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Atmospheric mercury concentrations: measurements and profiles near snow and ice surfaces in the Canadian Arctic during Alert 2000

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Abstract

Gaseous elemental mercury (GEM) concentration measurements were made during the Alert 2000 campaign in Alert, Nunavut, Canada, between February and May 2000. GEM exhibits dramatic mercury depletion events (MDE) concurrently with ozone in the troposphere during the Arctic springtime. Using a cold regions pyrolysis unit, it was confirmed that GEM is converted to more reactive mercury species during the MDEs. It was determined that on average 48% of this converted GEM was recovered through pyrolysis suggesting that the remaining converted GEM is deposited on the snow surfaces. Samples collected during this campaign showed an approximate 20 fold increase in mercury concentrations in the snow from the dark to light periods. Vertical gradient air profiling experiments were conducted. In the non-depletion periods GEM was found to be invariant in the air column between surface and 1–2 m heights. During a depletion period, GEM was found to be invariant in the air column except at the surface where a noticeable increase in the GEM concentration was observed. Concurrent ozone concentration profiles showed a small gradient in the air column but a sharp decrease in ozone concentration at the surface. Other profile studies showed a 41% average GEM concentration difference between the interstitial air in the snow pack and ~2 m above the surface suggesting that GEM is emitted from the snow pack. Further profile studies showed that during MDEs surface level GEM exhibits spikes of mercury concentrations that were over double the ambient GEM concentrations. It is thought that the solar radiation may reduce reactive mercury that is deposited on the snow surface during a MDE back to its elemental form which is then increasingly released from the snow pack as the temperature increases during the day. This is observed when wind speeds are very low. Crown Copyright © 2002 Published by Elsevier Science Ltd. All rights reserved.

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1. Introduction

Mercury is a persistent, toxic and bio-accumulative heavy metal that is of great interest in the Arctic because of the invasive impact it has on biota and northern peoples. In the atmosphere, mercury exists predominantly in the gaseous elemental phase (>95%) and may reside in the atmospheric environment for as long as 24 months (Schroeder and Munthe, 1998). In 1995, in the Canadian high Arctic at Alert, it was discovered that tropospheric mercury exhibits dramatic depletion episodes concurrent with the now well-known tropospheric ozone depletion events during and after the polar sunrise (Schroeder et al., 1998; Barrie and Platt, 1997). Recent studies have provided evidence that the mercury is converted from its elemental vapour (Hg(0)) form (gaseous elementary mercury, GEM) to a more reactive

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mercury species (Hg(II)) (reactive gaseous mercury, RGM) (Lindberg et al., 2001; Lu et al., 2001). This transformation may provide an important pathway for introducing this reactive mercury species into the biosphere thus impacting large areas of the Northern Hemisphere at a time when biota are preparing for peak summertime activity. The mechanism of the conversion of GEM into RGM is not well understood nor are the roles of the snow pack and the host of chemicals existing therein. For this reason various conversion and air–snow interaction studies were undertaken during the ALERT 2000 campaign conducted in Alert, Nunavut between February and May 2000.

2. Experimental

2.1. Sampling

All experiments were conducted near Alert, Nunavut, Canada which is located on the northern tip of Ellesmere Island. Sampling took place at three locations: the Global Atmospheric Watch (GAW) laboratory, the Special Studies Trailer (SST) and the Ice Camp. The GAW lab and SST are both situated 6 km south of the shoreline at 175 m ASL while the Ice Camp was 1.4 km north of the nearest land on the frozen Arctic Ocean. There were two intensive measurement campaigns: the "dark campaign" (in the absence of daylight) was from 7–21 February 2000 and the "light campaign" (in the presence of 24 h daylight) was between 17 April and 8 May 2000. During the light and dark campaigns, the air was sampled at the GAW lab and the SST. Additionally, during the light campaign, air sampling was conducted at the Ice Camp. In general, experiments were conducted either at the SST or at the Ice Camp but rarely at the same time due to the significant distance between sites.

