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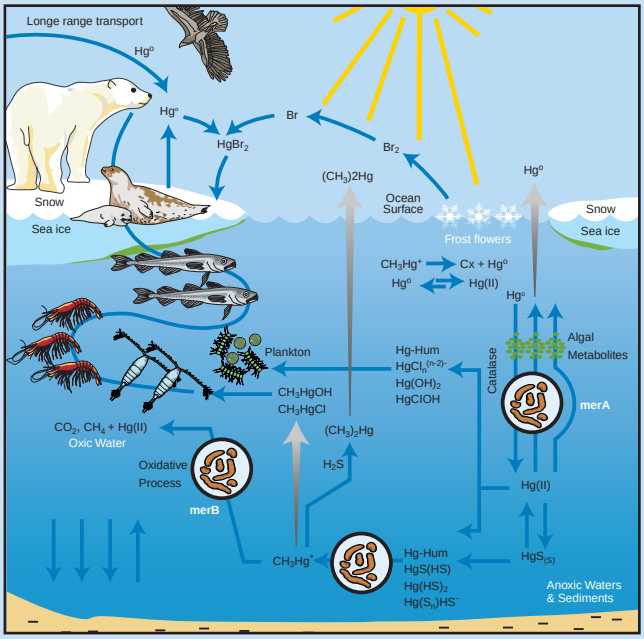
Fate of mercury in the Arctic (FOMA)

Sub-project Atmosphere

NERI Technical Report No. 533



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Data sheet

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Abstract:	The main source of mercury in the Arctic is long range transport from mid latitudes. In order to understand the dynamics of the source strength in the Arctic a series of analytical methods is developed. For example the first flux measurements ever of RGM have been carried out together with flux measurements of GEM. The results are used to make a new parameterisation of the chemical and physical processes and model calculations are performed for the first time of the input of atmospheric mercury to the Arctic.
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Contents

Preface

This report is the final report of the project FOMA; Subproject Atmosphere, funded by the Danish Environmental Protection Agency with means from the MIKA/DANCEA funds for Environmental Support to the Arctic Region. The aim of the project is to study the inter-compartment mercury transport chain in the Arctic area. This part of FOMA describes the atmospheric processes controlling Arctic mercury and atmospheric deposition of mercury on snow and ice surfaces leading to enhanced mercury levels in the Arctic environment.

The subproject Atmosphere was planned and carried out by Department of Atmospheric Environment at NERI in close co-operation with other Subprojects carried out by two other NERI departments: Dep. of Arctic Environment, and Dep. of Marine Ecology (Skov *et al.*, 2004a). Furthermore the University of Southern Denmark and the University of Copenhagen took directly part in the present subproject.

A series of international field campaigns was the basis for the subproject and it was carried out in close co-operation with the National Oceanic and Atmospheric Administration, USA; Oak Ridge National Laboratory, USA; NILU, Norway and Meteorological service of Canada. These activities were carried out under the umbrella of Arctic Monitoring and Assessment Programme (AMAP).

Chapter 1 of the report gives a short introduction to atmospheric mercury in the Arctic. A more thorough introduction is found in the FOMA report (Skov *et al.*, 2004a) where the atmospheric part is described together with the other matrixes (Marine and biological matrixes). Chapters 2, describes the measurement sites; Chapter 3 describes the analytical instruments and techniques used. Chapter 4 presents a discussion of the results. Chapter 5 sums up the conclusion of the work and Chapter 6 presents recommendations for future works.

Sammenfatning

Kviksølv har normalt en levetid på 1 år eller mere i atmosfæren. I det arktiske forår forkortes levetiden imidlertid til timer pga. hurtige kemiske reaktioner, der omdanner gasformigt elementært kviksølv (GEM) til reaktivt gasformigt kviksølv (RGM) under "Atmospheric mercury depletion events" (AMDEs). Efterfølgende deponerer RGM til sneoverflader, hvorved kviksølv fjernes fra atmosfæren. Det er vigtigt at få kvantificeret og forstået disse processer, da kviksølv har en negativ påvirkning på miljøet (Skov *et al.*, 2004a).

Der er derfor udviklet en serie nye metoder, der kan anvendes til at måle fraktioner af atmosfæriske kviksølv med en højere tidslig eller geografisk opløsning end hidtil muligt. Anvendes disse metoder sammen med mikrometeorologiske teknikker, kan man bestemme den totale afsætning af atmosfærisk kviksølv i Arktis med høj tidsopløsning. Dette er vigtigt dels for at etablere en massebalance for atmosfærisk kviksølv og dels for at få en god forståelse af de vigtigste atmosfæriske processer som efterfølgende anvendes, til en generalisering af resultaterne med atmosfærekemiske transportmodeller, eksempelvis Danish Eulerian Hemispheric Model (DEHM).

I rapporten præsenteres de første fluxmålinger i verden af reaktivt gasformigt kviksølv (RGM) og fluxen af gasformig elementært kviksølv (GEM). Fluxmålingerne viser, at AMDEs fører til en kraftig forøgelse af kviksølvbelastningen til det arktiske miljø. Dette sker gennem omdannelse af GEM til RGM efterfulgt af deposition af RGM til sne. Resultaterne er anvendt til at lave en ny parameterisering for afsætningen af atmosfærisk kviksølv i kemimodulet i DEHM. Til arealet nord for polarcirklen er der således estimeret en deposition på ca. 200 tons/år kviksølv, når AMDEs er medtaget. Til sammenligning giver modelberegninger uden en parameterisering af AMDEs en afsætning på kun ca. 90 tons/år.

Måleresultaterne i dette projekt viser, at dynamikken hvorved kviksølv afsættes, er mere kompliceret end beskrevet med den nuværende model parameterisering. GEM oxideres sandsynligvis tæt ved eller direkte på sneoverfladen. Havsalt fra frosne våger er ansvarlige for den samtidige fjernelse af både GEM og ozon. GEM omdannes til RGM, der efterfølgende hurtigt fjernes fra atmosfæren ved deposition til overfladen.

Eqikkaaneq

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Summary

Usually mercury has a lifetime of about 1 year or more in the atmosphere. However, during Arctic spring the lifetime is only a few hours due to fast atmospheric processes that convert gaseous elemental (GEM) to reactive gaseous mercury (RGM) during Atmospheric Mercury Depletion Events (AMDEs). Then RGM is removed quickly from the atmosphere by deposition. It is important to quantify and understand the processes responsible for the removal of mercury from the atmosphere due to the negative environmental impact of mercury (Skov *et al.*, 2004a).

A series of methods is developed for measuring the fractions of atmospheric mercury with higher temporal or spatial resolution than previously. If these new methods are applied together with micrometeorological methods, they make it possible to measure the flux of Total Atmospheric Mercury (TAM) in the Arctic. This is important in order to get a mass balance for atmospheric mercury and to establish a good understanding of the processes that can be used to make a new chemical parameterisation for the model. The parameterisation can then be used to generalise the results in atmospheric chemical transport models e.g. the Danish Eulerian Hemispheric Model (DEHM).

In the present project the first flux measurements ever of RGM have been carried out together with flux measurements of GEM. These measurements show that AMDEs increase the mercury burden in the Arctic. This occurs though the conversion of GEM to RGM followed by fast deposition of RGM to the snow. The results are applied in a new parameterisation of atmospheric mercury in DEHM. North of the Polar Circle about 200 tons/year is deposited when AMDEs are included. A model calculation of mercury deposition without AMDE only gave about 90 tons/year.

Experimental results show that the dynamics of atmospheric mercury are far more complicated than described by the present model parameterisation in DEHM. Most probably GEM is oxidised close to the surface or directly on the surface. Sea salt from refrozen leads together with sunlight are responsible for the GEM removal and for the simultaneously removal of ozone. In those processes RGM is formed. Subsequently RGM is quickly removed from the atmosphere by deposition.

1 Introduction

Main source

The main source of mercury in the Arctic area is due to long range transport from mid latitudes where most emissions are taking place (Pacyna & Pacyna, 2002a; Hylander & Meili, 2003). Gaseous elemental Mercury (GEM) dominates the emission of mercury from anthropogenic sources with minor contributions of reactive gaseous mercury (RGM) and fine particulate mercury (FPM).

Atmospheric lifetime

The lifetime of mercury outside the Arctic is a matter of dispute. Estimates vary between about 1 year (Lin & Pehkonen, 1998; Schroeder *et al.*, 1998a) and down to 15 days, where the lower limit is obtained using the results of the latest study of the rate constant of Hg^{o} with O_3 (Pal & Ariya, 2004). However, field observations of gaseous elemental mercury (GEM) (e.g. Skov *et al.*, 2004l) disagrees with the short lifetime of Pal and Ariya.

Re-emission

In any case, a significant amount of mercury deposited from the atmosphere is re-emitted to the atmosphere, the so-called hopping. In this way atmospheric mercury is transported over long distances. As a result mercury is an ubiquitous pollutant with a background concentration of close to 1.5 ng/m^3 throughout the year. Therefore, mercury is transported from the source areas at mid latitudes to the Arctic.

Arctic Spring

During arctic spring, atmospheric mercury depletion events (AMDE) have been observed where GEM is quickly converted to reactive gaseous mercury (RGM) (Schroeder *et al.*, 1998b; Lindberg *et al.*, 2001; Berg *et al.*, 2003; Skov *et al.*, 2004k) and in Subarctic spring, (Poissant & Pilote, 2003). RGM is either deposited to the surface leading to mercury accumulation in the Arctic or alternatively transformed into fine particulate mercury, FPM, with a longer atmospheric lifetime than RGM, see Figure 1.1.

Aim

The aim of the atmospheric part of FOMA is to determine the ratio between the mercury deposition and RGM transformation to FPM. The results from the study of the processes are in turn parameterised and implemented in the Danish Eulerian Hemispheric Model in order to make the first rough estimate to the Arctic (Christensen *et al.*, 2004c; Skov *et al.*, 2004j).

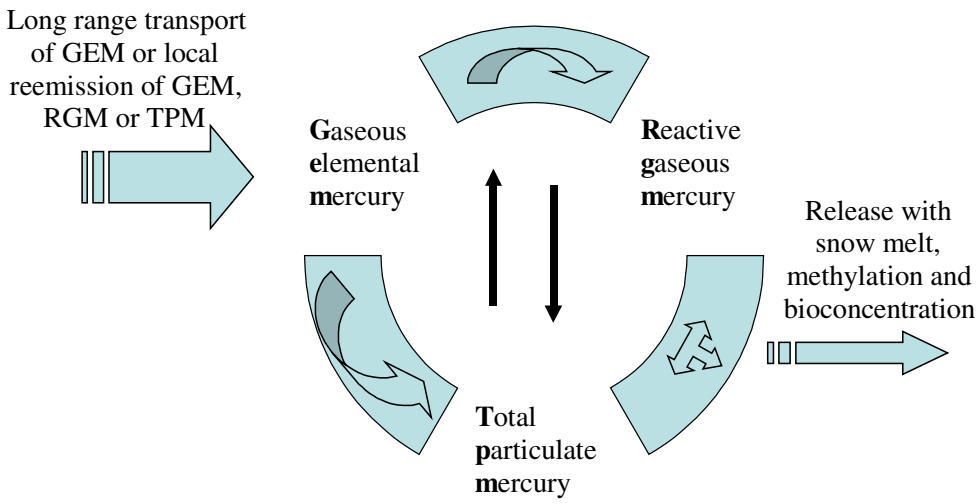

Figure 1.1 A simple scheme of the processes of atmospheric mercury in the Arctic during AMDE

Flux measurements

In 2001 flux measurements of RGM were carried out in a campaign at Barrow, Alaska for the first time in the world (based on basic funding and funds from SNF (Skov *et al.*, 2005). The work was a co-operative study between NERI and NOAA, ORNL and US-EPA, USA. In order to document the findings and further investigate the mechanisms of the deposition, additional campaigns were carried out in 2002, 2003 and 2004. The 2003 campaign at Barrow was fully financed by DANCEA and the 2004 campaign was partly financed by DANCEA. Furthermore, a 2002 campaign at Station Nord was partly financed by DANCEA

Final Report

The present report presents the obtained results and serves as the final report for the FOMA-subproject Atmosphere. The report focuses on the processes that are used to determine the input flux of mercury to the Arctic, but results from development etc. of new methods are also mentioned, as they are very important for future investigations of the fate of atmospheric mercury in the Arctic.

Co-operation

In this connection two studies were carried out in co-operation with University of Southern Denmark. In the first study a diffusive sampler is developed (Daugaard *et al.*, 2005) and in the other the methods already in use were tested and new methods were developed (Hansen *et al.*, 2005). These two studies are only briefly described in the report and shortly two manuscripts will be submitted to Journal of Environmental Monitoring.

2 Experimental sites

In this chapter the three sites are described where the fate of mercury is studied. Figure 2.1 shows the location on a map over the Northern Hemisphere with the most important Arctic Stations.

