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Observations of atmospheric Hg species and depositions in remote areas of China

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Abstract. From September 2007, we conducted continuous measurements of speciated atmospheric mercury (Hg) and atmospheric mercury depositions at five remote sites in China. Four of these sites were involved in the Global Mercury Observation System (GMOS) as ground-based stations. These stations were located in the northwest, southwest, northeast, and east part of China, respectively, which represent the regional atmospheric Hg budgets in different areas of China. The preliminary results showed that mean TGM concentrations were in the range of 1.60 – 2.88 ng m^{-3}, with relatively higher levels observed at sites in Eastern China and Southwestern China and lower levels at sites in Northeastern and Northwestern China. TGM concentrations at remote sites of China were also higher than those reported from background sites in North America and Europe, and this is corresponding very well with the Chinese great anthropogenic Hg emissions. Gaseous oxidized mercury (GOM) and particulate bounded mercury (PBM) were in the ranges of 3.2 – 7.4 pg m^{-3} and 19.4 – 43.5 pg m^{-3}, respectively. The preliminary result on precipitation showed mean precipitation THg concentrations were in the range of 2.7 – 18.0 ng L^{-1}.

Key words: Atmospheric mercury; China; Remote sites; Wet mercury deposition

Introduction

Mercury (Hg) is a persistent, bio-accumulative and toxic chemical in the environment and has potential adverse effects to human health. Hg in the atmosphere, which is derived from both anthropogenic and natural emission sources, is generally operationally defined into three major forms, namely elemental gaseous mercury (GEM), reactive gaseous mercury (RGM), and particulate mercury (PHg), with the sum of GEM and RGM known as total gaseous mercury (TGM). Due to the fast developing economy, anthropogenic Hg emission has been increasing for many years and is ranked as the world's largest anthropogenic source region of atmospheric Hg, with the annual anthropogenic Hg emissions of about 500 – 700 tons (Street et al., 2005; Wu et al., 2006). Long-term monitoring of TGM at remote sites is crucial to assess the regional atmospheric Hg budget. Since the end of last century, a numerous measurements with regard to speciated atmospheric Hg and atmospheric Hg depositions have been conducted in North America and Europe, which benefited many modeling communities and provide useful recommendations to the governments to better protect human heath and ecosystems over the world. However, such kinds of comprehensive measurements of atmospheric Hg and atmospheric Hg depositions were quite limited in China.

Materials and Methods

Five remote sites were selected to monitor atmospehrc Hg species and atmospheric depostions in different areas of China as follows:

(1) Mt. Waliguan Baseline Observatory (100º53′52.7″ E, 36º17′12″ N, 3816 m a.s.l.): which belongs to the World Meteorological Organization's (WMO) Global Atmospheric Watch (GAW) Baseline Stations and is situated at the summit of Mt. Waliguan at the edge of northeastern part of the Qinghai-Xizang (Tibet) Plateau. Speciated atmospehric Hg measuremnts were conducted at this statioin using a Tekran 2537/1130/1135 automated speciation analyzer system and a manual sampling and analysis method since September 2007 (Landis et al., 2001). Wet-only precipition sampling was also started from March 2012.

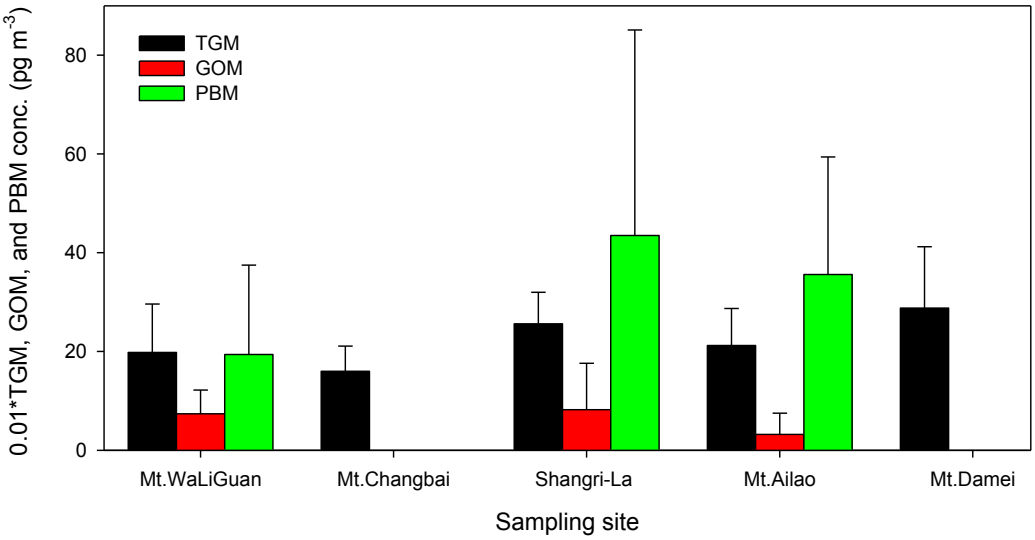

Fig. 1. Mean concentrations of TGM, GOM, and PBM at the five remote sites, China.

