Due to the fast developing economy, mercury (Hg) emissions to the atmosphere from Chinese mainland have increased rapidly in recent years. Consequently, this issue has received a considerable attention internationally. This paper reviews the current understanding of and knowledge on atmospheric Hg emissions, distribution and transport in China. The magnitude of Hg emissions to the atmosphere from Chinese anthropogenic sources has been estimated to be in the range of 500–700 tons per year, whereby comprising a significant proportion of the globe total anthropogenic emissions. Emissions of Hg from natural surfaces including bare soil, water, and vegetation covered soil tend in a comparison to be higher in China than in Europe and North America, indicating the importance of this source category. Atmospheric Hg exhibits a significant concentration variability among urban, semi-remote, and remote areas. Total Gaseous Mercury (TGM) concentrations in urban areas of China were often 1.5–5 folds higher compared to the corresponding settings in North America and Europe. In turn, particulate mercury (PHg) concentrations in urban areas of China were up to two orders of magnitude higher compared to North America and Europe. Atmospheric observations made at strictly remote sites in China also include the presence of occasional high concentrations of TGM, and the more short-lived fractions PHg and Reactive Gaseous Mercury (RGM). Accordingly, Hg deposition fluxes tended to be higher in China, with remote areas and urban areas being 1–2 times and 1–2 magnitude higher than those in North America and Europe, respectively.
wildlife. Unlike other heavy metals pollutants that tend to exist in the atmosphere as the particulate phase, Hg exists mainly in the gaseous phase (Wängberg et al., 2001; Poissant et al., 2005; Gabriel et al., 2005; Aspmo et al., 2006; Valente et al., 2007; Chand et al., 2008). In current speciation/fractionation schemes, atmospheric Hg can be divided into gaseous elemental Hg (GEM), reactive gaseous Hg (RGM), and particulate Hg (Phg) (Schroeder and Munthe, 1998). RGM and to some extent Phg have a high surface reactivity and water solubility and are readily scavenged from atmosphere via wet and dry deposition. However, GEM, the predominant form of atmospheric Hg (>90% of the total Hg in atmosphere), is fairly stable in the lower atmosphere with a residence time of several months to a year, with the exception of certain situations including polar regions, upper troposphere, and marine boundary layer (Schroeder and Munthe, 1998; Lu et al., 2001; Lindberg et al., 2007; Faïn et al., 2009; Obrist et al., 2010). This enables Hg⁰ to undergo long-range transport and eventually become well-mixed on a hemispherical scale.

China is the largest developing country worldwide. Due to the fast developing economy, China has emerged as the world’s biggest energy consumer. For example, coal combustion in China claims more than 25% of the total worldwide coal production. Combustion of fossil fuels represents the largest single anthropogenic source of Hg to the atmosphere (Pacyna et al., 2006). Moreover, other industrial activities including non-ferrous metal smelting and cement production prevalent in China also release large amounts of Hg to the atmosphere. Hence, China is considered to be the largest anthropogenic Hg source region over the world. Recently, significant efforts have been made to study the emissions of atmospheric Hg and its distribution in China. In order to provide a better understanding of current knowledge with respect to atmospheric Hg in China and promote future studies, this paper provides an overview of current atmospheric Hg studies in China.

2. Source of atmospheric Hg in China

2.1. Anthropogenic sources

China is the largest emitter of atmospheric Hg worldwide. The anthropogenic Hg emission from China was estimated to be at about 535 tons in 1999 (Street et al., 2005). Due to the increased energy consumption during the past decade, anthropogenic Hg emissions has rapidly increased with predicted amount of 695 tons for the year 2003, which is about 26% higher than that in 1995 and corresponds to an average annual increase of 3.0% (Wu et al., 2006). Anthropogenic Hg sources are irregularly spatially distributed over China. As can be seen in Fig. 1, the emissions from provinces of Guangdong, and Guizhou are the highest. The high anthropogenic Hg emissions of Liaoning and Guangdong were linked to non-ferrous smelters, while high Hg content of raw coal and relatively large amount of uncontrolled coal combustion affect the emissions from Guizhou (Street et al., 2005).

