

Measurements of atmospheric mercury species during an international study of mercury depletion events at Ny-Ålesund, Svalbard, spring 2003. How reproducible are our present methods?

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Abstract

Six groups participated in an international study of springtime atmospheric mercury depletion events (AMDEs) at Ny-Ålesund in the Norwegian Arctic during April and May 2003 with the aim to compare analytical methods for measurements of atmospheric mercury species and study the physical and chemical processes leading to AMDEs. Five groups participated in the method comparison that was conducted at three different locations within Ny-Ålesund. Various automated and manual instrumentation were used to sample, measure and compare gaseous elemental mercury (GEM), reactive gaseous mercury (RGM) and mercury associated with particles (Hg-P). The concentration of GEM was reproducible during background conditions. For the first time using ambient air, the statistics associated with round robin test procedures were applied. This was found to be an appropriate tool to investigate the reproducibility of GEM measurements in ambient air. The precision for each group measuring GEM concentrations was found to be consistently good (within 5%). Five AMDEs were recorded during the study. Using four different methods, including single and replicate samples, all groups recorded higher values of RGM and Hg-P during AMDEs. The results show that measuring comparable atmospheric mercury species at both the same and different locations (within the Ny-Ålesund area) is difficult. Not only do site location and site characteristics create challenges when trying to intercompare results but there are difficulties, as well, in obtaining comparable results with similar sampling and

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analysis methods. Nevertheless, with our current procedures for atmospheric mercury identification we can differentiate with certainty between “high” and “low” concentration values of RGM and Hg-P.

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1. Introduction

Mercury is an environmental pollutant of global concern because of its exceptional behavior due to volatility and ability of methylation (Slemr, 1985). Mercury is among the most highly bioconcentrated trace metals in the human food chain, especially in marine ecosystems (AMAP 2002; Steffen et al., 2002). In the environment, mercury is present in various physical and chemical forms which have different characteristics of transport, deposition and impacts on ecosystems (Schroeder and Munthe, 1998). The speciation of atmospheric mercury is important in order to understand the fate and impact of this pollutant on the environment. Prior to 1995, elemental mercury (hereafter called gaseous elemental mercury (GEM)) was normally the most abundant form of mercury in the atmosphere with a northern hemispheric background concentration of approximately 1.7 ng m^{-3} (Slemr et al., 2003). GEM was also thought to have a residence time of about 6–24 months, allowing it to be globally transported far from its emission sources (Wängberg et al., 2001). Oxidized mercury compounds, such as the operationally defined reactive gaseous mercury (RGM) and mercury associated with airborne particulate matter (Hg-P), are normally found at much lower concentrations (in the pg m^{-3} range) in the air.

The reactivity of GEM in the atmosphere is weak except under special conditions in which GEM can be rapidly oxidized. These fast atmospheric processes, known as atmospheric mercury depletion events (AMDEs), have been observed throughout the Polar Regions (Alert (Schroeder et al., 1998), Barrow (Lindberg et al., 2002), Ny-Ålesund (Berg et al., 2001, 2003a), Greenland (Skov et al., 2004), Amderma (Steffen et al., 2004) and Antarctic (Ebinghaus et al., 2002)). The lifetime of the oxidized mercury fractions is moderate and they have a higher affinity to condensed phases, making them more prone to deposition (Lindberg and Stratton, 1998). The chemical composition and physical structure of RGM and Hg-P cannot be exactly identified by the experimental methods available today, but are instead characterized by their properties and capability to be collected by different sampling systems. Munthe et al. (2001) defines RGM as inorganic water-soluble mercury species with a sufficiently high vapor pressure to exist in the gas phase, and Hg-P as inorganic

(including Hg^0) and organic mercury species bound to and/or adsorbed to airborne particulate matter. Several chemical components are proposed as likely candidates for RGM and Hg-P, such as HgBr_2 , HgCl_2 and HgO (Munthe et al., 2001; Calvert and Lindberg 2003; Goodsite et al., 2004).

Since the discovery of AMDEs in 1995 at Alert (Schroeder et al., 1998), a lot of work has been carried out to develop methods to identify and quantify the different atmospheric mercury species produced during these events. Additionally, the processes behind these events and their impacts on the Arctic environment are under much investigation (Schroeder et al., 2003). During the past few years, new manual and automated methods to measure GEM, RGM and Hg-P have been developed and are now commercially available. An historic review of the development of methods for speciation of mercury in air is given by Munthe et al. (2001) and Landis et al. (2002). Currently, no calibration standards are available for these operationally defined methods as the chemical composition of the oxidized mercury fractions are presently not known, thus making field intercomparison studies a necessary tool to investigate the performance of the methods.

Field intercomparison is an established practice to determine the degree of comparability of measurement results by comparing the sampling and analytical procedures used by different groups around the world (Ebinghaus et al., 1999). In practice, precision often varies when replicates are performed by different laboratories or even within the same laboratory at different times. This also happens during field intercomparisons thereby making it necessary to distinguish between the repeatability and the reproducibility of the individual results. Several field intercomparison studies of atmospheric mercury species have been performed (Schroeder et al., 1995; Lu et al., 1998; Ebinghaus et al., 1999; Munthe et al., 2001; Landis et al., 2002; Wängberg et al., 2003) at temperate latitudes, but a very limited number of field intercomparison studies have been performed in the Arctic, even less during AMDEs. At present, there is a general comprehension regarding the reliability of methods during background conditions, but the performance of the currently used methods during AMDEs has never been documented. Previous studies under arctic conditions have indicated insufficient comparability of RGM and Hg-P measurements

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