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Simultaneous monitoring of total gaseous mercury at four urban monitoring stations in Seoul, Korea

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ABSTRACT

The monitoring of total gaseous mercury (TGM) concentrations together with other trace gases and meteorological parameters was made at hourly intervals over 2-year period (1 January 2010 to 31 December 2011) at four urban monitoring sites: Guro-gu (G), Nowon-gu (N), Songpa-gu (S), and Yongsan-gu (Y) in Seoul, Korea. The mean concentrations of TGM at these sites were found to span the range of 3.28 (Y) to 3.47 ng m^{-3} (N). Inspection of the seasonal patterns indicates that the maximum concentrations (ng m^{-3}) occur at different times of year across the four sites: winter at N (3.67 ± 1.77), fall at S (3.64 ± 1.12), summer at G (3.61 ±1.51), and spring at Y (3.40 ± 1.26). The long-term trend in Hg concentrations, when also considering data sets from previous studies, suggests modest reductions in concentrations at all four sites, from 2.89 to 4.49 ng m^{-3} in 2004 and from 2.49 to 3.42 ng m^{-3} in 2011.

1. Introduction

Mercury (Hg) is a well-known harmful and toxic pollutant that can cause adverse effects to human health and wildlife. It exists in several forms and phases: (1) elemental (also known as metallic mercury) or oxidized, (2) vapor in the air or dissolved in liquid matrices, and (3) inorganic or organic (with all species having different toxic effects (WHO, 1991)). Because of its high vapor pressure, elemental mercury can be easily vaporized and transported to other environmental compartments. The distribution of mercury in air thus depends on its chemical and physical form and the rate of its interconversion between different species in the various environmental compartments.

Mercury is emitted into the atmosphere by both natural and anthropogenic processes. According to the global emission inventory of mercury for 2005, approximately 1930 tons of mercury was emitted from anthropogenic sources, two-thirds of which came from Asian countries (Pacyna et al., 2010). The combustion of fossil fuels is estimated to account for 45% of anthropogenic sources worldwide. Other anthropogenic sources include such artisanal/small-scale gold mining (18%), industrial gold production (5–6%), other mining and metal production activities (10%), and cement production (10%) (Pacyna et al., 2010). It is estimated that anthropogenic sources may comprise 70% of total mercury emissions to the atmosphere (Schuster et al., 2002). The mercury emission inventory in Korea shows a pattern of emission profiles similar to the global inventory (Pudasainee et al., 2009). According to Kim et al., (2010), thermal power plants were the largest source of Hg emission in Korea, accounting for about 3.30 tons (26%). Oil refineries, cement kilns, medical waste incinerators, iron manufacturing,

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and sludge incinerators were responsible for about 3.26 (25%), 2.69 (21%), 1.13 (9%), 0.91 (7%), and 0.60 tons (5%), respectively. The remaining sources (crematoria, pulp and paper manufacturing, nonferrous metals manufacturing, and mobile sources) contributed 0.92 tons (7%) (Kim et al., 2010).

Human activities are considered as the major sources of Hg emission in urban areas. This can lead to enhanced Hg levels relative to the local regional background (Lee et al., 1998; Fang et al., 2010; Nguyen et al., 2007, 2010b; Liu et al., 2011). The environmental behavior of Hg species in urban areas has been investigated by a number of previous studies (e.g., Kim and Kim, 2001a, 2002; Kim et al., 2011). Long-term trends of Hg concentration in an urban area were also examined to assess its relationship to changes in source processes (Kim and Kim, 2002; Shon et al., 2008). For instance, a significant reduction in Hg concentrations seen throughout the 1980s and 1990s in Korea was ascribed to decreases in the consumption of anthracitic coal. Choi et al. (2009) suggested that the wintertime rise in total gaseous mercury (TGM) and carbon monoxide (CO) concentrations in a southern part of Seoul was caused by increased emissions (due to the high consumption of fossil fuel) and low mixing heights.

We have been monitoring concentrations of TGM at four urban sites in Seoul, Korea, since 2004 (Kim et al., 2011). The present study is a significant extension of our previous efforts to collate Hg concentration data in urban areas by continuing the monitoring at these four urban sites. In this study, Hg data measured at these sites during 2010 and 2011 were analyzed to elucidate recent trends and evaluate the relationship with previous years.