Coal combustion is the most important anthropogenic source of Hg in China. According to the studies by Wu et al. (2006) and Pirrone et al. (2010), coal combustion in China released approximately 256–268 tons of Hg to the atmosphere in 2003, accounting for about 40% of the total anthropogenic emissions in China. Hg emission factors for coal combustion show a large span in China from 1.5 to 350 mg ton⁻¹ (Tang et al., 2007; Zhang et al., 2008; Wang et al., 2010), which was comparable with the global mean estimates (40–300 mg ton⁻¹, Street et al., 2009; Pacyna et al., 2010). Coal type and coal Hg content are important factors affecting Hg emission and its speciation from coal combustion. Anthracite coal tends to have a higher concentration of Hg than bituminous coal and is therefore suggested to release more Hg during combustion (Wang et al., 2010). Moreover, the Hg emission factor from coal combustion is evidently influenced by the implementation of air pollution control devices. Wang et al. (2010) investigated Hg emission factors from six typical coal-fired power plants of China and found the average Hg removal efficiency was at 73% in the plants including a combination of electrostatic precipitators (ESP) and wet flue gas desulfurization (FGD). In addition, Tang et al. (2007) reported that Hg emission factors for small industrial coal-fired boilers decreased by about 40% when wet FGD systems were in operation.

Non-ferrous metal smelting also constitutes an important anthropogenic Hg source category in China. This sector was estimated to release about 230 tons of Hg to the atmosphere in 1995, which was revised up to 320 tons for 2003 (Wu et al., 2006). In the global perspective, the non-ferrous metal smelting in China makes up a much larger portion of the domestic anthropogenic Hg emissions than most other countries. However, it should be noted that the estimates of Hg emission from non-ferrous smelting in China are associated with significant uncertainties due to various smelting methods and pollution-control techniques applied. Among the non-ferrous metals, the contribution from zinc smelting emerges as the largest single source (Street et al., 2005; Wu et al., 2006). Li et al. (2010a) estimated Hg emission factors for industrial-scale zinc smelting plants in China to be in the range 5.7–122 g ton⁻¹ (mean = 33.1 g ton⁻¹) using the mass balance method. Of the several plants investigated, the Hg emission factors related to artisanal and imperial smelting process operations were found to be the highest. On the basis of the calculated emission factors, Li et al. (2010a) estimated that zinc smelting released 80–104 tons of Hg annually to the atmosphere during the period 2002 to 2006, which is much lower than the estimates obtained from previous studies (Street et al., 2005; Wu et al., 2006). Again, the employment of air pollution control devices will significantly affect Hg emissions from zinc smelting. Wang et al. (2010) measured Hg concentrations of flue gas in a hydrometallurgical zinc smelter and found that most of emitted Hg was removed by air pollution control devices. The emission factor for this hydrometallurgical zinc smelter was as low as 0.5 g Hg ton⁻¹ zinc.

Cement production, Hg mining, biofuel combustion, and household waste burning constitute in China comparatively minor sources of atmospheric Hg, which combined accounted for the release of more than 80 tons of Hg to the atmosphere in 2003 (Wu et al., 2006).

2.2. Emissions of Hg from natural surfaces

Table 1 summarizes measurement studies of Hg air–natural surface exchange performed in typical landscapes of China. Some
conclusions can be drawn on the basis of the cited studies: (a) Exchange fluxes of Hg between atmosphere and landscapes are bi-directional. Nevertheless, natural surfaces predominantly act as net sources of Hg to the atmosphere. Deposition fluxes occur more frequently during cold seasons with low air temperature, low solar radiation, and elevated atmospheric Hg\(_0\) concentrations (Feng et al., 2005; Wang et al., 2007a). (b) In general, the magnitude of Hg flux from natural surfaces of China is significantly higher compared to the observations made in North America and Europe. Average Hg fluxes from background bare soils in the United States were reported in the range of 0.1–2.7 ng m\(^{-2}\) h\(^{-1}\) (Erickson et al., 2006). In Canada, emission fluxes of Hg from aquatic environments and forest soils were in the range of 0.3–5.6 ng m\(^{-2}\) h\(^{-1}\), and forest soils in the range of 0.0–0.2 ng m\(^{-2}\) h\(^{-1}\) (Ci et al., 2011a).

