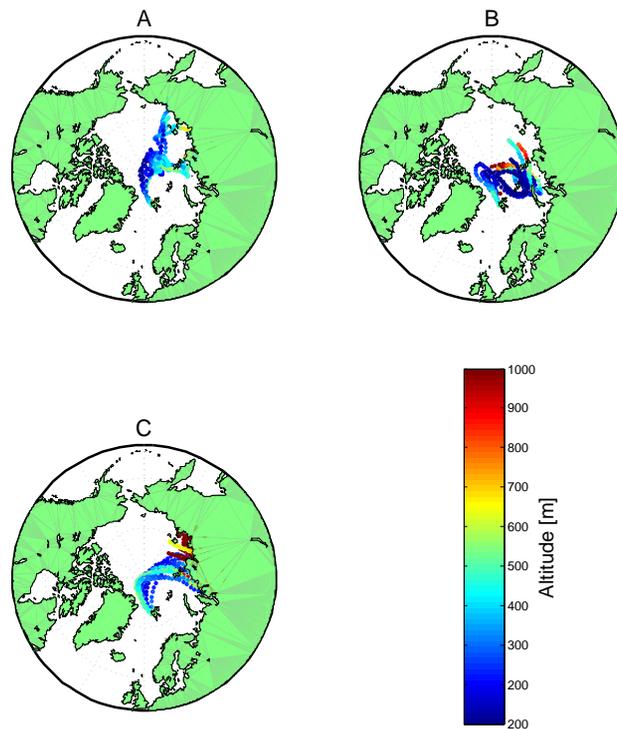


Fig. 7. 7-day back trajectories arriving in Ny-Ålesund during the strongest AMDEs in 2008 ($GEM < 0.54 \text{ ng m}^{-3}$). **(A):** 24 March–25 March, **(B):** 17 April–18 April, **(C):** 21 April–23 April.