2.2. Analysis methods

All measurements of mercury compounds were made using a Tekran™ 2537A Mercury Vapour Analyser (Tekran Inc., Toronto). The instrument employs selective concentrations of gaseous phase mercury by amalgamation onto gold cartridges with subsequent thermal desorption and cold vapour atomic fluorescence spectrophotometric detection. The detector is specific for measuring the elemental form of mercury (GEM). The flow rate of sampled air was set at $1.5\,\mathrm{l\,min^{-1}}$ and concentrated for either 5 or 30 min. A Teflon filter (47 mm diameter, 0.2 μm pore) was installed at the front of the sample lines as well an additional filter is always located at the back of the Tekran instrument at the sample inlet.

The concentration of "total airborne mercury" (TAM) species present in ambient air was determined by installing a Cold Regions Pyrolysis Unit (Tekran Inc., Toronto) as a front-end unit to a Tekran 2537A monitor. This unit was located at the GAW laboratory at a height of 3 m (on a walk-up tower). Using the pump of the 2537A analyser, ambient air first enters the inlet of the CRPU and is drawn through a quartz tube, filled with quartz chips, that is enclosed in a heated oven maintained at 900°C. By heating the incoming air to this temperature gas phase and particle-associated organic and inorganic Hg compounds (denoted PAM) are converted to GEM for detection. The conversion efficiency of the system was >99% (determined by the manufacturer). The system was operated either with a Teflon filter installed at the inlet to prevent the particulate species from entering the system, or it was left unfiltered to measure all mercury species in the ambient air. The sampled air was pulled through a common inlet and then split into ambient (unheated) and pyrolysed air streams where, two different heated (20°C) Teflon lines (10.7 m in length) introduced the samples into two separate 2537A GEM monitors. The GEM concentrations in air from the two instruments were directly compared.

Snow samples were collected into ultra-cleaned, wide mouth, glass jars with solid PFA Teflon lids. Snow samples were kept frozen until they were weighed and low-mercury HCl and BrCl were added to preserve the Hg in the samples and as a digestant for total mercury (Szakacs et al., 1980; Bloom and Crecelius, 1983). Total mercury was determined by cold vapour atomic fluorescence spectrometry after stannous chloride reduction of the melted snow samples, as described elsewhere (Vermeir et al., 1991).

2.3. Vertical air concentration profile measurements

Vertical profile data for GEM were collected at the SST and Ice Camp. At the SST, sample intake lines were positioned at 1, 39, 98 and 190 cm above the snow surface, while at the Ice Camp the intake levels were at 1, 10, 50 and 120 cm. The sample lines (unheated) were Teflon (0.635 cm o.d.) and ~15 m in length. At each site a synchronised switching device was installed which allowed alternate sampling at two levels. By manual manipulation, the sampling procedure could be expanded to 4-level sampling. Each level was sampled sequentially for various times between 10 and 30 min. At the Ice Camp, ozone was measured concurrently next to the GEM intake levels of 1, 10 and 50 cm, as well as from a 10 m tower at 4.5 and 7 m (for details see Fuentes et al., 2002). The intent of these profile studies was to determine vertical gradients above the snow surface. However, due to a combination of light snow fall and snow drifts during the month of March, when the instrument at the SST ran in unattended automatic mode, a unique series of 2-level profiles was obtained when the bottom inlet became buried under a layer of snow (~10 cm from the surface). At this time, the upper inlet was set at ~180 cm above the snow surface. After 11 April, the bottom inlet at the SST was dug out so that it was positioned at 1 cm above snow surface, the top level remained at 180 cm above the snow surface. All times reported in this paper are in Greenwich Mean Time (GMT).