Hg emissions to air from natural surfaces have been considered in some regional atmospheric Hg budget studies of China. Feng et al. (2005) estimated that Hg emission from soil in Guiyang made up ~64% of the anthropogenic Hg emissions in the city. Wang et al. (2006a) measured Hg emission fluxes from bare soil, forest soil, grassland, and water surfaces in Chongqing city in southwestern China. On the basis of the observations, annual emission of Hg from natural surfaces in Chongqing was estimated to be 1.8 tons (Wang et al., 2006a). Shetty et al. (2008) developed a chemical–physical model coupled with detailed GIS data and satellite LAI;

### Table 1

<table>
<thead>
<tr>
<th>Location</th>
<th>Classification</th>
<th>Type</th>
<th>Time</th>
<th>Flux (ng m(^{-2}) h(^{-1}))</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Lanmuchang, Guizhou</td>
<td>Mining area,</td>
<td>Bare soil</td>
<td>Dec 2002</td>
<td>482–919</td>
<td>Wang et al., 2005</td>
</tr>
<tr>
<td>Wuchuan, Guizhou</td>
<td>Mining area,</td>
<td>Bare soil</td>
<td>May 2003</td>
<td>247–2283</td>
<td></td>
</tr>
<tr>
<td>Wanshan, Guizhou</td>
<td>Mining area,</td>
<td>Bare soil</td>
<td>Nov 2002</td>
<td>−550–140</td>
<td>Wang et al., 2007a</td>
</tr>
<tr>
<td>Guiyang, Guizhou</td>
<td>Urban area</td>
<td>Bare soil</td>
<td>May and Jun 2003</td>
<td>0.4–44.4</td>
<td>Feng et al., 2005</td>
</tr>
<tr>
<td>Chongqing</td>
<td>Urban area</td>
<td>Forest soil</td>
<td>Jan, Aug, and Sep 2003</td>
<td>3.4–8.4</td>
<td>Fu et al., 2007</td>
</tr>
<tr>
<td>Changchun</td>
<td>Urban area</td>
<td>Bare soil</td>
<td>Apr–Jun 2005</td>
<td>7.8–41.5</td>
<td>Wang et al., 2006a</td>
</tr>
<tr>
<td>Hongfeng lake, Guizhou</td>
<td>Semi-remote</td>
<td>Bare soil</td>
<td>Jul 2002</td>
<td>5.8–21.5</td>
<td>Feng et al., 2003</td>
</tr>
<tr>
<td>Mt. Gongga, Sichuan</td>
<td>Remote</td>
<td>Bare soil</td>
<td>Sep 2005</td>
<td>3.3–7.6</td>
<td></td>
</tr>
<tr>
<td>Longtanzi Reservoir, Chongqing</td>
<td>Urban area</td>
<td>Water</td>
<td>Oct 2003; Nov 2003</td>
<td>5.9–7.5</td>
<td>Wang et al., 2006a</td>
</tr>
<tr>
<td>Hongfeng lake, Guizhou</td>
<td>Semi-remote</td>
<td>Water</td>
<td>Jan 2003; March 2003; March 2004; Nov 2004</td>
<td>1.5–20.1</td>
<td>Feng et al., 2008</td>
</tr>
<tr>
<td>Baihua lake, Guizhou</td>
<td>Semi-remote</td>
<td>Water</td>
<td>May 2002; March–Apr; 2003, April 2006</td>
<td>1.5–20.1</td>
<td>Feng et al., 2004a</td>
</tr>
<tr>
<td>WJD Reservoir, Guizhou</td>
<td>Remote</td>
<td>Water</td>
<td>May 2006</td>
<td>3.3–14</td>
<td>Fu et al., 2010a</td>
</tr>
<tr>
<td>SFY Reservoir, Guizhou</td>
<td>Remote</td>
<td>Water</td>
<td>Jan 2007</td>
<td>6.5–14</td>
<td>Fu et al., 2010b</td>
</tr>
<tr>
<td>South China Sea</td>
<td>Remote</td>
<td>Sea water</td>
<td>Aug 2007</td>
<td>4.5±3.4</td>
<td>Ci et al., 2011a</td>
</tr>
<tr>
<td>Yellow Sea</td>
<td>Remote</td>
<td>Sea water</td>
<td>Jul 2010</td>
<td>18.3±11.8</td>
<td></td>
</tr>
</tbody>
</table>

