Atmospheric Environment 43 (2009) 5604–5610

Contents lists available at ScienceDirect

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv

Dynamic exchange of gaseous elemental mercury during polar night and day

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ARTICLE INFO

Article history:
Received 10 February 2009
Received in revised form
28 July 2009
Accepted 29 July 2009

Keywords:
Arctic
Gaseous elemental mercury
Flux gradient
Ny-Ålesund

ABSTRACT

From February 29 until June 15 2008 gaseous elemental mercury (GEM) fluxes above a snow covered surface was measured in Ny-Ålesund, Svalbard using a GEM flux gradient method. A clear seasonal pattern in the meteorological variables associated with the GEM flux was observed. For the first time in Ny-Ålesund a net deposition of GEM was recorded during polar night, despite the lack of Atmospheric Mercury Depletion Events (AMDE). 7500 ng m^{-2} GEM was emitted from the surface snow to the air during the entire study. The depositions of GEM and reactive gaseous mercury (RGM) were calculated to be 1500 and 1000 ng m^{-2}, respectively, during the same time period. The GEM fluxes reported in this study were found to be comparable to GEM fluxes measured at other Arctic locations (i.e. Alert and Barrow), suggesting that GEM acts in a similar way throughout the Arctic. An assessment of the GEM flux gradient method used discovered a non-linear GEM concentration profile. The nonlinearity was explained by a non-stationary turbulence regime. The GEM flux calculated was not found to be representative for the entire surface boundary layer.

1. Introduction

Mercury (Hg) as a global pollutant has been of great concern in Arctic environments ever since the discovery of the annual recurring spring time phenomenon labelled the Atmospheric Mercury Depletion Event (AMDE) (Schroeder et al., 1998). Furthermore, the occurrence of AMDEs is circumpolar, they have been observed at Barrow, Alaska (Lindberg et al., 2001), Ny-Ålesund, Svalbard (Berg et al., 2003), Kuujjuarapik, Quebec (Poissant, 2003), Station Nord, Greenland (Skov et al., 2004), Amderma, Russia (Steffen et al., 2005) and Andøya, Norway (Berg et al., 2008) in addition to Neumeyer, Antarctica (Ebinghaus et al., 2002). It is agreed that gaseous elemental mercury (GEM) is converted to reactive gaseous mercury (RGM) during an AMDE. RGM is subsequently either deposited to snow and ice surfaces or associated with aerosols (PHg) (Steffen et al., 2008). It is estimated that AMDEs can lead to the deposition of up to 300 tonnes of Hg per year to the Arctic (Ariya et al., 2004). Driven by the fact that mercury (Hg) has strong toxic properties the post depositional fate of Hg needs to be specified. Since the first observation of an AMDE researchers have been interested in determining the air to surface exchange (i.e. flux) of Hg. In Ny-Ålesund GEM fluxes have previously been measured by the chamber technique (Ferrari et al., 2005, 2008; Sommar et al., 2007). Additionally, vertical GEM gradients indicating a possible GEM flux have been measured (Sommar et al., 2007; Anne Orderdalen Steen, unpublished results). These measurements were conducted for a few weeks, during spring only.

The development of a micrometeorological approach for measuring GEM fluxes was motivated by the need for an improved method to study fluxes from Hg-emitting surfaces. At present, the intake height used in the GEM concentration gradient sampling system employed in GEM flux gradient measurements are not standardized. Steffen et al. (2002) used intakes at 1, 39, 98 and 190 cm, respectively at Alert. Brooks et al. (2006) reported GEM fluxes over a 20 m continuously flushed gradient system at Barrow. Cobbett et al. (2007) used mainly 2.5 cm and 100 cm as lower and upper intake heights at Alert. The study performed by Sommar et al. (2007) in Ny-Ålesund used intakes at 20 and 180 cm, respectively. Long-term GEM flux measurements are lacking. The only study to date was performed by Cobbett et al. (2007).

This study which discusses the longest-running series of GEM flux measurements in Ny-Ålesund, reveals a seasonal pattern in both the GEM flux itself and the meteorological variables associated with this flux. Additionally we assess the GEM flux gradient method used to quantify the GEM exchange between the air and the surface snow. The behaviour of the GEM concentration gradient

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doi:10.1016/j.atmosenv.2009.07.069

profile is discussed extensively. Finally, recommendations for future applications of the method and suggestions for prospective work are proposed.

2. Experimental

2.1. Study site

Ny-Ålesund International Research and Monitoring Facility (78°54′N, 11°52′E) is located next to the southern shore of Kongsfjorden on the west coast of Spitsbergen (Fig. 1). The settlement is surrounded by steep mountains ranging in altitude from 500 to 1000 m to the south and east of Kongsfjorden, and open sea to the west and north. The instrumental setup for the GEM flux gradient was located 200 m from the settlement in Ny-Ålesund (Fig. 1). RGM measurements were performed at the Zeppelin air monitoring station at 474 m above sea level. A detailed description of the sampling locations used is given in Aspmo et al. (2005). Although the snow started to melt in late May, the surface was covered with snow during the entire study.