3. Results and discussion

3.1. Determination of fractional composition of Hg in ambient air

The cold regions pyrolysis unit (CRPU) was installed in Alert to determine the concentrations of total airborne mercury in the environment before, during and after polar sunrise. Pyrolysis is known to convert any inorganic and organo-metallic airborne mercury species that are not GEM (e.g. RGM or PAM) into GEM (Schroeder and Jackson, 1984). A mercury depletion event (MDE) is operationally defined in this paper as GEM concentration $<1.0\,\mathrm{ng\,m^{-3}}$. Two types of experimental set-ups were employed for the CRPU experiments: (1) when the incoming air to the CRPU was unfiltered and (2) when the incoming air to the CRPU was filtered. Results from the first experimental set-up showed that when there was no MDE occurring, the pyrolysed air and ambient air (non-pyrolysed) samples generally yielded the same GEM concentration. This confirmed that most of the total airborne mercury at that time was GEM. However, using the same set-up during MDEs showed that GEM in the two streams differed substantially. GEM concentrations, measured from the ambient air sample inlet, typically decreased to well below $1\,\mathrm{ng\,m^{-3}}$, but the pyrolysed air mercury concentrations remained higher. These results indicated that during depletion events mercury species other than GEM were present in the air, presumably RGM and PAM. Importantly, it was observed that not all the missing GEM during the depletion event was recovered through pyrolysis. Using the second CRPU experimental set-up, when the CRPU incoming air was filtered, it was observed that the GEM concentrations measured in both the pyrolysed and ambient air streams were identical whether it was during a depletion event or not. Hence it was deduced that the inlet filter was retaining PAM and/or RGM species.

Fig. 1 shows the unfiltered ambient mercury concentration data from 7 March to 19 April and the per cent recovery of missing GEM determined by the CRPU during that time where eight significant mercury depletion episodes occurred. The per cent of mercury recovered from the depleted ambient air through pyrolysis was estimated according to the following equation (where $1.75\,\mathrm{ng\,m^{-3}}$ is the mean winter-time concentration of GEM in the year 2000):

$$\frac{[\mathrm{GEM}]_{\text{pyrolysed air}} - [\mathrm{GEM}]_{\text{ambient air}}}{1.75 - [\mathrm{GEM}]_{\text{ambient air}}} \times 100\% = \text{Percent Recovered.}$$

This percentage was calculated for the amount of mercury recovered by pyrolysis during MDEs (ambient $\mathrm{GEM} < 1.0\,\mathrm{ng\,m^{-3}}$). Using the data from 3 March to 19 April 2000 it was found that by pyrolysing unfiltered air, on average 48.5% (ranging from 11% to 87%) of the oxidised elemental mercury was recovered during the various depletion periods. This shows that a significant amount of the total airborne mercury was "unaccounted

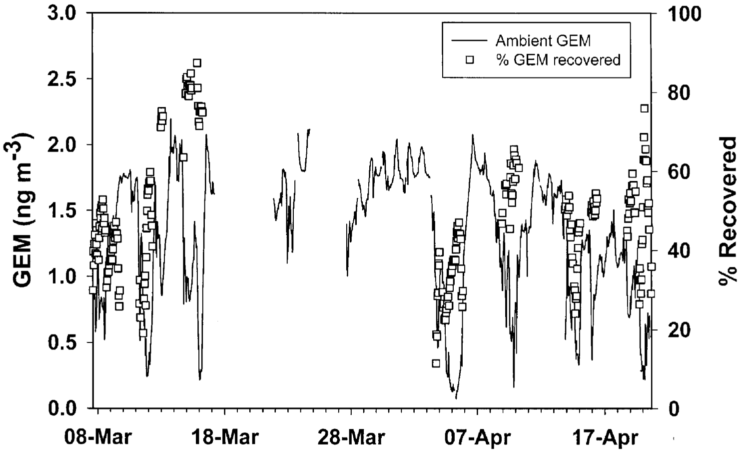

Fig. 1. Ambient GEM concentrations and per cent recovered of missing GEM during MDEs determined using the Cold Regions Pyrolysis Unit between 7 March and 19 April 2000 at the GAW lab.