### Fig. 2

Relationship analysis between the logarithm of Hg flux versus the logarithm of substrate Hg concentration for field chamber measurements. Data were from Lanmuchang (Wang et al., 2005); Wushan (Wang et al., 2007a); Guiyang (Feng et al., 2005), Qingshen (Feng et al., 1996), and Hongfenghui (Wang et al., 2004) in Guizhou province and Mt. Gongga (Fu et al., 2008a) in Sichuan province.
products to estimate natural emissions of Hg in China and Asia. It was estimated that the total Hg emissions from natural surfaces of China was 462 tons in 2001, which comprise nearly 80% of the total anthropogenic Hg emissions in China (Wu et al., 2006).

3. Distribution of atmospheric Hg species in China

3.1. Distribution of atmospheric Hg in urban and industrial areas

Atmospheric Hg concentrations showed a clear spatial distribution pattern in China (Table 2). TGM concentrations reported for urban and industrial areas of China were in the range of 2.7–35 ng m$^{-3}$, which were about 1.5 – 7 folds higher than the values reported from North America and Europe (Carpi and Chen, 2002; Lynam and Keeler, 2005; Stamenkovic et al., 2007). For urban areas of China, relatively low TGM concentrations were observed in the coastal cities of Shanghai and Ningbo (2.7 ± 1.7 and 3.79 ± 1.29 ng m$^{-3}$ respectively), which may correspond to the influence of relatively clean marine air masses. On the other hand, elevated TGM concentrations were observed at some inland cities, which generally are co-located with large point sources (e.g. coal-fired power plants, non-ferrous metal smelting, etc.) and residential coal burning (Liu et al., 2002; Fu et al., 2009; Fu et al., 2011). In general, TGM concentrations in urban and industrial areas of China are higher than those reported for adjacent Asian countries. Kim et al. (2009) reported that the mean TGM, PHg, and RGM concentrations in Seoul, Korea were 3.22±2.10 ng m$^{-3}$, which was about 0.4% of the observed TGM (Fu et al., 2011a). Highly elevated levels of speciated atmospheric Hg in urban and industrial areas in China indicate that they are important source regions of atmospheric Hg. Generally, urban areas of China are densely populated, and there is a huge demand for coal consumption which is a large source of atmospheric Hg. For example, the coal consumption in Guiyang in 2003 was 9,230,000 tons, which was predicted to release about 817 kg of GEM, 813 kg of RGM, and 267 kg of PHg to the atmosphere (Tang et al., 2007).

Table 2
Statistic summary of atmospheric speciated mercury concentrations in China.