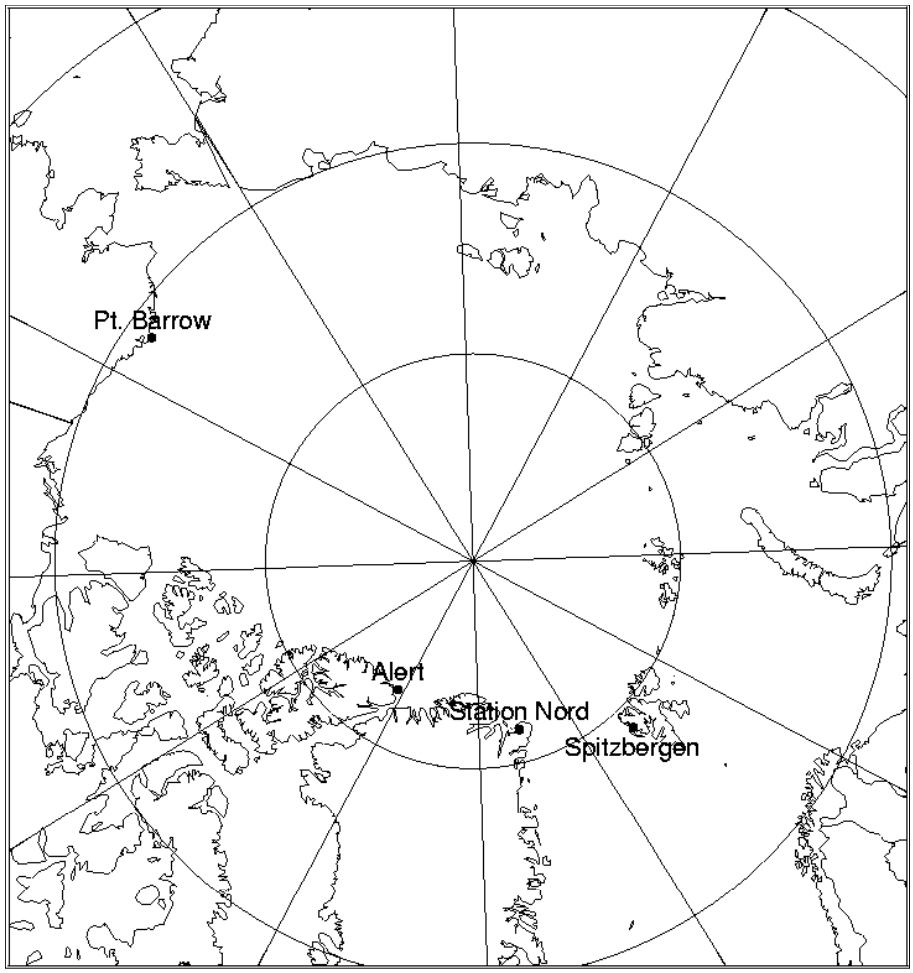

Figure 2.1 The Northern Hemisphere with the Arctic Stations. Barrow and Station Nord are used in the mercury study presented here.

2.1 Barrow

CMDL site

The monitoring site is a NOAA Climate Monitoring and Diagnostic Laboratory (CMDL) hut at 71°19′ N, 156°37′ W. About 10 km Southwest of CMDL is the town Barrow. The CMDL is located near the peninsula at Point Barrow, approximately 2 km inland from the shoreline, and is surrounded primarily by water to the north, east, and west.

Activities

At the station there is a 20 m mast where flux measurements of RGM were carried out using relaxed eddy accumulation (REA) and gradient measurements of RGM together with gradient measurements of GEM and ozone. At the station basic meteorology was available and finally GEM, RGM and in periods also FPM were measured.

2.2 Lille Malene Mountain, Nuuk

Location

Lille Malene Station is located at 345 m above sea level and 64°10'46.4"N, 51°43'33.9"E and is thus a Sub-Arctic station, see Figure 2.2. Lille Malene is close to Nuuk, the capital of Greenland with 13,500 inhabitants and to the airport of Nuuk.

Figure 2.2 Greenland with main cities including the Capital Nuuk.

Activities

In the spring 2004 a field campaign was carried out where new methods were tested for measuring the various fractions of mercury. Ozone, GEM and NOx measurements were started at Lille Malene, Nuuk in January 2002 but due to technical problem measurements were first continuously performed from 2003.

2.3 Station Nord

Location

The monitoring has taken place at Station Nord, a small military air field located in north-eastern Greenland at 81^{0}36' N, 16^{0}40'W and only accessible by courtesy of the Royal Danish Airforce, see Figure 2.3a and b.

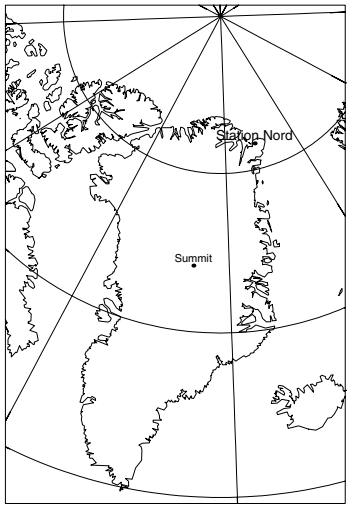

Figure 2.3a Greenland with the location of Station Nord.

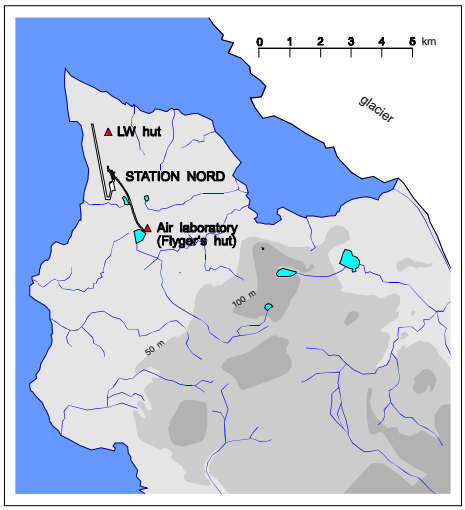

Figure 2.3b The position of the air monitoring site Flyger's Hytte at Station Nord.

Minimum of local sources

The location of the AMAP station has been chosen on the basis of the characteristics of the site, described previously (Heidam *et al.*, 1999), so as to minimise influence from any local air pollution. The measurements were carried out at the site 'Flyger's Hut', which is located approximately 3 km south of the central complex of buildings, as shown on the map in the Figure 2.3. The building is supplied with electricity and heat from Station Nord's local diesel generator and served as the main base for the Danish AMAP Air Monitoring Programme until July 2002.

3 Experimental section

Ozone

Ozone was measured with an UV absorption monitor, API, with a detection limit of 1 ppbv and an uncertainty of 3 % for concentrations above 10 ppbv and 6 % for concentrations below 10 ppbv, see Table 3.1 (all uncertainties are at a 95% confidence interval) (Skov *et al.*, 1997).

GEM

A TEKRAN 2537A mercury analyser measured GEM. The principle of the instrument is as follows: A measured volume of sample air is drawn through a gold trap that quantitatively retains elemental mercury. The collected mercury is desorbed from the gold trap by heat and is transferred by argon into the detection chamber, where the amount of mercury is detected by cold vapour atomic fluorescence spectrometry. The detection limit is 0.1 ng/m^3 and the reproducibility for concentrations above 0.5 ng/m^3 is within 20% (at a 95% confidence interval) based on parallel measurements with two TEKRAN 2537A mercury analysers. It is not at present possible to give the combined uncertainty of the method following the guidelines of ISO 14956, as the exact identity of the measured mercury is unknown, though GEM is determined as the dominant compound. In order to protect the instrument against humidity and sea salt, a soda-lime trap was placed in the sample line just in front of the analyser before the 2001 season to avoid passivation of the gold traps (Skov *et al.*, 2004i), and a heated sample line was used as well. However, no change in the level of GEM at Station Nord was observed after the installation of the trap and heated line. Parallel measurements of GEM in Denmark at a site not directly influenced by sea spray with and without soda lime trap showed perfect agreement within the experimental uncertainty.

Table 3.1 The uncertainty of the measurements of the various species at Barrow, Alaska. For all the mercury species the uncertainty is estimated from reproducibility experiments

	Ozone monitor	Tekran GEM	Tekran RGM	Tekran FPM	RGM Flux	GEM Gradient
Unit	ppbv	ng/m^3	pg/m^3	pg/m^3	pg m^{-2} min^{-1}	ng/m^3
Range	0-50	at 0.5	0-1000	0-50	-22 to 30	at 0.5
std. dev.	6%	20%	20%	40%	52%	33%

Ozone gradient

Ozone gradients were measured by sampling at 20 m and 0.1 m above the snow surface. The sample lines were of equal length. An external pump provided a constant flow in the two tubes of 20 Litre/min. A Thermo Environmental Instruments model 49 Ozone Analyzer measured ozone alternating 10 minutes at 20 m and then 10 minutes at 0.1 (Brooks *et al.*, 2005; Skov *et al.*, 2004a).

The principle of RGM Flux using REA

RGM fluxes were measured by relaxed eddy accumulation, REA, see Figure 3.1. A sonic anemometer measures the vertical wind speed and through the connection to a fast shifting valve system, air samples can be sampled in upward and in downward air. The difference in the concentrations in the two channels is proportional to the flux, F as expressed in equation 3.1.

$$F = \beta\sigma_w(C_{up} - C_{down}) \qquad (3.1)$$

where β is an empirical constant, σ_w is the standard deviation of the vertical wind velocity, and C_{up} and C_{down} are respectively the concentrations in the upward and downward air. When the vertical wind speed was close to 0 cm/sec a third channel was opened, the so-called dead band. Annular denuders for the chemical sampling were used (Landis *et al.*, 2002e).

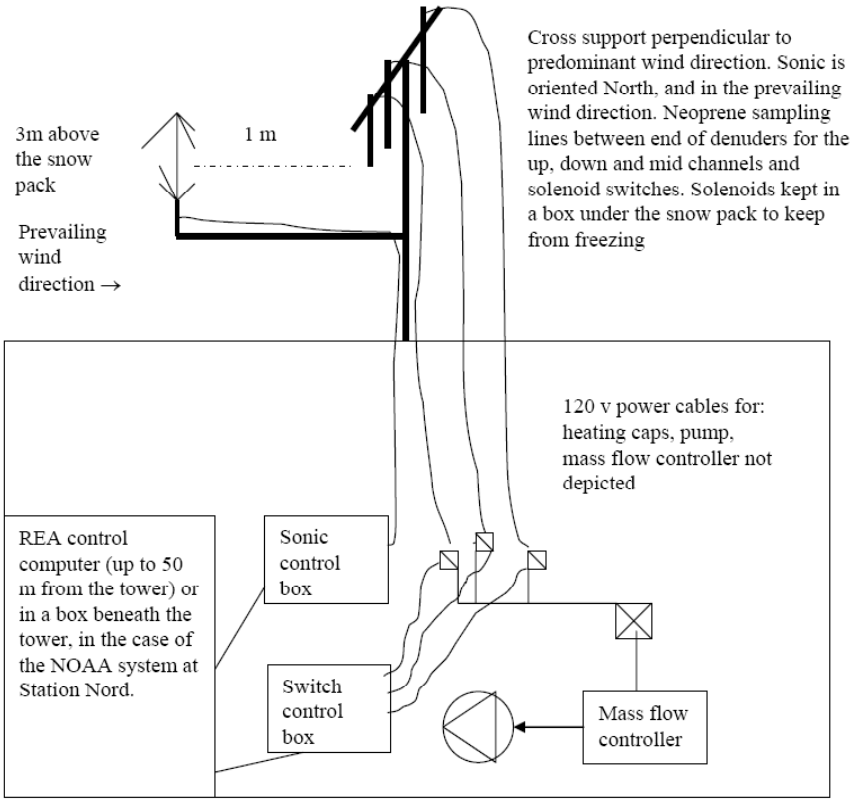

Figure 3.1 A sketch of the RGM-REA system (Goodsite, 2003))}.

Sonic anemometer

A sonic anemometer (in 2001 a Metasonic and the rest of the years a RM Yong) provided atmospheric airflow data with a frequency of 10Hz but in the present configuration only a 1 Hz signal output was used. In this way 95% of the turbulence was captured (Tilden Meyers Private communication). This was the best compromise between the meteorological measurements and the chemical sampling. The slow shifting frequency ensured laminar conditions in the denuders, which is necessary for them in order to work properly.

RGM

RGM was measured and analysed by using the method of (Landis *et al.*, 2002d). The Landis et al. method for RGM determination uses a KCl coated quartz annular denuder sampling chain heated to 50 ^{0}C to sample mercury in air. The detail is described in Landis et al. and the method will only be briefly described here.

RGM denuders

At the inlet, there is an elutriator and an impactor, with impactor plate. The elutriator is coated with cross-linked Teflon since RGM is considered very "sticky" but will not adsorb to the cross- linked Teflon. The elutriator straightens the flow and accelerates it, by forcing it through an orifice onto a roughened impactor plate, which is not coated on the surface. The cut-off diameter is 2.5 µm, so only PM2.5 (particles with a diameter less than 2.5 µm) flows past the active area of the denuder. The sample flow is 10 litres per minute. The flow is controlled just prior to the denuder chain with a "dry cal" flow meter before and after sampling.

Immediately following the impactor, there is a dead space prior to the annulus. This allows for expansion of air, from ambient to 50°C, since KCl optimally collects RGM at this temperature (Landis *et al.*, 2002c); as well as a proper development of laminar flow, which is a necessary condition for proper functioning of denuders.