(2) Mt. Changbai Station (42°24′0.9″ N, 128°06′45″E, 741 m a.s.l): This station is located in a naturally preserved forest on the north slope of Mt. Changbai, Northeastern China. Atmospheric TGM concentrations were contineously measured using a Tekran 2537 A antomated Hg vapor analyzer from October 2008. Wet-only precipitation sampler were intalled in the station from March 2012.

(3) Shangri-La station (99.733 º E, 28.0167 º N, 3580 m a.s.l.): This station is a regional atmospheric background station in the Southwestern China which was established by the China Meterologocal Administration. Atmospheric TGM concentrations and Hg wet depositions were contineously monitored from November 2009 to Noverber 2010, and GOM and PBM were contineously measured for a duration of two weeks at the interval of very three-month using a manual sampling and analysis technique (Landis et al., 2002).

(4) Mt. Ailao station (101.010 º E, 24.320 º N, 2450 m a.s.l.): This station is situated at one of summits of Ailao Mountain range in the central of Yunan province, Southwestern China. The surrounding areas of the site are naturally preserved forest. Atmospheric TGM concentrations and Hg wet depositions were contineously monitored from May 2011, and GOM and PBM were contineously measured for a duration of two weeks at the interval of very three-month using a manual sampling and analysis technique (Landis et al., 2002).

(5) Mt. Damei Station (121.565 º E, 29.632 º N, 550 m): This station is situated at one of the summits of Damei Mountain in the coastal areas of Yangtz River delta, which is one of the most polulated and industrilized regions of China. Atmospheric TGM concentrations and Hg wet depositions were contineously monitored from April 2011.

For the five remote sites, Mt. Waliguan, Mt. Changbai, Shangri-La and Mt. Ailao stations were involved in the Global Mercury Observation System (GMOS) network. Standard Operating Procedures (SOPs) and QA/QC protocols were implemented at all the sampling sites. Data quality of atmospheric TGM concentration was controlled via periodic internal recalibration with a 25 h interval, and the emission rate of internal permeation source was calibrated every 4 months. Precipitation samples were collected using automated wet-only precipitation collectors, and samples colllected were preserved in pre-cleaned Teflon bottles in refrigerators at the temperature of ~ 4ºC. The precipitation samples were rapidly brought to lab for THg analysis. THg concentrations in precipitation were determined following US EPA Method 1631 (USEPA, 1999).

Results and Discussion

Averaged TGM concentration in ambient air at Mt. Waliguan, Mt. Changbai, Shangri-La, Mt. Ailao, and Mt. Damei stations were 1.98 ± 0.98 ng m^{-3}, 1.60 ± 0.51 ng m^{-3}, 2.56 ± 0.64 ng m^{-3}, 2.12 ± 0.75 ng m^{-3}, and 2.88 ± 1.24 ng m^{-3}, repectivley. These levels were slightly lower than those observed at Mt. Gongga (3.98± 1.62 ng m^{-3}, Fu et al., 2008a) and Mt. Changbai (3.58± 1.78 ng m^{-3}, Wan et al., 2009), which were impacted by strong regional anthropogenic Hg sources and local emisisoins in the vicinity of the sites. On the other hand, the mean TGM concentrations in this study were consistent with the results obatained from remote sites of Mt. Leigong, Southern China (2.80 ± 1.51 ng m^{-3}, Fu et al., 2010) and Chengshantou, Eastern China (2.31 ± 0.74 ng m^{-3}, Ci et al., 2011). On the other hand, TGM concentrations from remote areas of China were relativley higher than those obtained from remote sites in North America and Euope (1.5 – 2.0 ng m^{-3}, Lindberg et al., 2007). These observations indicates that China likely has relatively higher anthropogenic Hg emission intensity compared to North America and Europe, which is consistent with the