<table>
<thead>
<tr>
<th>Locations</th>
<th>Classification</th>
<th>Time</th>
<th>Method</th>
<th>TGM (ng m$^{-3}$)</th>
<th>PHg (pg m$^{-3}$)</th>
<th>RGM (pg m$^{-3}$)</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Guiyang</td>
<td>Urban</td>
<td>Nov 2001 to Nov 2002</td>
<td>Tekran 2537</td>
<td>8.40 ± 4.87</td>
<td></td>
<td></td>
<td>Feng et al., 2004b</td>
</tr>
<tr>
<td>Beijing</td>
<td>Urban</td>
<td>TGM: Jan, Feb, and Sep in 1998</td>
<td>TGM: Tekran 2537; 1130, and 1135</td>
<td>9.7 ± 10.2</td>
<td>3.36 ± 676*</td>
<td>35.7 ± 43.9</td>
<td>Fu et al., 2006b</td>
</tr>
<tr>
<td></td>
<td>Suburban</td>
<td>Tekran 2537</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Remote</td>
<td>PHg: Jan–Dec 2003</td>
<td>PHg: Manual sampling and acid digestion</td>
<td>Tekran 2537</td>
<td>7.9–345</td>
<td>1180 ± 820</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>5.3–12.4</td>
<td>680 ± 620</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>2.5–5</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Shanghai</td>
<td>Urban</td>
<td>March 2002 to Sep 2003</td>
<td>Tekran 2537</td>
<td>2.7 ± 1.7</td>
<td>3.79 ± 1.29</td>
<td>6.74 ± 0.37</td>
<td>Xiu et al., 2009</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Aug–Sep 2009</td>
<td>Manual sampling and acid digestion</td>
<td></td>
<td></td>
<td></td>
<td>Friedli et al., 2011</td>
</tr>
<tr>
<td></td>
<td>Beijing</td>
<td>Oct 2007 and Jan 2008</td>
<td>Lumex RA-915 +</td>
<td></td>
<td>3.36 ± 676*</td>
<td>35.7 ± 43.9</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Changchun</td>
<td>Aug 2006 to Sep 2007</td>
<td>Lumex RA-915 +</td>
<td></td>
<td></td>
<td></td>
<td>Nguyen et al., 2011</td>
</tr>
<tr>
<td></td>
<td>Urban</td>
<td>Jul 1999 to Jan 2000</td>
<td>Manual sampling and thermal desorption</td>
<td>Tekran 2537</td>
<td>2.7 ± 1.7</td>
<td>3.79 ± 1.29</td>
<td>Yang et al., 2009</td>
</tr>
<tr>
<td></td>
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<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Su et al., 2007</td>
</tr>
<tr>
<td>Wuhan</td>
<td>Urban</td>
<td>–</td>
<td>Manual sampling and thermal desorption</td>
<td>Tekran 2537</td>
<td>18.4</td>
<td>276</td>
<td>Fang et al., 2004</td>
</tr>
<tr>
<td></td>
<td></td>
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<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Mt. Gongga</td>
<td>Urban</td>
<td>May–Jun 2007</td>
<td>Tecran 2537</td>
<td>7.7–20.1</td>
<td></td>
<td></td>
<td>Fu et al., 2009</td>
</tr>
<tr>
<td></td>
<td>Chengdu</td>
<td>Jul 2007 to May 2009</td>
<td>Lumex RA-915 +</td>
<td>2.21 ± 0.74</td>
<td></td>
<td></td>
<td>Ci et al., 2011b</td>
</tr>
<tr>
<td></td>
<td>Pearl River Delta</td>
<td>Nov–Dec 2007</td>
<td>Tecran 2537</td>
<td>2.94 ± 2.02</td>
<td></td>
<td></td>
<td>Li et al., 2011</td>
</tr>
<tr>
<td></td>
<td>Mt. Changbai</td>
<td>Aug 2005 to Jul 200</td>
<td>Tecran 2537, 1130, and 1135</td>
<td>3.58 ± 1.78</td>
<td>77 ± 136*</td>
<td>65 ± 84</td>
<td>Wang et al., 2009a, b</td>
</tr>
<tr>
<td></td>
<td>Mt. Changbai</td>
<td>Oct 2008 to Oct 2010</td>
<td>Tecran 2537</td>
<td>1.60 ± 0.51</td>
<td></td>
<td></td>
<td>Fu et al., in preparation</td>
</tr>
<tr>
<td></td>
<td>Mt. Gongga</td>
<td>May 2005 to July 2007</td>
<td>Phg and RGM: Manual sampling and thermal desorption</td>
<td>Tekran 2537</td>
<td>3.98 ± 1.62</td>
<td>30.7 ± 32.1</td>
<td>Fu et al., 2008b, c</td>
</tr>
<tr>
<td></td>
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<td></td>
</tr>
<tr>
<td>Mt. Leigong</td>
<td>Remote</td>
<td>May 2008 to May 2009</td>
<td>Tecran 2537</td>
<td>2.80 ± 1.51</td>
<td></td>
<td></td>
<td>Fu et al., 2010c</td>
</tr>
<tr>
<td>Mt. Wulingan</td>
<td>Remote</td>
<td>Sep 2007 to Aug 2008</td>
<td>Phg and RGM: Manual sampling and thermal desorption</td>
<td>Tekran 2537</td>
<td>1.98 ± 0.98</td>
<td>19.4 ± 18.1*</td>
<td>Fu et al., 2011b</td>
</tr>
<tr>
<td>Shangri-La</td>
<td>Remote</td>
<td>Nov 2009 to Nov 2010</td>
<td>Tecran 2537</td>
<td>2.59 ± 1.33</td>
<td>43.5 ± 41.6*</td>
<td>8.2 ± 9.4</td>
<td>Zhang, 2011</td>
</tr>
<tr>
<td>South China Sea</td>
<td>Remote</td>
<td>Aug 2007</td>
<td>Lumex RA-915 +</td>
<td>2.62 ± 1.13</td>
<td></td>
<td></td>
<td>Fu et al., 2010b</td>
</tr>
<tr>
<td>Yellow Sea</td>
<td>Remote</td>
<td>Jul 2010</td>
<td>Lumex RA-915 +</td>
<td>2.61 ± 0.50</td>
<td></td>
<td></td>
<td>Ci et al., 2011a</td>
</tr>
</tbody>
</table>