Denuders had a collocated precision of 15.0±9.3 % with 2 times standard deviation in agreement with the findings reported in Landis et al.

The heating mantels employed for the sampling system are different from those used by Landis et al., since the mantels were judged to be too bulky for use in the flux system.

The heating mantles consist of a PVC inner tube that encloses the denuder from the tip of the inlet to the top of a filter pack at the outlet. The outer portion of the pipe is wrapped with a silver tape to ensure heat transfer from self-regulating heat tape.

The tube was placed inside a larger PVC pipe, allowing 5 cm spacing between the tubes. The overall diameter was approximately 9.6 cm. The space between the two shells was then filled with self-expanding polyurethane foam for thermal insulation. Upon drying, the insulation was cut, so that top and bottom end caps will fit snugly. The insides of the top and bottom caps are filled with foam cut to fit the inlet and outlet of the denuder. The heating mantle is sealed at each end with silicone, so that it is watertight. Other shells than PVC may be used but the present solution was chosen for cost efficiency reasons. The result is a self-regulating heating mantle capable of producing the typical 80°C effect required in the Arctic spring for an efficient active coating temperature of 50°C. Therefore the mantle temperature was constant during sampling in the campaign.

RGM analysis

After sampling, the quartz denuders were closed immediately with plastic caps equipped with Teflon inner seals, and taken into the laboratory for thermal desorption and detection with a TEKRAN 2537A following the procedure of (Landis *et al.*, 2002b).

During analysis, care was taken to keep the ends of the denuders cooled while the active area was being heated. At the outlets of the oven the denuder tube was isolated with quartz pads and the ends of the denuder that extend out of the oven were cooled using co-axial fans. In this way, only RGM captured on the active area was desorbed.

Field blanks

For all measurements a field blank was obtained by handling a denuder in the field. Hg mass from this field blank was subtracted from the measured Hg masses on the exposed denuders. If there was any indication of Hg(0) adsorption, for example with sudden sharp increases in Hg amounts then the denuder was cleaned and re-coated, since as pointed out by (Sheu & Mason, 2001) just 1% of Hg(0) adsorption on a denuder is enough to compromise RGM measurements.

Uncertainty

The accuracy of the denuders was found to be in agreement with those reported by (Landis *et al.*, 2002a). In the campaign, the US-EPA manual denuders exhibited a precision of 10%, based on co-located parallel measurements, and were on average within 25% of the automated RGM sampling system running separately. In 2004 several RGM systems measured in parallel and the agreement between those measurements was very poor and furthermore the deviation was random. The difference between the RGM measurements might be due to contaminants internal in the TEKRAN monitors.

Unfortunately the NERI instrument broke down after it had been running over night at –20°C.

Gradients as supplement

The REA system broke down as well on one of the first days of the 2004 campaign due to extreme weather conditions with temperatures increasing from –20°C to –4°C and as a consequence the temperature in the box rose to above 40°C. The –4°C was record high temperature on the day. Fortunately, gradient measurements of RGM were carried out in 2004 for supporting the REA results. RGM was sampled at 4.15 m, 8.69 m and 14.15 m height above the snow surface.

Mercury fractionation

Concentrations of GEM, RGM and in periods at Barrow also FPM were measured by the set-up from TEKRAN consisting of TEKRAN 1130 unit for RGM, followed by a TEKRAN 1135 for FPM and with a TEKRAN 2537A mercury analyser. The principle is shown in Figure 3.2

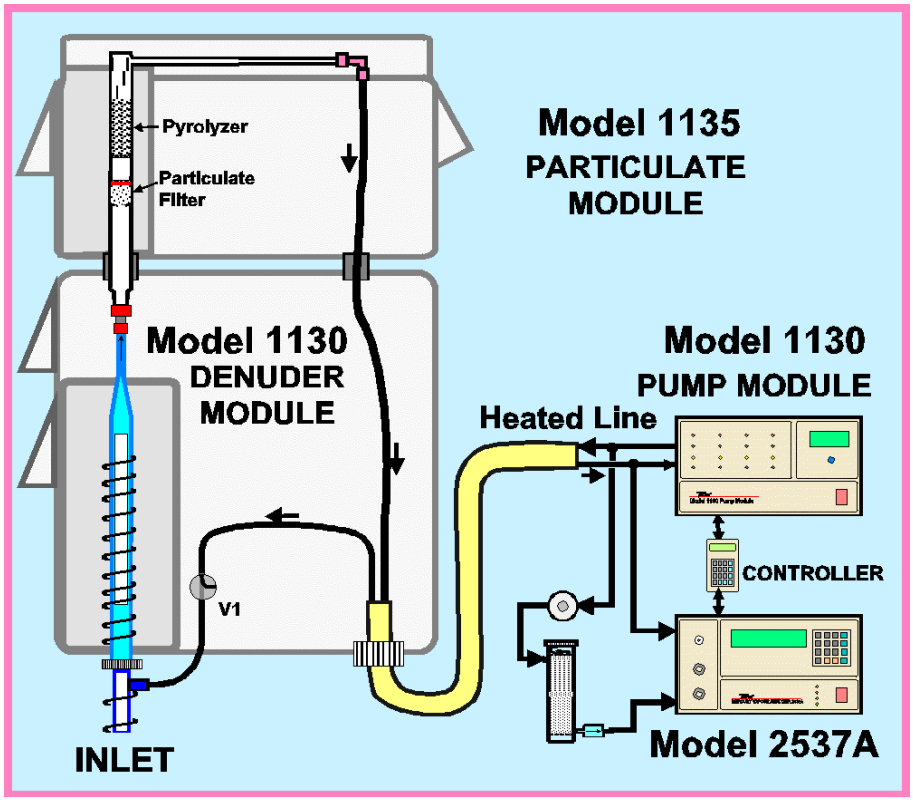

Figure 3.2 TEKRAN fractionation system consisting TEKRAN 2537A cold vapor atomic fluorescence mercury analyzer, equipped with TEKRAN 1130 RGM sampler and 1135 FPM sampler. From www.tekran.com.

4 Results and discussion

4.1 Atmospheric mercury during springtime in Nuuk, Greenland

Continuous monitoring of GEM and ozone

Measurements of GEM and ozone have been performed since the beginning of 2003 on the Lille Malene Mountain just outside Nuuk the capital of Greenland. The results show that depletion of elemental mercury to some extend occurred at this Subarctic site during May in 2003 and in April 2004 (only 2003 data shown here).

Campaign

GEM, total atmospheric mercury (TAM), fine particulate mercury (FPM) and RGM were studied in a more intensive campaign in May 2004.

AMDE

AMDEs are also observed at Nuuk in the Sub-Arctic in the spring of 2003 and 2004, though much weaker than at more northern positions, see Figure 4.1.

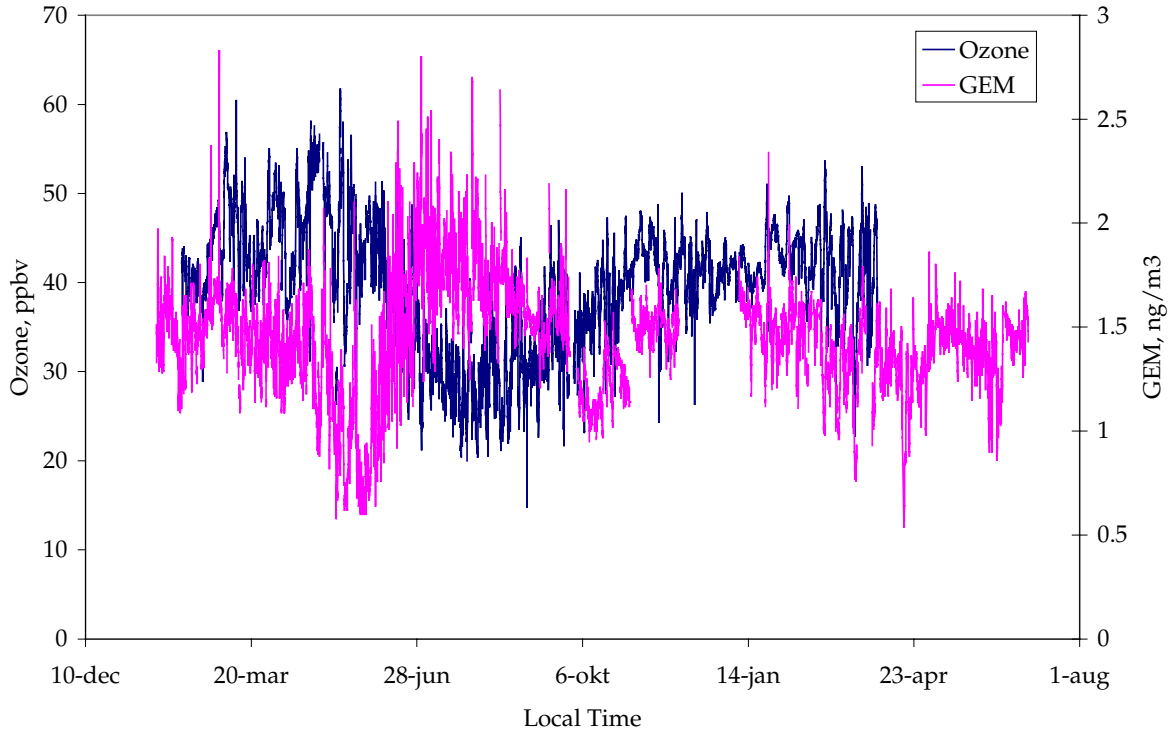

Figure 4.1 Ozone and Gaseous elemental mercury (GEM) at Lille Malene Station Nuuk Greenland from January 2003 to end of June 2004.

GEM and ozone are not correlated during AMDEs as in the Arctic. Long-range transport of air masses with depleted GEM concentrations appears to be a good explanation for AMDE at Nuuk. This interpretation is supported by the very low concentrations of filterable bromine at 1 ng/m^3 compared to 20 ng/m^3 observed at Station Nord (Skov *et al.*, 2004h).

4.1.1 Fractionation of atmospheric mercury

TAM

The fractionation of mercury has been investigated through measurements of GEM, TAM, RGM and fine particulate mercury. The measurements were performed in May 2004 and thus after the AMDEs took place. Atmospheric mercury was found to be almost solely GEM. RGM and FPM are in the range from 0-3 pg/m^3, which are typical background values outside the AMDEs. Slightly elevated RGM and TAM concentrations were observed when the measurement site was affected by the plume from the nearby waste incinerator or from the urban plume from Nuuk, this is illustrated for TAM in Figure 4.2.

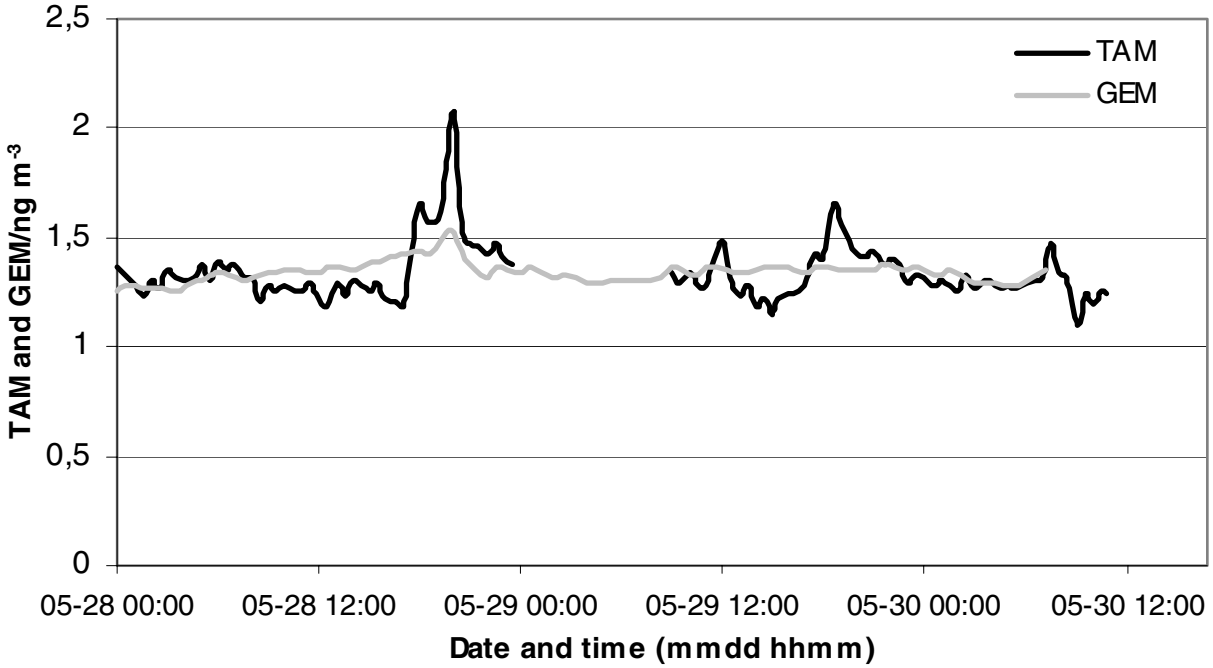

Figure 4.2 Concentrations of GEM and TAM measured in May 2004 at The Greenland Institute of Natural Resources in Nuuk.