for" and we postulate that this had been deposited to the snow and ice surfaces. In support of this hypothesis, we collected freshly fallen snow samples near the SST and near the Ice Camp. Near the SST, in the dark period the mean mercury concentration in freshly fallen snow was $5\,\mathrm{pg\,ml^{-1}}$ while in the light period, when there were frequent MDEs, the mean concentration was $121\,\mathrm{pg\,ml^{-1}}$. At the Ice Camp samples contained on average 21.7 and $182\,\mathrm{pg\,ml^{-1}}$ of mercury for the dark and light periods, respectively (Lawson, 2001). These results compare very well with similar studies at Alert in 1999 where average mercury concentrations increased from $4.9\,\mathrm{pg\,ml^{-1}}$ in the dark period to $185\,\mathrm{pg\,ml^{-1}}$ in the light period (Schroeder et al., 2000). As well, surface snow collected in 1998 over the frozen Beaufort Sea showed enhanced mercury concentration levels in the sunlit spring when compared to the dark winter period (Lu et al., 2001). While tempting, it is not justified to attempt a quantitative comparison between the unaccounted for mercury, as calculated above, and mercury concentrations in the snow. Firstly, the unaccounted for mercury consists of unknown fractions of both RGM and PAM, which are expected to have different scavenging efficiencies/deposition rates. Secondly, the actual scavenging is most likely to occur before the sampled air mass reaches the measurement site. Nevertheless, these data show that the snow pack at Alert and other Arctic locations is enriched in some form of mercury.

3.2. *Vertical gradient air profiling experiments*

The possible role of the Arctic snow pack in the depletion of tropospheric mercury and whether it acts as a source or sink was addressed by measuring surface-level air concentration profiles of GEM. Profiles were measured at the SST during both the dark and light intensive periods and at the Ice Camp during the light intensive period. Typical results from 4-level vertical concentration profiles of GEM are shown in Fig. 2. These gradients were measured for 3–5 h. Fig. 2A and B shows two cases of GEM concentrations in the air column when no appreciable depletion events occurred: (A) in February, in the absence of sunlight and (B) in April under 24-hour sunlight. These profile data showed no significant difference in GEM concentrations between near surface and those at 1.9 and 1.2 m heights at the SST and Ice Camp, respectively. In contrast, Fig. 2C shows profile data obtained at the Ice Camp on 3 and 8 May, while a depletion event occurred. For the most part, it can be seen that the GEM concentration in the air column above 10 cm remained invariant with height but close to the surface a noticeable increase in the GEM concentration was observed. Concurrent ozone profiles data are also shown in Fig. 2C. These profiles exhibited small but measurable mixing ratio gradients between the 10 and 120 cm height and a sharp decrease at the surface level. These data suggest that the snow pack was a source of GEM but a sink for ozone. Although mercury vapour and ozone have very different physical and chemical properties (Schroeder, 1982) they are known to correlate extremely well in the Arctic boundary layer in winter and spring when sampled at a height of 3 m ($R^2 > 0.8$) (Schroeder et al., 1998). Clearly this correlation does not pertain to the air close to the surface during a MDE. GEM behaves in a manner that is different from ozone as the two substances approach and interact with the frozen surface. The nature of this interaction is complex and is not yet fully understood at this time.

