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Shipboard and ground measurements of atmospheric particulate mercury and total mercury in precipitation over the Yellow Sea region☆

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ABSTRACT

The first ever shipboard measurements for atmospheric particulate mercury (Hg(p)) over the Yellow Sea and ground measurements for atmospheric Hg(p) and total mercury (THg) in precipitation at the remote sites (Deokjeok and Chengshantou) and the urban sites (Seoul and Ningbo) surrounding the Yellow Sea were carried out during 2007–2008. The Hg(p) regional background concentration of 56.3 ± 55.6 pg m^{-3} over the Yellow Sea region is much higher than the typical background concentrations of Hg(p) in terrestrial environments (<25 pg m^{-3}) which implies significant impact of anthropogenic mercury emission sources from East Asia. The episodes of highly elevated Hg(p) concentrations at the Korean remote site were influenced through long-range transport from source regions in the Liaoning Province - one of China's most mercury-polluted regions and in the western region of North Korea. Interestingly, wet scavenging of atmospheric Hg(p) is the predominant mechanism regulating concentration of THg in precipitation at the Chinese sites; whereas, wet scavenging of gaseous oxidized mercury (GOM) might play the more important role than that of Hg(p) at the Korean sites. The highest annual wet and dry deposition fluxes of Hg were found at the Ningbo site. The comparison between wet and dry deposition fluxes suggested that dry deposition might play the more important role than wet deposition in Chinese urban areas (source regions); whereas, wet deposition is more important in Korean areas (downwind regions).

1. Introduction

Atmospheric mercury typically includes three species: gaseous elemental mercury (GEM), gaseous oxidized mercury (GOM, a.k.a. RGM), and particulate mercury (Hg(p)) (Castro et al., 2012). Hg(p) consists of gaseous mercury species bound or adsorbed to atmospheric particles. These mercury species are characterized with the differences with respect to their atmospheric lifetime, transport, physical and chemical transformations, deposition, and influence on ecosystems (Lin and Pehkonen, 1999). Given the potential toxicity and bio-magnification of mercury, studies on its existence in the environment is essential (Poissant and Pilote, 1998). Dry and wet depositions have been both identified as principal pathways for scavenging of atmospheric mercury (Lin and Pehkonen, 1999). With respect to dry deposition, Hg(p) and GOM in ambient air can deposit and diffuse into both aquatic and terrestrial ecosystems. As GEM accounts for more than 95% of total Hg in the atmosphere, it could also play an important role in contributing to dry deposition of Hg over vegetated surfaces (Zhang et al., 2009). Wet deposition of atmospheric mercury also involves the collection of both particulate and gaseous phases. Despite a small portion (<5%) of the total atmospheric mercury, Hg(p) and GOM both play a significant role in wet deposition of atmospheric mercury because they are

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effectively scavenged by precipitation (Lin and Pehkonen, 1999). Whereas, GEM must first be oxidized to more soluble oxidation state before being efficiently scavenged by precipitation (Poissant and Pilote, 1998; Sakata and Asakura, 2007). Mercury wet deposition could be affected by several factors including precipitation, atmospheric oxidant concentration and mercury speciation, chemical/physical transformation, wet scavenging of atmospheric mercury species, in-cloud processes, and concentration of total mercury (THg) in precipitation (Lai et al., 2007; Xu et al., 2014). THg in precipitation could result from the wet scavenging of atmospheric Hg emitted by local and/or regional sources (Xu et al., 2014; Wang et al., 2014), and formation through in-cloud processes (Huang et al., 2015).

East Asia is considered as the largest source region contributing approximately 50% of global anthropogenic mercury emissions (UNEP, 2013). China has been regarded as the largest single emitter, contributing about 50% and 30% of Asian and global emissions, respectively (UNEP, 2013). During the last decade, coal combustion and non-ferrous metals smelting have accounted for 80% of total national mercury emissions and are expected to continue to be the two largest sources of China's mercury emissions (Wu et al., 2006). Based on AMAP/UNEP (2008), it is calculated that the total mercury emission amount in China was approximately 26 times higher than that in Korea in 2005. Due to the large difference in the emission amounts, dry and wet deposition mechanisms of atmospheric Hg species may be different between Korea and China.

The Yellow Sea could be one of the most polluted sea areas in the world since it is adjacent to the coast of eastern China with a lot of industrial activities. As westerly wind dominates most of seasons (except summer), the Yellow Sea region might be impacted by various Hg species emitted from highly polluted areas in China. Among various atmospheric Hg species, this study focuses on Hg(P) because the dry deposition velocity of GEM is typically low over water bodies (less than 0.03 cm s^{-1}; Fu et al., 2010) and the increase of Hg(P) and GEM concentrations, rather than GOM, are reported to be significant during long-range transport in Seoul, Korea (Kim et al., 2009). Although, it is important to study the long-range transport of Hg(p) in the Yellow Sea region, to our knowledge, no data on the Hg(p) levels over the Yellow Sea region has been reported.