4.1.2 Conclusion

AMDEs are also observed at Nuuk in the Sub-Arctic in the spring of 2003 and 2004. GEM and ozone are not correlated during depletion as in the Arctic and thus GEM and ozone are not directly coupled. Long-range transport of air masses where AMDEs have already taken place appears to be a good explanation for AMDE at Nuuk. Therefore it is impossible to investigate the mechanisms responsible for AMDE at this site. The speciation of mercury has been investigated through measurements of GEM, TAM, RGM and fine particulate mercury. All results indicate that mercury almost solely exists in its elemental and RGM and FPM are in the range from 0-3 pg/m^3, which are typical background values outside the AMDEs. Slightly elevated RGM and TAM concentrations could be observed when the measurement site was affected by the plume from the nearby waste incinerator or from the plume from Nuuk.

4.1.3 Development of a diffusive sampler for gaseous mercury

High uptake diffusive sampler

A high uptake diffusive sampler for atmospheric mercury is developed. This sampler can measure the low atmospheric concentrations of mercury with a time resolution of max 1 week. Previously, no other sampler has been able to do that. The sampler consists of a porous cylinder and a gold core. This type of sampler is a Radiello sampler, see Figure 4.3.

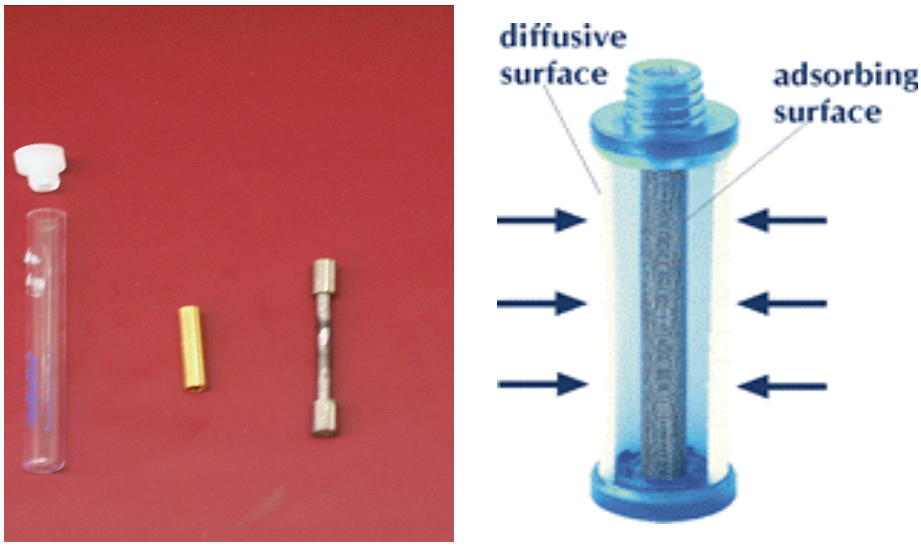

Figure 4.3 Left; Gold core that adsorbs mercury, supporting iron core and glass tube with plastic cap to store the gold core before and after sampling. Right; Radiello diffusive body in polyethylene.

Measurement sites

The sampler was first developed for aromatic species (Cocheo *et al.*, 2000) and here it is developed further to be used also to measure gaseous elemental mercury (Daugaard *et al.*, 2005). The sampler was used on Faeroe Islands, at Nuuk and at NERI Roskilde site. An uptake rate at room temperature is about 80 ml/min and thus 24 hour exposure should be sufficient. However, at present only 1 week samples have been carried out.

The diffusive sampler can in future studies be used to measure the geographical distribution of mercury concentrations in ambient air. It is thus planned to measure GEM as function of distance from an open lead. In spring 2005 we plan to carry out this project.

4.2 Station Nord Campaign

Measurement complications

The weather conditions during the spring of 2002 made it impossible to measure REA fluxes at Station Nord due to lack of wind simultaneously with the appearance of AMDE. The campaign was held from 1 to 27 April, 2002 and there were short depletion periods (from ozone measurements) throughout the campaign see Figure 4.4. However the first large depletion event came on the day of departure.

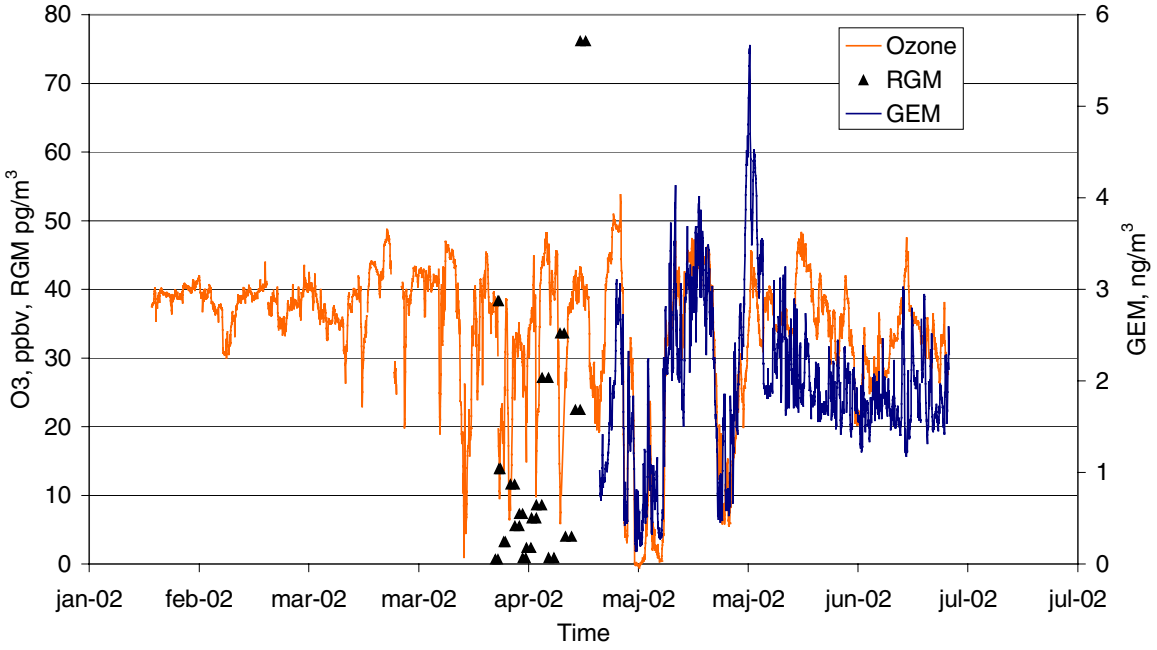

Figure 4.4 Ozone, GEM and RGM at Station Nord 2002.

Unfortunately there was a too large consumption of electricity that led to a slight decrease in the tension at the monitoring hut. As a consequence the TEKRAN 2537A did not work properly and many GEM data were discarded in the quality control after the campaign, so that there is only continuously GEM data from 27 June 2002 and onwards. The other instrumentation and meteorological equipment were unaffected.

Ozone and RGM

RGM and ozone measurements are seen in Figure 4.5 where the ozone values are proxy for GEM variation. The horizontal bars describe the sampling time of the RGM measurements, which varies between 4 and 24 hours. The maximum level is 76 pg/m^3 (24 hour average) which is significantly higher than observed in Denmark and Nuuk where the maximum level are at about 2-4 pg/m^3. Therefore RGM is produced in large quantities at station Nord compared with levels at mid latitudes, but at the same levels as observed at Barrow, Alaska, see section 4.3. Unfortunately, the long average sample time of RGM made it impossible directly to compare GEM and RGM values.

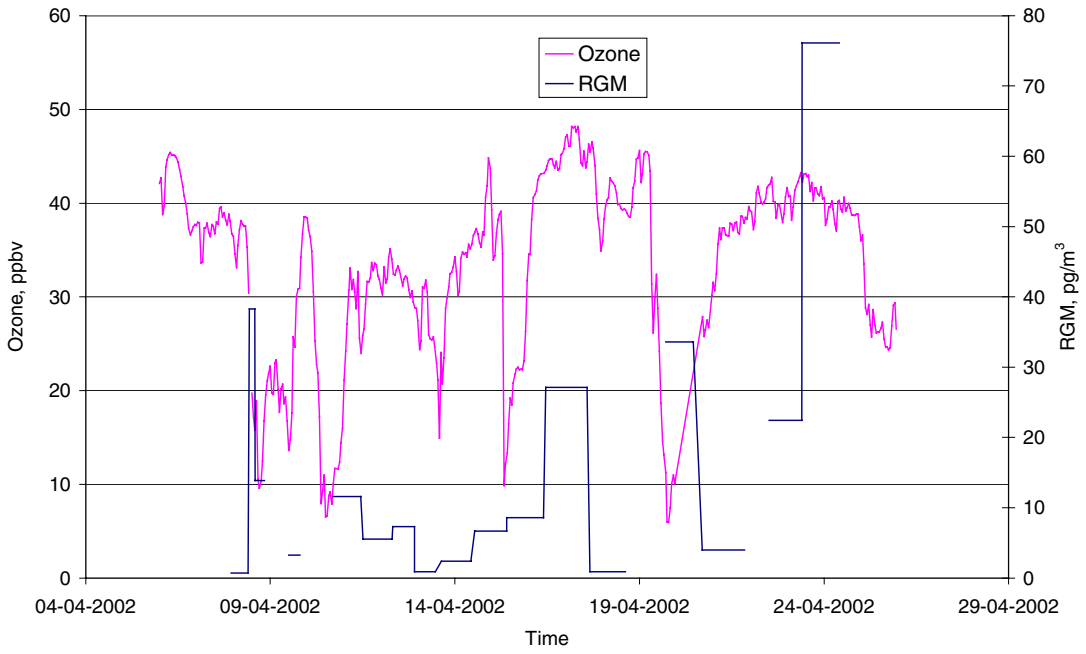

Figure 4.5 Ozone and RGM at Station Nord, Spring 2002. The horizontal bars of RGM show the duration of sampling.

4.3 Barrow campaigns

4.3.1 GEM and RGM continuous measurements

2003 campaign

GEM and RGM were measured during the campaigns in 2003 and 2004 using a TEKRAN 2537A equipped with a TEKRAN 1130 speciation equipment. The results for 2003 are shown in Figure 4.6.

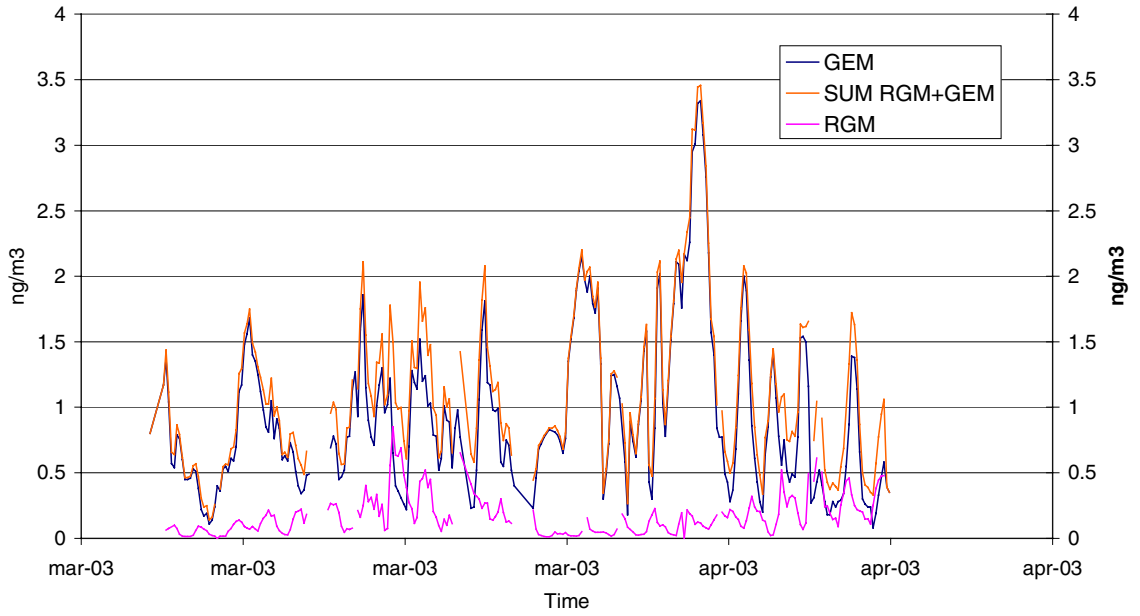

Figure 4.6 GEM, RGM and the sum GEM + RGM at Barrow, Alaska 2003

GEM and RGM diurnal pattern

The maximum RGM concentration is 850 pg/m^3 and the minimum is 0.5 pg/m^3. GEM varies between 0.08 and 3.34 ng/m^3. The variation shows a strong diurnal variation as further demonstrated in Figure 4.7 where all data are averaged over a specific time of the day.