In the period between the two intensive campaigns, 2-level GEM air concentration profiles continued at the SST. As mentioned earlier, due to occasional snowfall and snowdrift, the bottom inlet became buried in snow about 10 cm deep. Hence, between 10 March and 10 April 2000, the interstitial air in the snow pack was alternately measured with the air 180 cm above the snow surface. Results from these measurements showed that the concentration of GEM in the interstitial air in the snow pack was almost continually higher than in ambient air, especially when a MDE was experienced (but not limited to those periods). This observation can be contrasted with measurements of ozone in the snow interstitial air. On a number of occasions, the concentration of ozone at various snow depths in the snow pack was measured (Albert, 2002). The authors observed a decrease in ozone with depth into the snow pack indicating that the snow was a sink of ozone. The GEM concentration difference, during this period between campaigns, between the two inlet heights was found to range between -0.32 and $3.25\,\mathrm{ng\,m^{-3}}$ and the average per cent difference was 41%. This shows that GEM was almost continually released from the snow pack indicating that the snow was a source of mercury. Fig. 3 exemplifies this positive gradient by showing GEM concentrations at 180 cm above the snow surface and the difference of GEM concentrations between the interstitial snow pack air and the air at 180 cm from 12 to 20 March (differences are calculated as: GEM in snow−GEM in air). Snow pack temperature at a depth of –6 cm near the SST and wind speed and total incoming solar radiation data from the weather station at Alert are also shown in Fig. 3. Upon visual inspection, it appears that there is a distinct diurnal signature in the GEM concentration at the 180 cm inlet that somewhat correlates with solar radiation. At times, the snow temperature and GEM concentration differences follow similar temporal patterns suggesting mercury is being released from the snow pack when the snow temperature increases. Also notable is that the largest difference between GEM in snow interstitial air and at 180 cm above the snow pack occurs when the wind speed is low. Although we caution that the wind

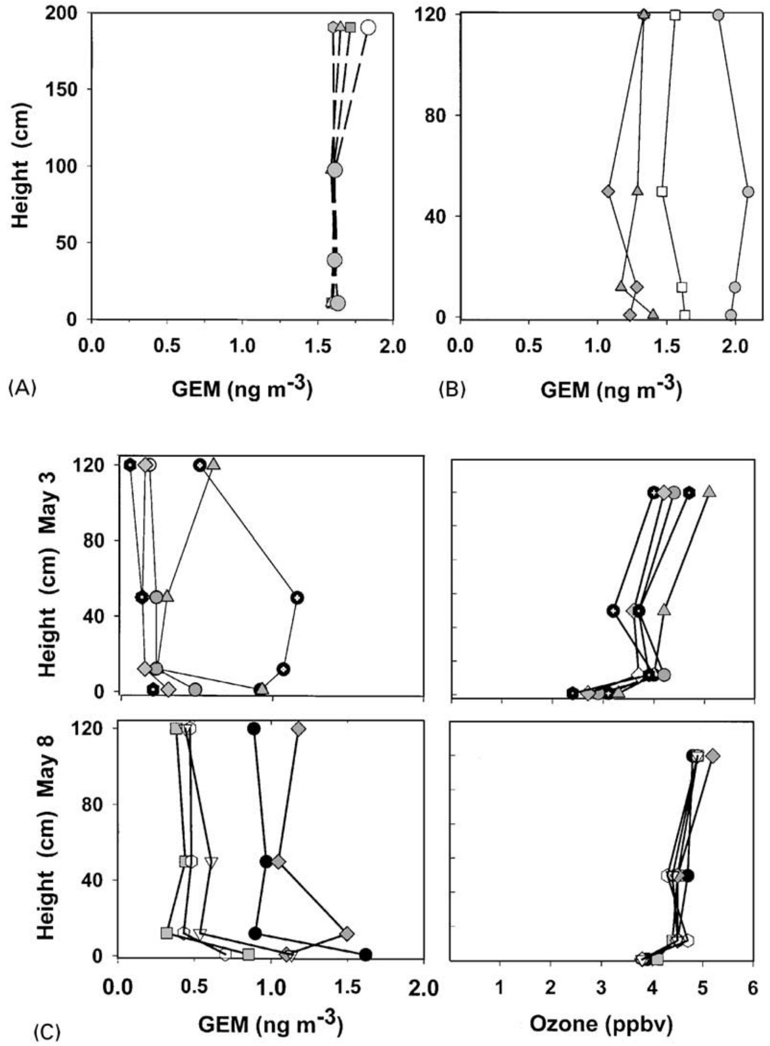

Fig. 2. Sequential 4-Level GEM concentration profiles during non-depletion events at the SST on 17 February (Panel A); at the Ice Camp on 22 and 23 April 2000 (Panel B) and GEM and ozone concentration profiles at the Ice Camp during MDEs on 3 May 2000 (Panel 1) and 8 May 2000 (Panel 2).