The first ever shipboard measurements of atmospheric Hg(p) over the Yellow Sea were carried out for the periods in fall and spring of 2007 and 2008 in this study. Ground measurements at the two Korean sites and two Chinese sites located in the Yellow Sea region were also performed during 2007–2008 to evaluate the spatial and temporal variations of atmospheric Hg(p) and THg in precipitation in this region. This study aimed to assess the concentration levels and seasonal variations of atmospheric Hg(p) and THg in precipitation over the Yellow Sea region; evaluate the influence of long-range transport to Hg(p) measured at the remote site in Korea; and estimate and compare the Hg wet and dry depositions between Korea and China.

2. Materials and methods

2.1. Shipboard measurements

The Yellow Sea is an inlet of the Pacific Ocean lying between mainland China on the west and north, and the Korean Peninsula on the east. Atmospheric Hg(p) levels over the Yellow Sea were obtained using an on-board ship measurement system. Measurements were carried out for three sea routes including Incheon-Jeju (10–14 Sep 2007), Incheon-Qingdao (16–20 Oct 2007), Incheon-Weihai (28 Apr-01 May and 13–16 Oct 2008), as depicted in Fig. 1. Incheon is Korea's third most populous city located in

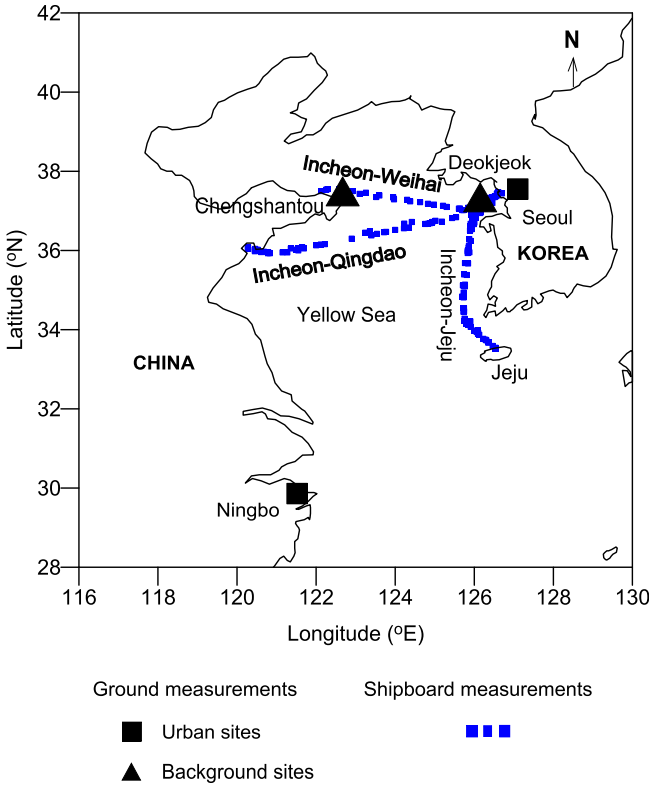

Fig. 1. Ground and shipboard measurements of atmospheric Hg(p) and THg in precipitation. Deokjeok and Chengshantou are remote sites; Seoul and Ningbo are urban sites. Three routes represent the shipboard measurements over the Yellow Sea: Incheon-Weihai, Incheon-Qingdao, and Incheon-Jeju.

midwestern Korea; Jeju is Korea's largest island; Qingdao is a major seaport and industrial center in eastern Shandong province on the eastern coast of China; and Weihai is a major seaport in eastern Shandong province. During each cruise, total suspended particle (TSP) samples were daily collected. Each sampling was performed for about 8–10 h. The sampling was started 1 h after the departure from each seaport and stopped 1 h before the arrival. The measurement instruments were placed at the front of the top deck of the ship with a vertical distance of approximate 20 m from the surface of the sea. To minimize the effect of ship emissions as much as possible, an automatic control system was used to stop sampling when wind comes from the stern.

2.2. Ground measurements

The collections of atmospheric particulate and precipitation samples were conducted at the two Korean sites (Seoul and Deokjeok) and the two Chinese sites (Ningbo and Chengshantou) during 2007–2008 (Fig. 1). Seoul, with a population of more than 10 million, is the largest metropolis and the capital of Korea. The major anthropogenic source of mercury emissions in Seoul is waste incinerators (Pan et al., 2008). The Seoul site was located in the residential areas in the northeast of Seoul. The Deokjeok site was located in the mountain area of Deokjeok island in Gyeonggi, Korea with area of 20 km^2 and population of approximate 1000 people. There are no major anthropogenic emission sources surrounding the Deokjeok site. The Ningbo site was located at the Institute of Environment Protection Science Research and Design in the downtown district of Haishu in Ningbo city - a sub-provincial city in northeast Zhejiang province, China. There are significant anthropogenic sources nearby Haishu District including a waste