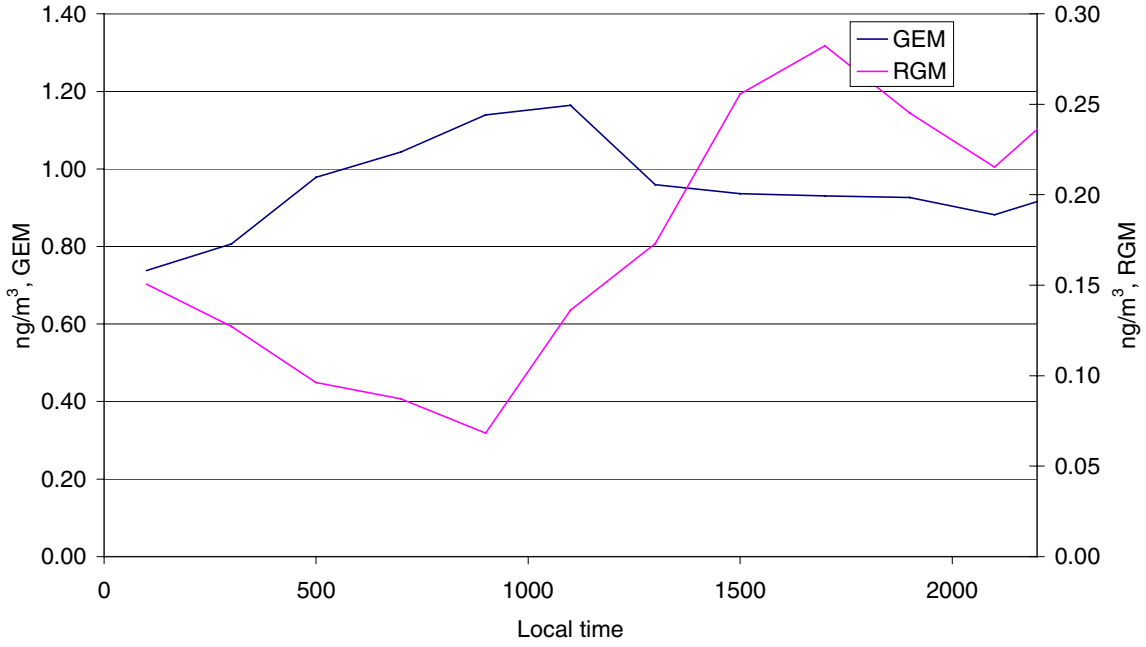

Figure 4.7 Average diurnal variation of GEM and RGM based on 2 hour average values at Barrow local time, 2003.

RGM concentrations peaked at 17.00 when correspondingly low GEM levels are measured. This pattern supports the hypothesis by (Lindberg *et al.*, 2002e) that RGM is formed photo-chemically. Brooks, (private communication) could fit the RGM concentration very well by introducing a function dependent on wind speed as a proxy for dry deposition and solar flux as a proxy for reactivity. However, the reactant responsible for the GEM removal and RGM formation is not yet identified even though there is strong evidence that it is related to Br atoms (Skov *et al.*, 2004g; Goodsite *et al.*, 2004b).

4.3.2 RGM, FPM and particle equilibrium

The longest depletion event ever was observed in 2004. It started March 25 and ended April 4, see Figure 4.8. During the depletion GEM and RGM concentrations were very low and FPM was high throughout the depletion period. Close to the end of the period the highest concentration ever of RGM was measured.

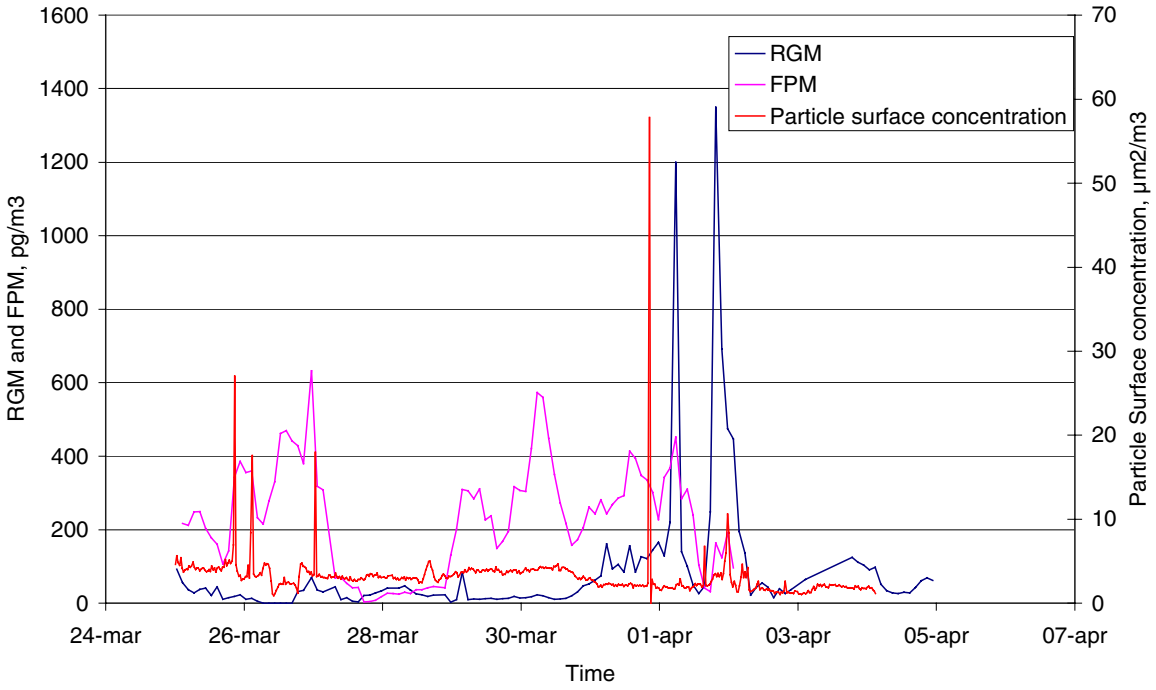

Figure 4.8 RGM, FPM and particle surface area concentration at Barrow 2004.

RGM, FPM and the size distribution of particles were measured in 2004 in order to describe the equilibrium between RGM, particles and FPM, see Figure 1.1.

$$\text{RGM} + \text{Particles} \rightleftharpoons \text{FPM} \tag{4.1}$$

The equilibrium constant K_{ads} for equation 4.1 is

$$K_{ads} = \frac{[FPM]}{[RGM][S_{particle}]} \tag{4.2},$$

where $S_{particle}$ is the particle surface area concentration

The temperature dependence of the equilibrium constant is expressed by Van Hoff's equation:

$$\ln K_{ads} = \ln\left(\frac{[FPM]}{[RGM][S_{particle}]}\right) = -\frac{\Delta H^0}{RT} + I \tag{4.3}$$

where T is the absolute temperature, ΔH^0 is the heat of adsorption, R is the ideal gas constant and I is a constant.

Correlation

Figure 4.9 shows a plot of $-\log_{10}([FPM]/([RGM][S_{particle}])$ versus 1/T. From equation 4.3 a straight line is expected. However it is clearly seen that there is not any correlation between the two parameters.

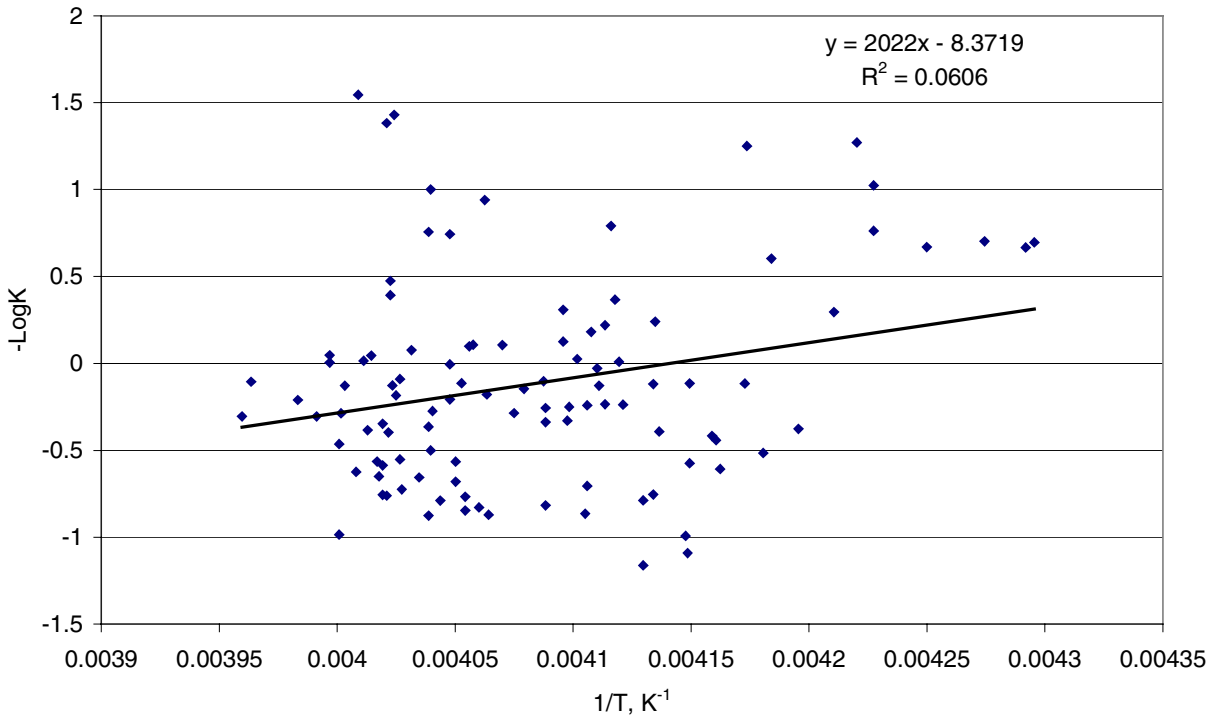

Figure 4.9 -$Log_{10}K$ (= -log ([FPM]/([RGM][$S_{particle}$]) and 1/T from Figure 4.8 together with regression line for the data –LogK and 1/T are poorly correlated.

However, during the depletion event a weak correlation is observed, see Figure 4.10 with correlation coefficient R^2 = 0.2881. Thus it is difficult to interpret the data. The parameter 1/T can only explain 29% of the variation in logK and the conclusion from the present data is that Van Hoff's equation is not fulfilled here.

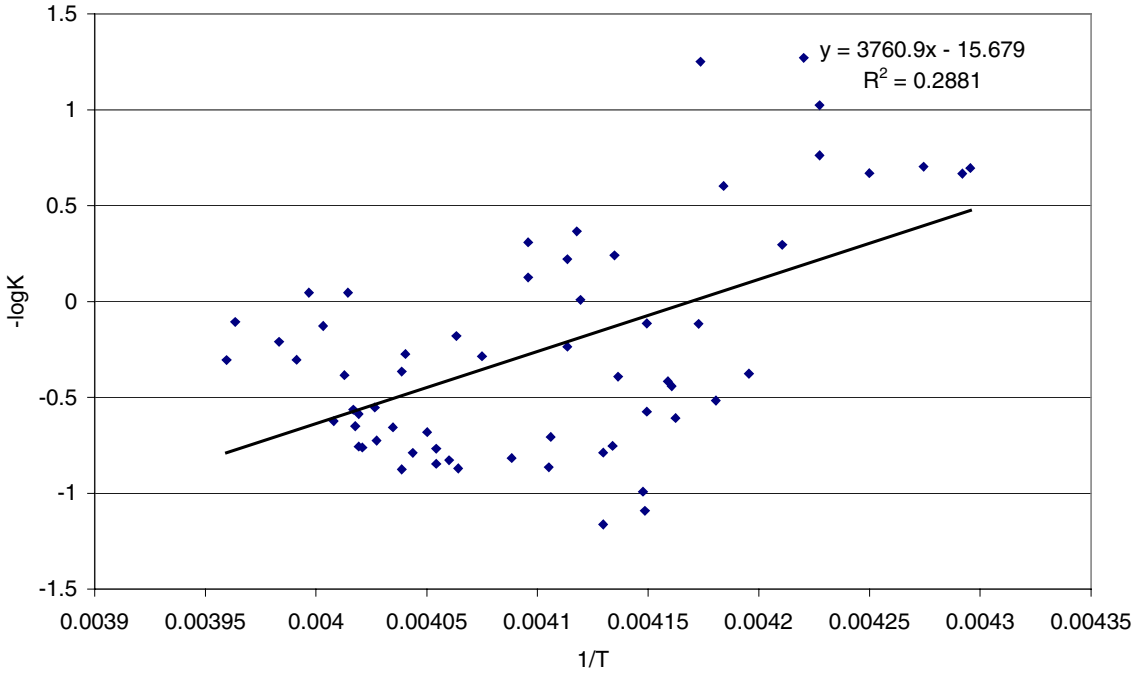

Figure 4.10 -$Log_{10}K$ (= -log ([FPM]/([RGM][$S_{particle}$]) and 1/T for the long AMDE period in Figure 4.7 together with regression line for the data –LogK and 1/T are poorly correlated, R^2 = 0.2881.

Thus equilibrium 4.2 is most probably of little importance for conversion and removal of RGM.