data shown here were not collected at the same location as the GEM measurements, it is notable that a similar trend is apparent in the vertical gradient data discussed in the next section. If the snow pack is a source of mercury then it is realistic to observe enhanced concentration profiles during calm conditions.

Another pattern to be discerned is the large temporary increase in the GEM observed in the snow pack during the recovery at the back end of a MDE. An example of this observation is shown in Fig. 4 where the GEM concentration at 180 cm is plotted as well as the GEM concentration difference between GEM in snow pack and at 180 cm. Meteorological information is also included as before. Between 4 and 5 April, the GEM in the ambient air (at 180 cm height) is almost completely depleted yet the GEM in the snow pack remains slightly higher (as shown by the GEM difference >0). However, on 6 April, while the concentration of GEM at the higher intake height returns to normal ambient concentrations ($\sim 1.75\,\mathrm{ng\,m^{-3}}$) the concentration of GEM in the snow pack increases to over twice that concentration and then returns to normal ambient levels, 12 h later. The large peak in the GEM concentration difference plot in Fig. 4 clearly shows this observation. This phenomenon is seen shortly after solar radiation and snow temperature have peaked for that day and when wind speeds decrease to very low. Similar trends can be seen in Fig. 3 for parts of 14 and 16 March.

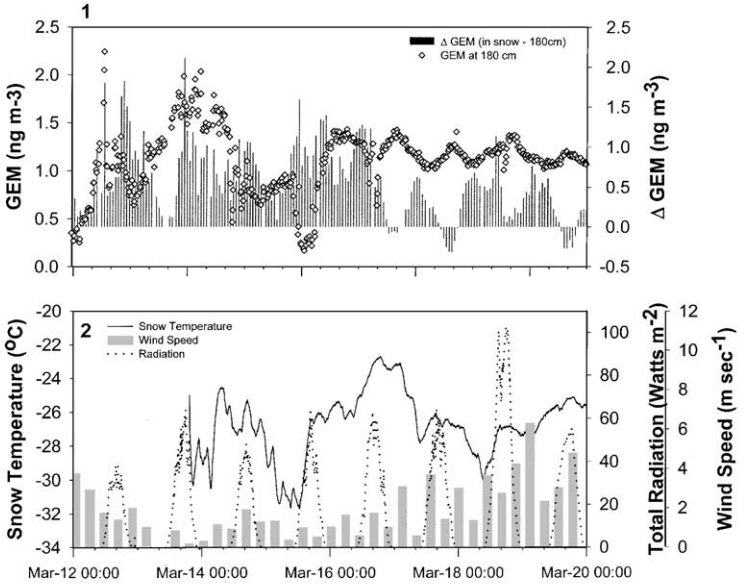

Fig. 3. Time series of GEM concentration at 180 cm above snow surface and of the difference between GEM concentrations in the snow pack and at 180 cm at the SST 12–15 March 2000 (Panel 1). Associated snow pack temperature, total radiation and wind speed (Panel 2).

