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Atmospheric particulate mercury in the megacity Beijing: Efficiency of mitigation measures and assessment of health effects



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HIGHLIGHTS

• Study of particulate mercury (HgP) in aerosols from 2005 to 2013 in summer and winter.

- Evaluation of short- & long-term success of mitigation measures during Olympic Games.
- HgP in Aug-08 decreased by 65% compared to previous years due to implemented measures.
- Decrease in winter indicated slight long-term improvement of HgP pollution in Beijing.
- But still high HgP levels regarding adverse health effects even after the reductions.

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ABSTRACT

Atmospheric particulate mercury (HgP) was studied before, during, and after the Olympic Summer Games in Beijing, China, in August 2008 in order to investigate the efficiency of the emission control measures implemented by the Chinese Government. These source control measures comprised traffic reductions, increase in public transportation, planting of vegetation, establishment of parks, building freeze at construction sites, cleaner production techniques for industries and industry closures in Beijing and also in the surrounding areas. Strictest measures including the "odd-even ban" to halve the vehicle volume were enforced from the 20th of July to the 20th of September 2008. The Olympic period provided the unique opportunity to investigate the efficiency of these comprehensive actions implemented in order to reduce air pollution on a large scale. Therefore, the sampling period covered summer (August, September) and winter (December and January) samples over several years from December 2005 to September 2013. Average HgP concentrations in total suspended particulates (TSP) sampled in August 2008 were 81 \pm 39 pg/m³ while TSP mass concentrations were 93 \pm 49 μ g/m³. This equals a reduction by about 63% for TSP mass and 65% for HgP, respectively, compared to the previous two years demonstrating the short-term success of the measures. However, after the Olympic Games, HgP concentrations increased again to pre-Olympic levels in August 2009 while values in August 2010 decreased again by 30%. Moreover, winter samples, which were 2- to 11-fold higher than corresponding August values, showed decreasing concentrations over the years indicating a long-term improvement of HgP pollution in Beijing. However, regarding adverse health effects, comparisons with soil guideline values and studies from other cities highlighted that HgP concentrations in TSP remained high in Beijing despite respective control measures. Consequently, future mitigation measures need to be tailored more specifically to further reduce HgP concentrations in Beijing.

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1. Introduction

Mercury transport in the atmosphere occurs predominantly as gas (volatilized Hg) but also adsorbed on particulate matter/aerosols (Fang et al., 2011) and its distribution covers the global hemisphere scale. The atmospheric residence time of gaseous elemental mercury (GEM) is at least a few months or even one to two years and, thus, it is much greater than for reactive gaseous mercury (RGM) with a few hours or days and for particulate mercury (HgP) with a residence time of a few weeks (e.g. Lindqvist and Rodhe, 1985; Schroeder and Munthe, 1998). Consequently, GEM can be transported over long distances, whereas HgP and RGM reflect more local and regional sources (e.g. Wängberg et al., 2003; Wang et al., 2006; Xiu et al., 2009). While HgP may constitute, in general, a small percentage by mass of total atmospheric mercury, it can be a very important form of atmospheric Hg under certain conditions, and play a strong role in the deposition of mercury to terrestrial and aquatic cosystems (Lu and Schroeder, 1999). Most researchers agree that HgP is critical in understanding the mercury cycle in the environment (Fang et al., 2001). In China, HgP normally constitutes a high proportion of urban airborne Hg, which during episodes may approach 10% (Fu et al., 2012). Streets et al. (2009) examined the effect of possible future scenarios (projections for 2050) on relative emissions of the different species of Hg. The authors concluded that there would be a relative trend toward reduced long-range transport and enhanced local deposition of Hg. Studies focusing on PHg will therefore gain further importance in future.