4.3.3 RGM and ozone flux

From 2001 to 2004 during a period of about one month each year, campaigns were carried out at Barrow, Alaska, where RGM fluxes were measured. In 2002 a similar campaign at Station Nord, Northeast Greenland was carried out as well. During all the years the work was carried out in co-operation with NOAA and ORNL. Furthermore, US-EPA participated the first year and NILU and MSC Canada were joining the campaign the last year and finally University of Grenoble participated at Station Nord in 2002. Students from both University of Copenhagen and University of Southern Denmark carried out additional supporting activities as well, e.g. see section 4.1.1 and 4.1.2.

Campaigns during March and April

All the campaigns were carried out over 1 month in the period from mid March to end of April where AMDEs are observed to be most frequent.

Barrow activities

In Barrow continuous measurements of GEM, RGM, FPM and Particle size distributions using commercial available instruments were performed. Furthermore, RGM and GEM fluxes (see section 4) were measured. RGM fluxes were measured by relaxed eddy accumulation, REA, see equation 3.1. GEM fluxes were measured by gradient method at about 20 m and 1 m above the ground in 2004 (Brooks *et al.*, 2005). Finally, gradient measurements of RGM were carried out in 2004 for supporting the REA results.

Uncertainties

The estimated uncertainties of the methods based on reproducibility experiments are listed in Table 3.1.

RGM fluxes and deposition velocity

In Figure 4.11 the results of the RGM flux measurements for 2001 to 2004 and the deposition velocities (V_d) calculated from equation 4.4 are shown.

$$V_d = F/C_{average} \tag{4.4}$$

where $C_{average}$ is the average of C_{upward} and $C_{downward}$.

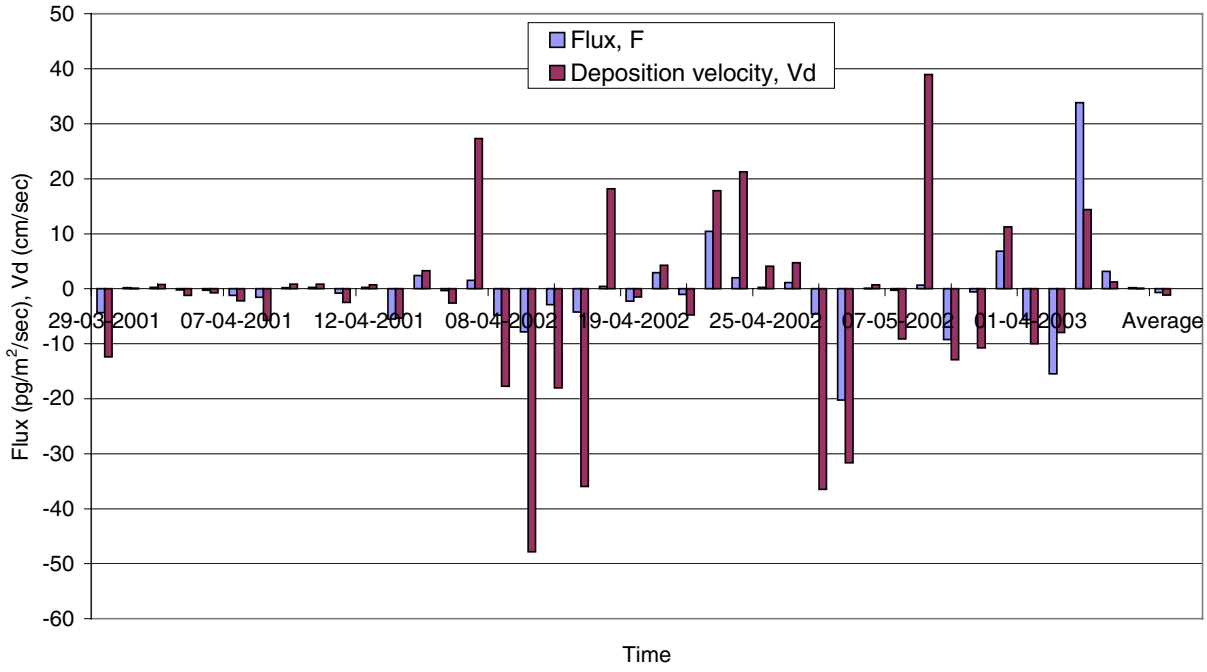

Figure 4.11 The measured flux at Point Barrow Alaska and the corresponding deposition velocities.

The flux measurements were carried out during AMDEs where large concentrations of RGM were build up. There is no flux data from St. Nord 2002 because of previously mentioned weather conditions and there is no RGM flux data from Barrow 2004 either. This time it was due to technical problems during the extreme weather conditions with temperatures varying from –20°C to -4°C. The record high temperatures gave the system a heat stroke. Figure 4.11 shows the flux measurements and the corresponding deposition velocity together with the average flux and the corresponding deposition velocity.

Reliability of measurements

There are periods with both emissions (positive values) and depositions (negative values). Before the campaigns, depositions were expected due to the observed short lifetime of RGM. So an obvious question is; whether the measured fluxes are real or due to artefacts and in case they are real what is the reason for and mechanism behind the emission.

Ozone gradient

Therefore ozone gradient measurements were carried out in both 2003 and 2004. Figure 4.12 shows the measured concentration gradients alternating between 10 min at 0.1 m and then 10 min at 20 m and so on.

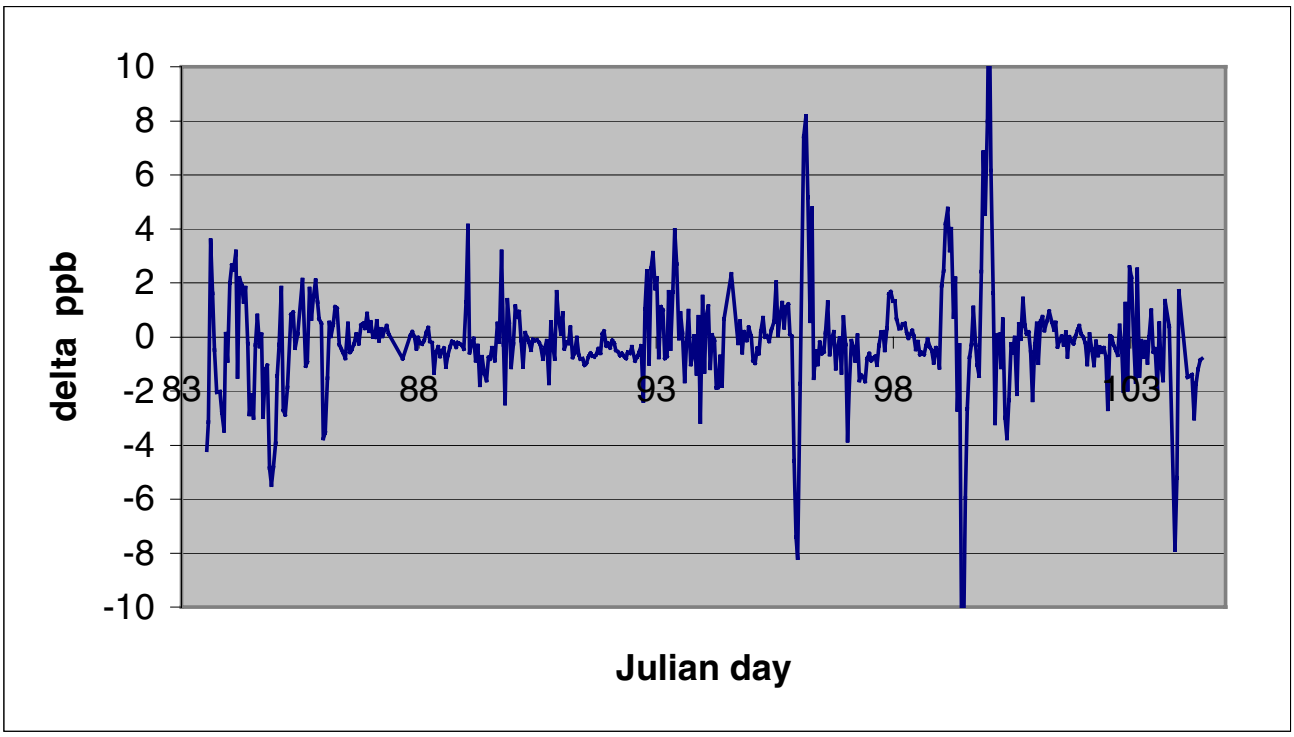

Figure 4.12 Ozone concentrations difference between 0.1 m and 20 m from Julian day 83 to 104 in 2003 .

In periods with AMDE there is a clear ozone gradient with the lowest height having the lowest value, whereas, there is no difference in periods without AMDE. Figure 4.13 shows the results for a period of 2.5 hours with depletion. A clear difference between the two heights is seen whereas there is no difference in Figure 4.14 for the period without AMDE.

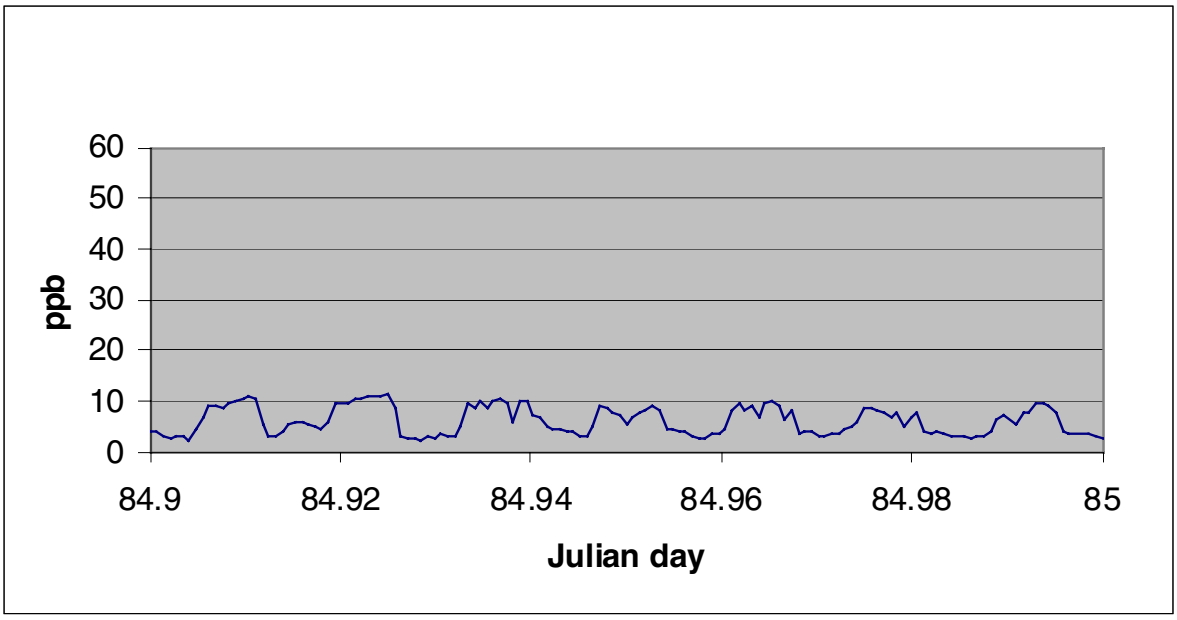

Figure 4.13 Ozone measurements alternating at 0.1 m for 10 minutes and at 20 m for 10 minutes and so on for a 2.5 h period on Julian day 84 with AMDE.

Chemical surface reaction

Ozone is known to have a very small deposition velocity to snow surfaces in agreement with Figure 4.13. Thus the only explanation for the gradient in Figure 4.11 is chemical removal near or at the surface.

Removal of GEM by Br

The connection between GEM and ozone depletion is very well established and there is strong evidence that the removal is due to competing reactions of ozone and GEM with Br (Skov *et al.*, 2004f; Goodsite *et al.*, 2004a) and leads to RGM (Lindberg *et al.*, 2002d).

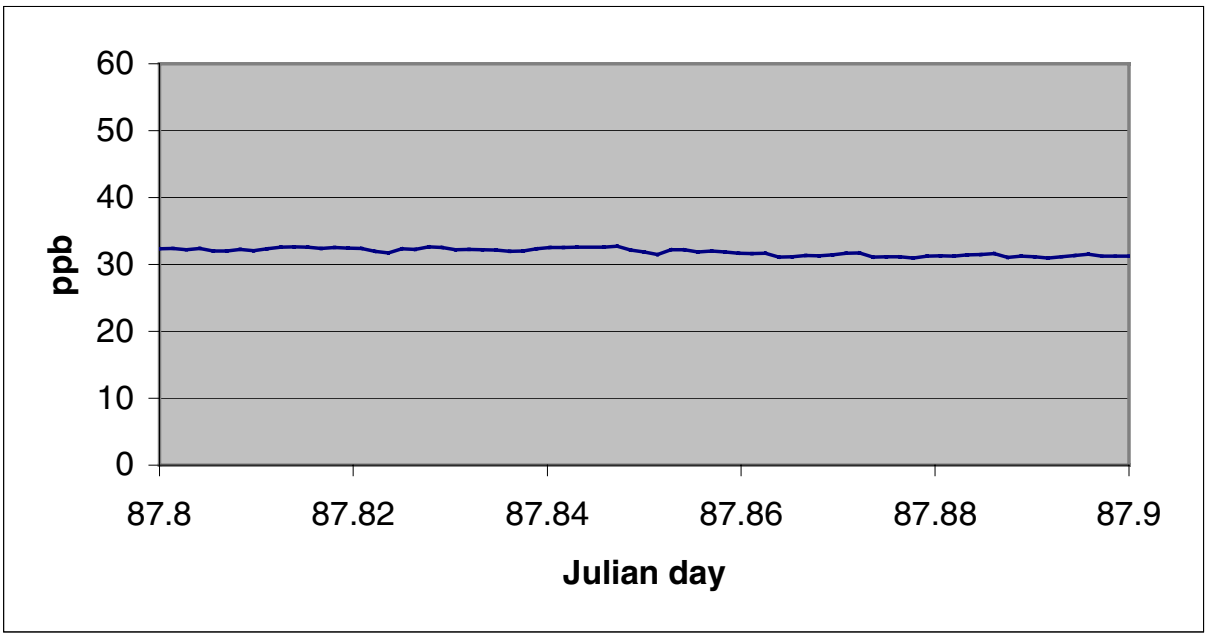

Figure 4.14 Ozone measurements alternating at 0.1 m for 10 minutes and at 20 m for 10 minutes and so on for a 2.5 h period on Julian day 87whithout AMDE.