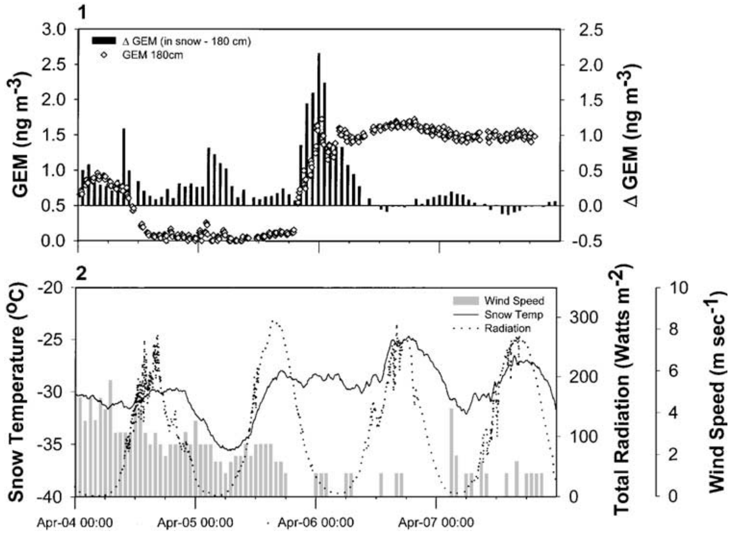

Fig. 4. Time series of GEM concentration at 180 cm above snow surface and of the difference between GEM concentrations in the snow pack and at 180 cm at the SST 4–7 April 2000 (Panel 1). Associated snow pack temperature, total radiation and wind speed (Panel 2).

3.3. Mercury behaviour revealed in the vertical gradient data during MDEs

An extended mercury and ozone depletion event commenced on 26 April and lasted until beyond the end of the measurement campaign (9 May). This allowed us to study the pattern of GEM vertical profiles in more detail during an extended MDE. As detailed by Bottenheim et al. (2002) and Strong et al. (2002), during this major ozone depletion episode winds impacting the Alert region originated from the NW and NNW directions, implying that the air mass emanated from the Arctic Ocean. GEM concentration profiles measured at two heights above the snow surface during this period showed several occurrences of a significant increase of GEM at the near surface level while the concentration at the higher intake level remained low. This "spiking" phenomenon was seen at both the SST and Ice Camp sites. Fig. 5 shows an example of this spiking at the SST between 4 and 6 May where the GEM concentration difference between 1 and 180 cm above the surface (calculated as: $\mathrm{GEM}_{1\,\mathrm{cm}}-\mathrm{GEM}_{180\,\mathrm{cm}}$) and GEM concentration at 180 cm are plotted. Total incoming solar radiation and wind speed (measured at the SST) are plotted as well in Fig. 5. In this case, the solar radiation pattern matches very well with the increased GEM concentration difference between the two inlet heights. This suggests that this spiking phenomenon could be (photo)-chemically induced. Unfortunately, snow temperature data are not available for this period. Wind speeds are consistently quite low but it can be seen that there is little difference in the GEM concentrations at the two heights with winds around $2\,\mathrm{m\,s^{-1}}$. However, when the wind decreased to below $1\,\mathrm{m\,s^{-1}}$ the GEM concentration around the snow surface increases almost threefold (as seen in the significant increase in GEM difference). This reinforces the notion that close to calm conditions were necessary to be able to observe these spikes in GEM. This is not to say that under higher wind conditions the spikes do not occur but rather that they are observed and measured when the wind speed is low. Similar results were obtained at the Ice Camp during this long depletion event. Contrary to the observations for this period at the SST, here there was little indication that the GEM increases were related to solar radiation. Both total UV and total global radiation were investigated for this time period and showed neither a different correlation nor any explanation for this anomaly. It is also interesting to note that ozone did not exhibit such "spiking" behaviour. As discussed previously in this paper, mercury concentrations in the snow are enhanced after MDEs because a significant fraction of GEM in the air is oxidised to RGM or PAM and subsequently deposited to the snow. One hypothesis to explain the GEM spikes is that the oxidised mercury on the snow surface is photo-(chemically) reduced back into its elemental form (GEM) where its physico-chemical characteristics then favour evasion from the snow pack or the surface itself. Supporting evidence for the (photo)-reduction of oxidised mercury to its elemental form in snow samples has been recently reported (Lalonde et al., 2000) as well as such evidence during the snow melt in the Arctic summer (Lu et al., 2001; Lindberg et al., 2001). However, it has not been reported previously in the interstitial snow pack air or in the air at surface levels during the MDE period. Only GEM is measured by our instrumentation thus the RGM and PAM that were deposited to the snow during a MDEs cannot be simply driven from the snow and collected by our instrumentation. We note that snow temperature indicated some relationship with the GEM spikes. A possible scenario is that solar radiation reduces the reactive mercury (RGM and PAM) that has been deposited on the snow surface (during the MDE) back to its elemental form (GEM). The reduced mercury is then increasingly released from the snow surface or snow pack as the snow temperature increases during the day.