Globally, artisanal and small-scale gold mining and coal burning are the major sources of anthropogenic mercury emissions to air (UNEP, 2013). China accounts for three-quarters of East and Southeast Asian anthropogenic Hg emissions, or about one third of the global total (UNEP, 2013). Streets et al. (2005) provide an inventory of Hg emissions from anthropogenic sources in China and estimated that in 1999 45% of the Hg came from non-ferrous metals smelting, 38% from coal combustion, and 17% from miscellaneous activities, of which battery and fluorescent lamp production and cement production were the largest. Particulate mercury emissions are high in China due to heavy burning of coal in residential and small industrial settings without PM controls (Streets et al., 2005). Streets et al. (2009) predict a further increase in Hg emission for the future with Asia as the main driving force. Sources for HgP are abundant in a Chinese megacity like Beijing. Coal combustion for industrial and residential use as well as coal-fired power plants constitutes a major source. Coal still is the dominant energy source in China, covering about 70% of the total energy consumption, which accounted for 2.6 billion tons Standard Coal Equivalent (SCE) in 2007 (National Bureau of Statistics of China, 2009). In Beijing, coal consumption reached almost 30 million tons of SCE in 2007 (Beijing Municipal Bureau of Statistics, 2009). Other potential sources are non-ferrous metal smelting, cement production, Hg mining, biofuel and biomass combustion (Fu et al. 2012). The manufacture of cement contributed about 10% (190 t) to the total global emissions of mercury to air from anthropogenic sources in 2005 and China, as the main contributor, was responsible for 45% of total emissions from cement manufacturing (UNEP, 2010). Mercury emissions from Zn production were estimated to be 80.7-104.2 t per year in China from 2002 to 2006 (Li et al., 2010). Also municipal solid waste incineration is a potential Hg source, which plays an important role in China (Chen et al., 2013).

The main focus of this study lies on the Olympic Summer Games in August 2008 in Beijing with its strictly enforced mitigation measures. The Olympic period (8th–24th of August 2008) provided the unique opportunity to investigate the efficiency of these comprehensive actions implemented in order to reduce air pollution on a large scale. The emission control measures comprised traffic reductions, increase and improvements in public transportation, planting of vegetation, establishment of parks, building freeze at construction sites, cleaner production techniques for industries and industry closures in Beijing and also in the surrounding areas (Fang et al., 2009; Schleicher et al., 2012). Strictest measures including the "odd-even ban" to halve the vehicle volume was enforced from the 20th of July to the 20th of September 2008. Several studies investigated the efficiency of the governmental control measures on precursor gases (Sun et al., 2011), black carbon (BC) (Wang et al., 2009a; Okuda et al., 2011; Schleicher et al., 2011a), and also metal concentrations (Schleicher et al., 2012; Chen et al., 2014). All these studies reported that mitigation measures were successful in improving air quality but had different effects on distinct air pollutants. For example, Schleicher et al. (2011a) showed that mass concentrations of particles of different size classes within the coarse mode $(2.5-80 \,\mu\text{m})$ during this period decreased, and that the coarser particles were reduced more efficiently than the finer ones. Another study by Schleicher et al. (2012) reported a reduction by 50-70% for elements predominantly from anthropogenic sources, such as S, Cu, As, Cd, or Pb, whereas elements mainly from geogenic sources, such as Fe, Rb, and Sr were reduced by only 30-50% during the Olympic Games. Chen et al. (2014) showed that concentrations of traffic-related elements, such as Pb, Sb, Sn, and Zn, varied with the strength of traffic restriction measures.

To the authors' knowledge, no study investigated how control measures affected HgP concentrations during the Olympic Games in Beijing. Wang et al. (2013) studied the effect of the shutdown of a large coal-fired power plant located in Rochester, New York, USA. on ambient mercury species. The authors reported a decrease of 25% for GEM, 74% for RGM, and 50% for HgP after its closedown for four months. However, it is of special interest to evaluate to what extent the comprehensive set of control measures during the Olympic Games, which was designed to reduce particulate air pollution but not HgP concentrations specifically, affected HgP pollution levels in Beijing. Therefore, the data set presented here includes weekly TSP samples of each August from 2006 to 2010 in order to gain detailed knowledge about HgP concentrations before, during, and after the Olympic Games period and, thus, to examine the efficiency of mitigation measures under realistic conditions. In August 2008, TSP samples were collected with a higher timeresolution of 24 h in order to assess day-to-day differences. Additionally, September samples until 2013 were analyzed in order to investigate the longer development after the Olympic Games. Furthermore, winter samples (December, January) from various years before and after the Olympic Games (winter 2005/2006 to 2010/2011) are included since HgP concentrations in Beijing are highest during winter season (Wang et al., 2006; Schleicher et al., 2015). With this comprehensive approach it was possible to estimate the short- and long-term effects of the emission control measures on HgP concentrations and to assess the health benefits for the affected population.

2. Methodology

2.1. Sampling

The sampling site, labeled as CRAES, is located in the north of the inner city of Beijing close to the Central Olympic District (COD, Fig. 1). The site is placed on the roof of the Chinese Research Academy of Environmental Sciences (CRAES) at a height of about 20 m. At this site, weekly TSP samples were collected continuously since October 2007. Weekly samples represent filters that were collected over the complete 168 h, i.e. integrating the whole period. In August 2008, sampling was carried out with a higher time-

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