Surface reaction of GEM

Therefore the fast removal reaction of GEM forming RGM is most probable at the snow surface or at least very close to the surface. This is important, as equation 3.1 then no longer is valid. Instead of C_{up} we have measured the sum of C_{up} and C_{chem}, where C_{chem} is the contribution from RGM due to a surface related reaction. The sum is denoted C_x

$$C_x = C_{up} + C_{chem} \quad (4.5)$$

and thus equation 3.1 has to be modified to equation 4.6

$$F = \beta\sigma_w(C_x - C_{chem} - C_{down}) \quad (4.6)$$

Unfortunately C_{chem} is unknown at present and we have only C_x. Therefore we can only express an upper limit for the flux (a lower limit of the deposition):

$$F = \beta\sigma_w(C_x - C_{down}) \quad (4.7)$$

In Figure 4.1 three positive fluxes (emissions) are seen in 2003. All three appear to be in periods with a negative ozone gradient, which confirms the interpretation that chemical formation of RGM close to the surface gives a significant contribution to C_x. The conclusion is that the flux value is biased due to the contribution of C_{chem} and thus C_x is measured instead of C_{up}.

Therefore it is only possible to give an upper limit for the deposition (of the negative fluxes) based on the average of a whole campaign period. The average fluxes are listed in Table 4.1 for the three years 2001, 2002 and 2003 together with average value of all the measurements.

Yearly average

Furthermore the average values are shown where positive values are removed.

Table 4.1 The average flux, concentration and deposition velocity of RGM at Barrow Alaska for the 3 campaigns in spring. The results in parenthesis are calculated from negative fluxes only.

Year	Average F		Caverage		Vd	
	pg/m2/sec		pg/m3		cm/sec	
2001	-0.67	(-1.42)	43.56	(34.07)	-1.54	(-4.16)
2002	-1.54	(-4.92)	29.74	(39.93)	-5.17	(-12.31)
2003	1.63	(-7.73)	145.93	(81.84)		(-9.45)
Average*	-0.67	(-4.45)	56.78	(46.24)	-1.17	(-9.63)

*Average is the average of all measured fluxes and concentrations and not only the three values listed here. V_d is calculated from the average flux and concentration.

Upper limit

Thus the obtained average F values have to be considered as upper limits. This is also the case for those where positive values have been removed from the average value. As a consequence the same is valid for the deposition velocity V_d also shown in Table 2.2.

Ignoring positive values

The average fluxes without positive fluxes are -1.42 pg/m^2/sec, -4.42 pg/m^2/sec and -7.73 pg/m^2/sec respectively for the campaigns in 2001, 2002 and 2003. The average flux for all campaigns is –4.10 pg/m^2/sec and the corresponding deposition velocity is calculated according to equation 4.3 to be –9.63 cm/sec (the negative sign indicates that it is a downward velocity).

Surface resistance

The deposition velocity is an important feature though it cannot be used directly in physical chemical transport models as e.g. DEHM (Christensen *et al.*, 2004b). Instead the resistance R is used, which is the inverse of V_d;

$$R = 1/V_d \quad (4.8)$$

The resistance is divided into the aerodynamic resistance R_a, the laminar resistance R_b and the surface resistance R_c. R_a is related to the meteorological conditions and R_b is related to the surface conditions

and meteorological conditions. The surface resistance is a parameter specific for the properties of the compound and the surface:

$$R = R_a + R_b + R_c \tag{4.9}$$

Assumptions

R_b is to a good approximation 0 for a snow surface and R_a can be calculated from the meteorological data available:

$$R_a = U/(U^*)^2 \tag{4.10}$$

where U is the wind speed and $-(U^*)^2$ is the vertical momentum (Asman et al. 1994). R_a is calculated to be 106 sec/m in 2003 and R is calculated from equation 4.9 to be 10.4 sec/m. It is clear from equation 4.10 that there is a discrepancy between the results for R and R_a because R_c cannot be negative. In fact the fast surface related reaction breaks down the necessary assumption for equation 4.9 that the flux is the same over the three resistances ($F = F(R_a) = F(R_b) = F(R_c)$).

Surface resistance

However, the very low values for R indicate that in any case *R* must be very small consequently the first estimate is close to zero. Thus *R* is only dependent on the aerodynamic resistance R_a.

RGM gradients

The gradient measurements at bottom 3.68, mid 8.19m and top 13.65m height are shown in Figure 4.15 and 4.16. They were carried out in 2004 to support the interpretation of REA measurements. Unfortunately the REA system broke down as previously mentioned.

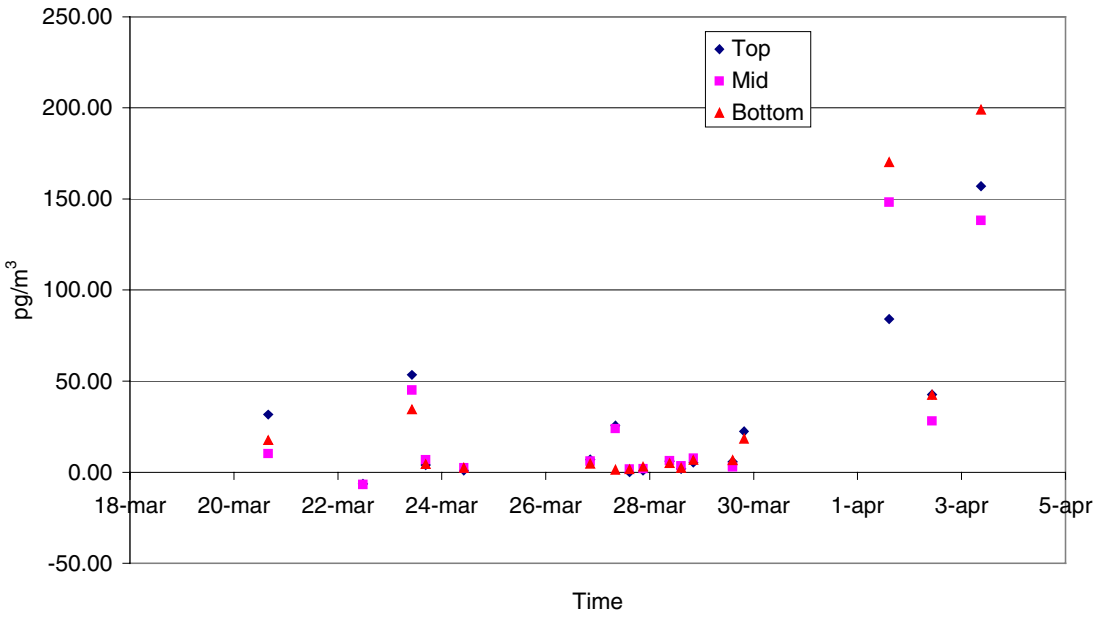

Figure 4.15 Measurements of RGM at bottom 3.68, mid 8.19m and top 13.65m heights.

Very low RGM concentrations are observed in the period from 20 to 30 April. At the end of the period there were strong GEM concentration fluctuations and high concentrations of RGM were observed, see Figure 4.8. The high RGM values are coincident with large positive gradients (emissions) that qualitatively agrees well with a chemical production of RGM close to the surface, see Figure 4.15

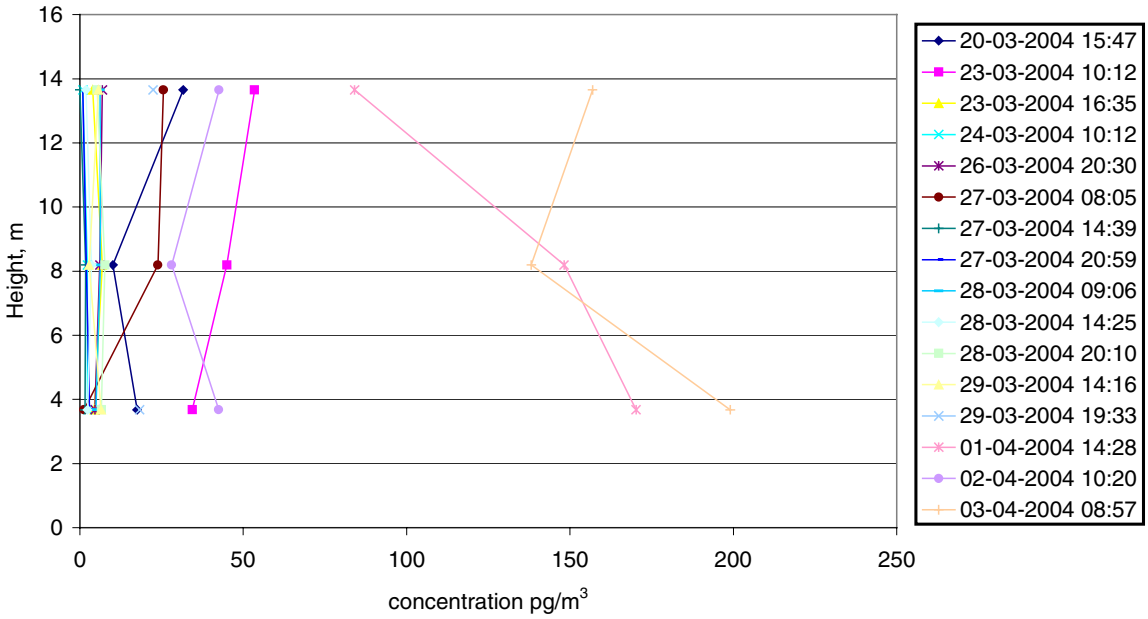

Figure 4.16 Height profiles of the measurements shown in Figure 4.

Previous description

The flux measurements show that RGM deposits quickly to snow surfaces at an inland station influenced by marine air. A simple process with GEM being oxidised into RGM followed by a fast deposition of RGM on the surface might not be correct.

Br chemistry

The critical processes responsible for mercury depletion most probably involve bromine in processes related to snow surfaces. Bromine reacts fast with ozone

$$Br + O_3 \rightarrow BrO + O_2 \quad (4.11)$$

BrO is fast recycled e.g.

$$BrO + BrO \rightarrow 2\ Br + O_2 \quad (4.12)$$

and thus Br has a catalytic cycle where ozone is removed (Mcconnell *et al.*, 1992). This catalytic circle proceeds in parallel with the competing reaction with Hg.

$$Hg + Br \rightarrow HgBr\bullet \quad (4.13)$$

followed by a series of reaction steps eventually leading to RGM.

Alternatively Br reacts with for example formaldehyde forming HBr that is removed fast from the gas phase e.g. by deposition

$$Br + H_2CO \rightarrow HBr + \bullet CHO \quad (4.14)$$

Reactivation of Br

HBr can be reactivated to Br_2 or BrCl on snow surfaces (Foster *et al.*, 2001) and Br is formed again by fast photolysis (Calvert & Lindberg, 2003).

$$Br_2 + h\nu \rightarrow 2\ Br \quad (4.15)$$

$$BrCl + h\nu \rightarrow Br + Cl \quad (4.16)$$

The reactivation of Br on surfaces might then be the limiting factor for deposition of atmospheric mercury, involving GEM conversion to RGM followed by deposition of RGM in agreement with the observations of RGM fluxes presented in this section. However, a very local process cannot be ruled out where RGM is produced at or close to open leads followed by advection to the CMDL site.

4.4 Model calculations of the mercury load to the Arctic using DEHM

Already after the first campaign in 2001 it was clear that the deposition of RGM was fast and. In literature it is suggested that the process is connected to sea ice (Lindberg *et al.*, 2002c). Furthermore analysis of GEM and ozone indicates that Br was the responsible reactant for converting GEM to RGM (Skov *et al.*, 2004e). This new knowledge has been used to make a new parameterisation describing the fast processes of AMDE. This new parameterisation was used in DEHM (Christensen *et al.*, 2004a; Skov *et al.*, 2004d) and the first calculations ever were carried out of the atmospheric burden of mercury to the Arctic environment with a model including the fast removal of atmospheric mercury during AMDE. The results are presented in this section.