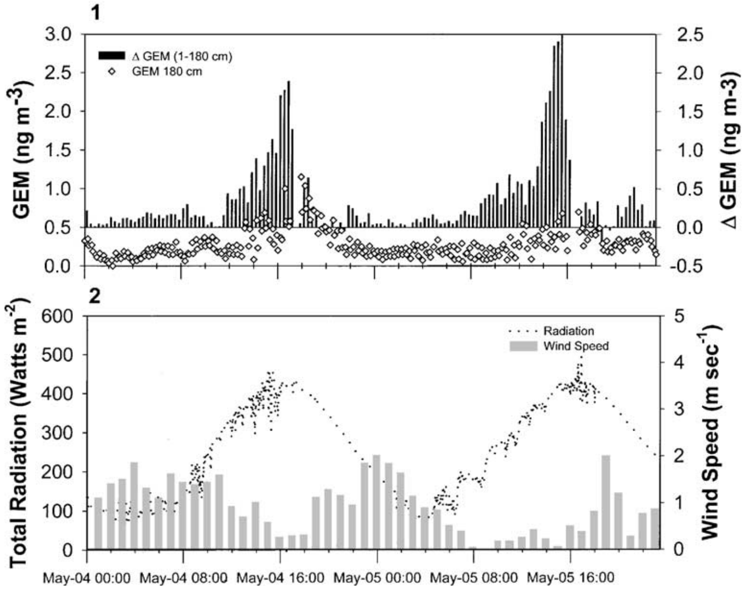

Fig. 5. Time series of GEM concentration at 180 cm above the snow and of the difference in GEM concentration between 1 and 180 cm from the snow surface during an extended MDE at the SST 4–5 May 2000 (Panel 1). Associated radiation and wind speed (Panel 2).

4. Summary and conclusions

The role of GEM in the Arctic environment around polar sunrise was investigated during the Alert 2000 campaign. The CRPU confirmed that GEM is converted to more reactive mercury species during the MDEs and determined that not all the reactive mercury species in the air during MDEs were recovered through pyrolysis. It was postulated that the remaining "unaccounted for" mercury species are deposited on the snow and ice surfaces. Snow collected during this campaign showed substantial increases in the mercury concentrations from the dark to light periods. This finding substantiates the argument that the converted GEM during MDEs is onto the snow.

Vertical gradient air profiling experiments showed that in the non-depletion periods GEM was found to be invariant in the air column between surface and ~ 2 m. However, during the depletion period GEM concentrations sharply increased at the surface. Concurrent ozone concentration profiles show a small gradient in the air column but a sharp decrease in ozone concentration at the surface. This indicates that the snow pack may be a source for mercury and a sink for ozone. Other profile studies between the interstitial air and air above the snow pack show a positive gradient suggesting that GEM is emitted from the snow pack. Further profile studies showed that during MDEs near surface level GEM exhibits spikes of mercury concentrations that over double the ambient GEM concentrations. It is thought that the solar radiation may reduce reactive mercury that is deposited onto the snow surface during a MDE back to its elemental form. An increase in snow temperature may then stimulate the exchange of GEM from the snow pack to the air throughout the day. This is the first report of such an occurrence of mercury in the interstitial and surface air around Arctic snow and further research to better understand this phenomenon is warranted.

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