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GEM fluxes and atmospheric mercury concentrations (GEM, RGM and Hg^p) in the Canadian Arctic at Alert, Nunavut, Canada (February–June 2005)

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Abstract

Five months of gaseous elemental mercury (GEM), reactive gaseous mercury (RGM) and particle bound mercury (Hg^p) concentrations as well as fluxes of GEM were measured at Alert, Nunavut, Canada above the Arctic snow pack. The study spanned February to June of 2005 to capture the effects of polar night, the transition period between night and day as well as polar day on the behaviour of mercury in the near surface atmosphere.

A micrometeorological approach was used to infer the flux of GEM using a continuous two-level sampling system to measure the GEM concentration gradient. The required turbulent transfer coefficients were derived from meteorological parameters measured on site. The flux of GEM was approximately zero during atmospheric mercury depletion events (AMDEs) demonstrating that mercury is not being deposited as GEM to the snow pack. Following AMDEs, there was no evidence of a net emission of GEM. The highest depositional fluxes of GEM occurred during polar night and the largest emission occurred when the tundra was first visible, followed by significant emission and deposition fluxes during the snow melt. Fluxes continued until the snow had completely melted (~JD 170) before returning to near zero.

Average concentrations of RGM ($44.4 \pm 49.8\,pg\,m^{-3}$), Hg^p ($102.6 \pm 124.9\,pg\,m^{-3}$) and GEM ($1.0 \pm 0.4\,ng\,m^{-3}$) were variable throughout the study due to the dynamic nature of atmospheric mercury during AMDEs. Increases in Hg^p preceded elevated levels of RGM during AMDEs by 60 days, yielding peak levels at 694 and $344\,pg\,m^{-3}$, respectively. Elevated concentrations of Hg^p typically occurred when the specific humidity dropped below $0.75\,g\,kg^{-1}$, winds were light ($<3\,m\,s^{-1}$) and the air temperature dropped below $-20\,^{\circ}C$. Increased levels of RGM were also noted when the winds were light ($<3\,m\,s^{-1}$) but when the temperature increased above $-10\,^{\circ}C$ and the specific humidity was in the range of 1 and $3\,g\,kg^{-1}$. As different environmental conditions were observed for the elevated concentrations of Hg^p versus RGM at Alert, it suggests that the formation mechanisms for each species may be different but tied to the atmospheric temperature and water content.

Total mercury (TM) levels in fresh snow measured approximately $5–10\,ng\,l^{-1}$ during AMDEs and reached nearly $80\,ng\,l^{-1}$ outside of depletion events, suggesting that wet deposition may not be a significant removal mechanism of GEM

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during depletion events. An unusually high concentration of TM was measured during a non-depletion event which coincided with the transitional period where atmospheric loading of RGM exceeded levels of Hg^p.

Keywords: Arctic; Flux gradient; Gaseous elemental mercury; Mercury depletion events; Micrometeorology; Particle bound mercury; Reactive gaseous mercury

1. Introduction

Mercury (Hg) in the Arctic is of considerable interest due to its toxicity and presence in a location far removed from anthropogenic emission sources. Atmospheric mercury is found predominantly (~95%) as gaseous elemental mercury (GEM) and is susceptible to long range transport due to its atmospheric residence time of approximately 1–2 years (Slemr et al., 1985; Schroeder and Munthe, 1998). In 1995, Schroeder et al. (1998) first noted atmospheric depletion of GEM at Alert, Nunavut, Canada following polar sunrise which are now termed atmospheric mercury depletion events (AMDEs). Less abundant forms of Hg also exist in the atmosphere as reactive gaseous mercury (RGM) and particle bound mercury (Hg^p) although the distribution between the three species changes dramatically during AMDEs between March and June at Alert, Nunavut.

GEM has been measured continuously at Alert since 1995 and is ongoing (Steffen et al., 2005). During the 1998 polar sunrise, a pyrolysis unit was employed at Alert and it was confirmed that some of the GEM was being converted to a more reactive form during depletion events and subsequent increases of total mercury (TM) in snow were noted (Steffen et al., 2002). During the Alert 2000 campaign, GEM profile measurements were made and suggested that concentrations were invariant with height between the snow surface and 2 m during non-depletion events although, during depletion events there was a noticeable increase at the snow surface (Steffen et al., 2002). In the spring of 2001, a Tekran 1130/35 mercury speciation unit was installed at Alert and increases in Hg^p and RGM were noted during AMDEs.

From what is currently known of the atmospheric chemistry of mercury in the Arctic, measuring mercury speciation concentrations is insufficient to determine the chemical mechanisms occurring and the long-term GEM flux measurements concurrent with mercury speciation measurements are essential in understanding the fate of mercury in the Arctic. Long-term GEM flux measurements have not been completed in the Arctic and most GEM flux studies occur following polar sunrise. The first goal of this study was to determine the magnitude and significance of GEM concentrations and fluxes as well as the concentrations of RGM and Hg^p above the Alert snow pack through three distinct periods (polar night, transition from night to day and polar day). The second goal was to provide insight into AMDEs from information provided by continuous GEM flux and micrometeorological measurements.

2. Experimental

2.1. Sampling

Five months of GEM, RGM and Hg^p concentrations as well as fluxes of GEM were continuously measured at Alert, Nunavut, Canada (82°58′N, 62°38′W) between February and June 2005 (Fig. 1, upper left). For the purposes of this research, an AMDE was considered to occur if the concentration of GEM decreased below 1 ng m^{-3} and the Hg^p or RGM concentrations reached levels greater than the mean (complete study mean for each species) plus 1 standard deviation. Snow table sampling for TM was also completed on a snow event basis. The measurement campaign covered three distinct periods including polar night (Julian day 31–62), the transition period from night to day (Julian day 63–94) and polar day (Julian day 95–174).

All measurements were conducted at the Special Studies Trailer (SST) (Fig. 1, upper right) located 6 km south of the Canadian Forces Station (CFS) Alert located on the Arctic Ocean shoreline (Ridley et al., 2000). Alert is a rural-remote site located at the northern tip of Ellesmere Island on a plateau 205 m above sea level (Steffen et al., 2005). There are four hills approximately 800 m in height located 5–6 km to the south of the SST and a mountain range about 40 km to the west (Ridley et al., 2000). The surface surrounding the study site is tundra which consists of dark soils, dominated by mosses, lichens, herbs and subsoil that is permanently