Model construction

The model system consists of two parts: a meteorological part based on the PSU/NCAR Mesoscale Model version 5 (MM5) (Grell *et al.*, 1995) and an air pollution model part, the DEHM model. The model system is driven by global meteorological data obtained from the European Centre for Medium-range Weather Forecasts (ECMWF) on a $2.5^{\circ} \times 2.5^{\circ}$ grid with a time resolution of 12 hours.

DEHM

The DEHM model is based on a set of coupled full three-dimensional advection-diffusion equations, one equation for each species. The horizontal mother domain of the model is defined on a regular 96x96 grid that covers most of the Northern Hemisphere with a grid resolution of 150 km × 150 km at 60°N. The vertical resolution is defined on an irregular grid with 20 layers up to about 15 km reflecting the structure of the atmosphere.

Chemical module

The chemistry scheme of mercury in the atmosphere outside the Arctic region includes 13 mercury species: 3 in gas-phase (Hg^0, HgO and $HgCl_2$), 9 species in the aqueous-phase and 1 in particulate phase and is adopted from the literature (Petersen *et al.*, 1998). Within the Arctic region an additional 1. order reaction of GEM was added to the chemical scheme where GEM reacts to form RGM with properties

determined experimentally in the Arctic, see description below. The reaction takes place inside the boundary layer over sea ice during sunny conditions in order to reproduce AMDEs in accordance with obtained experimental results (Lindberg *et al.*, 2002b; Skov *et al.*, 2004c). The reaction rate constant for the 1. order removal is based on the observed removal rates of mercury at Station Nord, (Skov *et al.*, 2004b). The measurements gave a 1. order lifetime of GEM between 3 and 10 hours, so scenario calculations with this range of lifetime were performed. The fast first order oxidation stopped, when surface temperature exceeded -4°C. This temperature appears to be crucial for the presence of AMDE as Br_2 and BrCl are formed at the surfaces of refreezing leads (Lindberg *et al.*, 2002a). Except for the AMDEs, the fate of GEM is controlled by a slow chemical removal in the gas phase and uptake by cloud water. The present version of the chemical scheme is the first step towards a chemical scheme, which simulates the chemical processes during AMDEs.

The dry deposition velocities of the RGM species formed during AMDEs are based on the resistance method, where the surface resistance is 0 s m^{-1} in accordance with the experimental results. The wet deposition of reactive and particulate mercury is parameterised by using a simple scavenging coefficients formulation with different in-cloud and below-cloud scavenging coefficients (Christensen, 1997).

The emissions of anthropogenic mercury are based on the global inventory of mercury emissions for 1995 on a 1°x1° grid (Pacyna & Pacyna, 2002b) including emissions of GEM, RGM and total particulate mercury (TPM). The model does not contain any re-emissions from land and oceans. Instead, a background concentration of 1.5 ng/m^3 of Hg^0 is used as initial concentrations and boundary conditions. The mercury model has been run for the period October 1998 to October 2002 without AMDEs and with AMDEs.

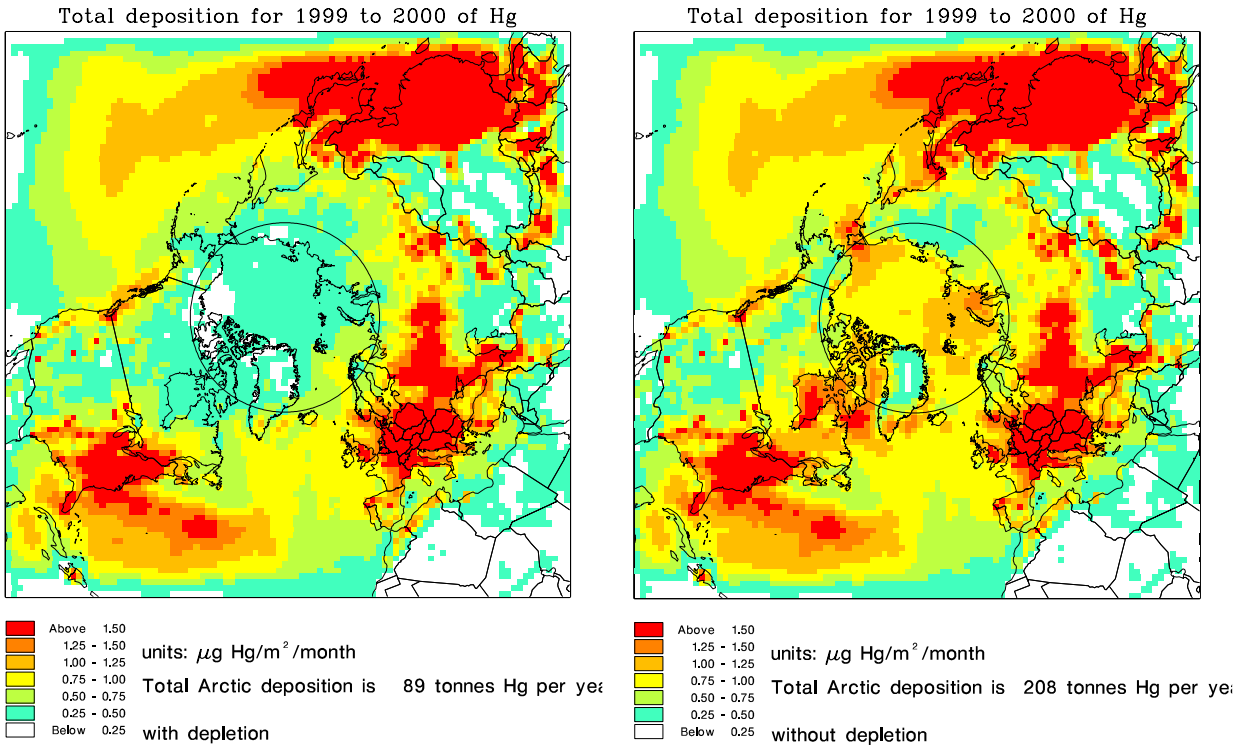

Figure 4.16 The total annual average deposition of mercury without Arctic mercury depletion (left) and with (right) in µg Hg/m^2/year for the years 1999 and 2000. The total annual deposition north of the polar circle is indicated as well

The total amount of mercury deposited in the Arctic area north of the polar circle is thus calculated to be 200 tons per year including AMDEs. This is more than the double of what is calculated to be deposited when AMDEs are not included in the model (89 tonnes/year), see Figure 4.16.

In connection with this value, re-emission of elemental mercury is not taken into account. GEM gradients were measured in 2003 and 2004 ((Brooks *et al.*, 2005). The first analysis of the re-emission indicates that about 30% of the deposited mercury is re-emitted, see Figure 4.17.

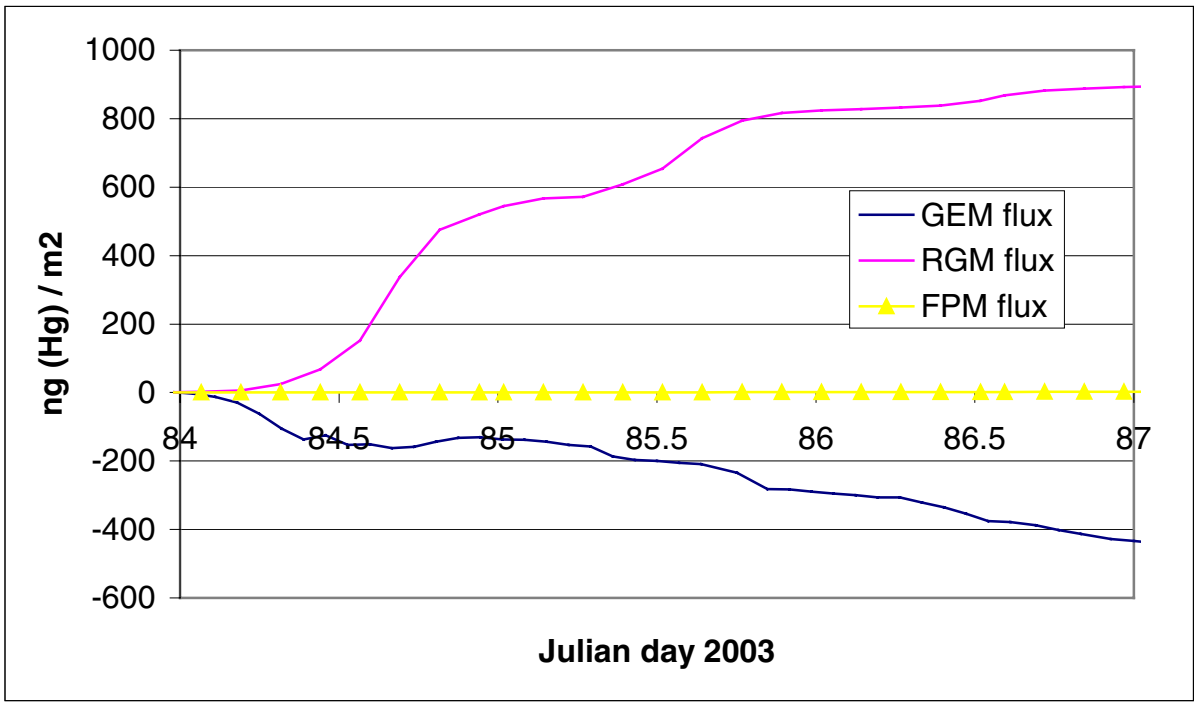

Figure 4.17 The fluxes of RGM, GEM, and FPM for Julian day 84 to 86 (April 1-3). Positive fluxes are depositions and negative fluxes are emissions. GEM flux is by gradient method, whereas RGM and FPM are estimated from the determined Vd for RGM and for particles together with inversion height and atmospheric turbulence (Brooks *et al.*, 2005).

It is difficult to extrapolate the result in Figure 4.17. They represent only 3 days at one arctic location. We do not know if the re-emission has a seasonal variation. At Alert, Canada they have also measured GEM, RGM and FPM and they found that FPM was much higher than RGM which means a completely different dynamics and thus a geographical extrapolation is connected with large uncertainty.

DEHM assumes that RGM formation is occurring equally within the marine boundary layer and it is known from the campaign that this cannot be the case. The surface related processes observed here also extend to inland locations exposed to air of marine origin. In fact elevated mercury concentrations in snow was recently found to extend 40 km inland from the seashore (Douglas & Sturm, 2004).

The mechanisms behind this observation are not known. It might be explained by deposition of bromine species followed by reactivation of bromine at the surface or it might be explained by horizontal transport of mercury by blowing snow or simple advection of marine air masses without vertical mixing.

5 Conclusion

Pyrolysis system

New methods have been developed and new and "old" methods have been tested. A pyrolysis system was tested and found to give reliable results of TAM. This is very important for further investigations because the TAM system can be used to obtain flux values for the net mercury flux with high time resolution. Furthermore the system can be run continuously and therefore much more data can be obtained compared to manual measurements. In this way it will be possible to obtain a mass balance for the inter-compartment fluxes from the atmosphere to the surface.

Diffusive sampler

For the first time a diffusive sampler has been developed which can be used for measurements of GEM during a time resolution of at least one week at the low ambient air concentrations.

RGM and ozone flux

The first measurements ever of RGM fluxes have been carried out. The RGM flux measurements in the Barrow campaigns combined with model calculations using DEHM have demonstrated that AMDEs more than doubles the input of mercury to the Arctic environment through the transformation of GEM to RGM followed by deposition of RGM to the snow. About 200 tons/year of mercury are deposited to the Arctic north of the polar circle.

The processes controlling atmospheric mercury are very complex and many questions are still unanswered. RGM is most probably formed close to the surface and not uniformly in the boundary layer as previously believed. A significant part of deposited mercury is reduced and re-emitted as GEM. This re-emission was determined for the first time. Finally the geographical extension of AMDEs is not well described either. The bottom line is that the processes and dynamics of AMDEs are still not sufficiently known and thus the first estimate of atmospheric deposition to the Arctic is encumbered with a very large uncertainty. This is illustrated by a recent paper that has estimated the input to the Arctic to be 325 tons/year (Ariya *et al.,* 2004) more than 50% higher than the one obtained here.

6 Recommendations

Based on the work performed so far we have made the first estimate of the Hg burden to the Arctic environment from the atmosphere. However, the dynamics between the various atmospheric mercury species are not yet understood and deposition and reemission of the various mercury species are not fully understood. There is a strong need to understand processes controlling these processes. In particular it is important to understand the role of snow air interaction determining for example a possible reactivation of bromine.

The central question is what is the mass balance for atmospheric mercury on an annual basis. Future monitoring should apply the new more cost efficient methods developed in FOMA and be carried out on a more long term basis so seasonal behaviour can be determined as well as long time trends.

Mercury in snow

Mercury in snow has never been determined in Greenland and the seasonal as well as the geographical distribution are strongly needed.

Input to marine system from melting snow and ice

Finally, the input of mercury to the marine system from melting snow and ice have to be determined, as no such data have yet been obtained.

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The main source of mercury in the Arctic is long range transport from mid latitudes. In order to understand the dynamics of the source strength in the Arctic a series of analytical methods is developed. For example the first flux measurements ever of RGM have been carried out together with flux measurements of GEM. The results are used to make a new parameterisation of the chemical and physical processes and model calculations are performed for the first time of the input of atmospheric mercury to the Arctic.

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