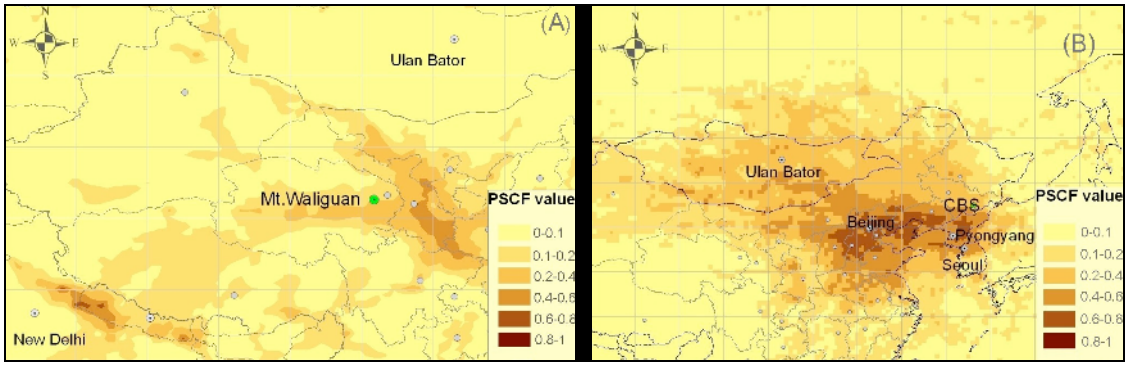

Fig. 2. Likely source regions of atmospheric TGM at Mt. Waliguan and Mt. Changbai (CBS) stations.

global anthropogenic Hg inventroies (Pacyna et al., 2010; Pirrone et al., 2010).

Spatial distribution patern of atmospheric TGM concentratins were observed at the studied sites (Fig. 1). Sites in Eastern China and Southwestern China, which were generally impacted by more regional anthropogenic Hg sources (Street et al., 2005), showed relativley higher levels of TGM compared to sites in Northeastern and Northwestern China.

These contineous observations also helps to gain a better insight to the effects of long-range atmospheric mercury transport on TGM variations in ambient air. Fig. 2 shows the possible source regions and pathways of atmospheric TGM at Mt. Waliguan and Mt. Chang stations identified by the PSCF analysis (Fu et al., 2012a,b). The major source regions of Mt. Waliguan were located to the east of the site, which includes western Qinghai, southern Gansu, and western Shaanxi. These areas are normally characterized as the most populated and industrial regions in Western China. Northwestern India was also an important source region of Northeastern Qinghai-Tibetan Plateau (Fig.2A), indicating this area might also have relatively higher anthropogenic Hg emissions. For the Mt. Changbai station, long-range air masses originated from Northern China as well as Southern North Korea likely contributed significantly to elevated levels of TGM at the monitoring site. There are normally many large coal fired power plants in these two indentified areas, indicating coal burning is an important source of atmospheric Hg.

Mean GOM and PBM concentrations were7.4 ± 4.8 pg m^{-3} and 19.4 ± 18.1 ng m^{-3} at Mt. Waliguan station, 8.2 ± 9.4 pg m^{-3} and 43.5 ± 41.6 ng m^{-3} at Mt. Shangri-La station, and 3.2 ± 4.3 pg m^{-3} and 35.6 ± 23.8 ng m^{-3} at Mt. Ailao station, respectively. In general, PBM concentrations in remote and urban areas of China showed much higher PBM concentration than GOM (Fu et al, 2008b, 2011, 2012c), and this is contrast with many observations conducted at remote sites in North America and Europe (Valente et al., 2007; Engle et al., 2010). This indicates China many have different anthropogenic Hg emission sectors with North America and Europe (Pacyna et al., 2010). This might be also caused by the serious pollution with repect to fine particulate matters with diameter < 2.5 μm (PM2.5) in China which results in more gas-to-partilce production of PBM (von Donkelaar et al., 2010).

Preliminary results on precipitation samples showed mean total mercury (THg) concentrations of 18.0 ± 41.2 ng L^{-1} and 2.7 ± 1.8 ng L^{-1} at Shangri-La station and Mt. Ailao station, respectively. Elevated THg concentration in rain samples at Shangri-La was mainly due to the fact that there was few rain events during the sampling campaign, and this led to elevated total particluated bounded mercury concentration in the atmospheric column which concequently entered into precipitaitons via rain scavenging processes. The THg concentration in precipitaitons at Mt. Ailao is comparable to that observed at the summit of Mt. Leigong (4.0 ng L^{-1}, Fu et al., 2010), indicating THg concentrations in precipitation at mountain tops are normally lower than other sites located in lower-altitude areas. It is speculated that GOM and PBM in atmospheric column may significantly increase precipitaiton THg concentrations during scavenging processes. This is different from sites located at the summit of mountain, where THg in precipitation was mainly originated from clouds in the free troposphere.

Conclusion

This study presents preliminary results of measurements of atmospheric Hg species and depositions at remote sites of China. The speciated atmospheric Hg concentrations observed were relatively higher than North America and Europe. Spatial distribution patterns with elevated TGM concentrations in Eastern and Southwestern China were observed. The levels and spatial pattern were consistent with anthropogenic Hg emission inventories and modeling studies, indicating great anthropogenic Hg emissions in China.

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