2. Materials and methods

2.1. Site characteristics

In this study, the concentration of TGM (or Hg unless otherwise stated) was measured at four monitoring sites in Seoul, Korea, as shown in Fig. 1. Concentrations of Hg in air at these monitoring stations were measured throughout 2010 and 2011. These monitoring stations were operated and managed by the Seoul Metropolitan Research Institute of Public Health and Environment (SRIE).

Detailed information on the operations of these four monitoring stations has previously been given (Kim et al., 2011). The four study sites are referred to by the initials of each district name: Guro-gu (G), Nowon-gu (N), Songpa-gu (S), and Yongsan-gu (Y) (Fig. 1). Their precise locations in terms of GPS longitude and latitude are Y (127:00:18 and 37:32:23), N (127:04:06 and 37:39:31), G (126:53:33 and 37:28:48), and S (127:07:27 and 27:31:17).

2.2. Sampling and analysis

The concentrations of Hg at each site were measured every hour for the entire period from 1 January 2010 to 31 December 2011. The hourly Hg concentration data were measured by sampling outdoor air through a 2-m-long sampling line comprising Teflon tubing (30 mm diameter) into an automatic analytical system (AM-3 model, the Nippon Instrument Co., Japan), which contained both combined sampling and analytical devices. This sampling tubing was placed to stick out of a wall of the building without any obstacles in the forefront (>10 m). During each hour, Hg was collected onto an Au-amalgam trap at a constant flow rate of 1.0 L min^{-1}. As TGM is an operationally defined fraction passing through a filter or some other simple filtration device (e.g., quartz wool plug), a 47-mm-diameter Teflon pre-filter was used to protect the sampling system against contamination by particulate matter (Munthe et al., 2001). At the end of each hour, the Hg collected was desorbed thermally and detected at wavelength of 253.7 nm by a non-dispersive double beam, flameless cold vapor atomic absorption spectrometry (CVAAS) system. The absolute detection limit of the system was found out to be approximately 1–2 pg of Hg, yielding a mass concentration detection limit in air of between approximately 15 and 30 pg/m^3. The relative precision of the analytical system averaged 0.3–0.6% at the concentrations measured. The analytical system was validated against several National Institute of Standards and Technology (NIST) standard reference materials (e.g., NIST SRM 1632d), and the mean accuracy was in the 3–5% range. The calibration of each system deployed at all four locations was made manually by withdrawing Hg vapor generated from a bell-jar calibration apparatus on a quarterly basis. The concentrations of all selected air pollutants ($PM_{2.5}$, PM_{10}, TSP, CH_4, CO, NO, NO_2, NO_x, O_3, SO_2, total hydrocarbon (THC), and non-methane hydrocarbon (NMHC)) and relevant meteorological parameters (temperature (TEMP), relative humidity (RH), wind speed (WS), and ultraviolet (UV)) were also monitored at each site.

2.3. Statistical analysis

In this study, the statistical analysis of our measurement data was performed using Microsoft Excel software. The diurnal variations of Hg (and other trace gases) at the four sites were examined using a *t*-test to judge the statistical significance between daytime and nighttime concentrations. In addition, the possible relationship between Hg and other measured parameters was also examined by Pearson's correlation analyses at significance levels of $p < 0.05$ or $p < 0.01$.

3. Results and discussion

3.1. Hg distribution at the four urban sites

All data were evaluated after calendar daily average had been produced from the series of hourly values (unless otherwise stated). A statistical summary of the daily averages of all different parameters is provided in Table 1. The mean concentrations of Hg obtained throughout the 2-year period were in a fairly narrow concentration range around 3 ng m^{-3}: N (3.47 ± 1.48 ng m^{-3}), S (3.43 ± 1.24 ng m^{-3}), G (3.33 ± 1.39 ng m^{-3}), and Y (3.28 ± 1.27 ng m^{-3}). These Hg concentrations are lower than those measured during 2004–2009, which fell in the range 3.36 (Y) to 3.98 ng m^{-3} (N) (Kim et al., 2011). Given the standard deviations on the concentrations measured in 2010–2011, it is not possible to conclude that these are significantly different to those measured in the preceding 6 years.

The average concentrations of Hg (ng m^{-3}) in this study were considerably lower than those measured recently in other Asian cities, for instance in Chinese cities such as Chongqing where concentrations were 6.74 ± 0.37 (Yang et al., 2009) and