2.2. GEM flux gradient measurements

The first long-term GEM flux gradient measurements between the air and the surface snow in Ny-Ålesund were performed from February 29 until June 15 2008.

The aerodynamic flux gradient approach, which measures the turbulent transfer coefficient (K), is based on Monin–Obukov (MO) similarity involving near-simultaneous measurements of GEM at two heights above the surface (Edwards et al., 1997; Edwards, 2005). The GEM concentration gradient was recorded between intakes at 1 cm and 50 cm above the surface snow using a Tekran 2537A instrument (cold vapour atomic fluorescence spectrometry). A synchronized switching device (Tekran 1110) allowed for alternate sampling between the two intakes.

The average flux is expressed as:

$$F' = -K\frac{\partial c}{\partial z} \approx \frac{u_* k(c_2 - c_1)}{\ln[z_2 - d/z_1 - d] - \psi h2 + \psi h1} \quad (1)$$

Where, F' is the GEM flux (ng m^{-2} s^{-1}); K is the turbulent transfer coefficient (i.e. eddy diffusivity) (m^2 s^{-1}); $\delta c/\delta z$ is the GEM concentration gradient (ng m^{-1}); u_* is the surface friction velocity (m s^{-1}); k is the von Karman's constant (0.4) (unitless); z_2 is the upper GEM intake height (m); z_1 is the lower GEM intake height (m); c_2 is the GEM concentration measured at height z_2; c_1 is the GEM concentration measured at heights z_1; d is the zero plane displacement height (i.e. the snow depth) (m); ψ_{h1} and ψ_{h2} are the integrated stability functions for heat at z_1 and z_2.

For further details concerning the calculations of the GEM fluxes, see Edwards et al. (2005).

In this paper F′ is multiplied by 3600. Subsequently the GEM fluxes are reported as ng m^{-2} h^{-1}.

A Sonic anemometer (CSAT3, Campbell Scientific) was mounted 125 cm above the surface with a consistent fetch of 100 m. Micrometeorological values were sampled with a frequency of 100 Hz and averaged over 20 min. These yielded values for sensible heat flux and surface friction velocity (CS5000, Campbell Scientific).

The GEM concentration gradient was based on 5 min cycles for the collection of GEM on gold traps, followed by thermal desorption and the detection of GEM by the Tekran 2537A instrument. GEM was measured for two cycles (i.e. for 10 min) at each height to allow for bias between the traps. Both sample intakes were continuously flushed with ambient air. The sample flow rate was 1.5 l min^{-1}. Teflon lines (¼" outer diameter) and fittings were used. Teflon

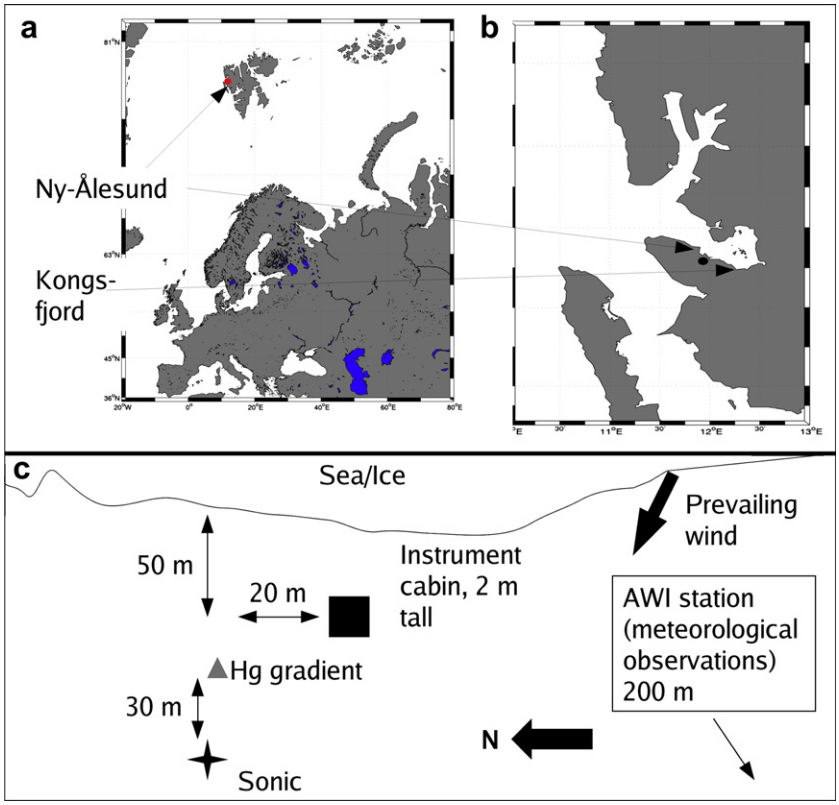

Fig. 1. Map indication (a) Svalbard in Europe, (b) the location of Ny-Ålesund on Svalbard and (c) a detailed description of the instrument locations.