a Fine fraction (< 2.5 μm) PHg.
Some measurements close to large cities and industrial areas exhibited highly elevated atmospheric Hg concentrations. For example, observations of atmospheric TGM and PHg concentrations at a site located 25 km to the southwest of Guiyang city were 6.15 ± 2.95 ng m⁻³ and 660 ± 80 pg m⁻³, respectively (Zheng et al., 2006). Measurements in semi-rural areas near Beijing and Changchun also showed elevated TGM concentrations (Liu et al., 2002; Fang et al., 2004).

### 3.2. Atmospheric Hg distributions in remote areas

Observations of TGM in remote areas of China are generally higher than the background concentration levels of the Northern hemisphere (1.5–1.7 ng m⁻³, Lindberg et al., 2007), with the exception of Mt. Changbai (1.60 ± 0.51 ng m⁻³, Fu et al., in preparation). TGM concentrations measured in Mt. Gongga area (3.98 ± 1.62 ng m⁻³) was significantly higher than those observed at other remote sites in China (Table 2). The elevated TGM level at Mt. Gongga was attributed to emissions of Hg from local domestic coal combustion and regional non-ferrous metal smelting activities. Coal and biofuel burning are prevalent in most areas of China. There is no pollution control for non-ferrous metal smelting activities. Coal and biofuel burning was signified to emissions of Hg from local domestic coal combustion and regional non-ferrous metal smelting activities. Coal and biofuel burning are prevalent in most areas of China.

Atmospheric Hg emissions (>50 tons, Wu et al., 2006) including large mercury mines, which potentially explaining the TGM level observed. In addition, there is an evidence that the site is a frequent receptor for long range transport of atmospheric Hg pollution from central China (E.g. Henan, Hubei, and Hunan provinces).

One year monitoring study of atmospheric speciated mercury has been conducted by (Fu et al., 2011b) at Mt. Waliguan Observatory operated as one out of the 24 baseline Atmospheric Global Watch (GAW) monitoring sites of the World Meteorological Organization (WMO). This high alitudinal station (3816 m a.s.l.) located at the edge of northeastern Qinghai–Tibetan Plateau is relatively isolated from industrial point sources and populated regions. Annual mean TGM, PHg, and RGM concentrations at this site were 1.98 ± 0.98 ng m⁻³, 19.4 ± 18.1 pg m⁻³, and 7.4 ± 4.8 pg m⁻³, respectively (Fu et al., 2011b), which were slightly higher than those reported from remote areas of North America and Europe (Choi et al., 2008a; Sigler et al., 2009; Engle et al., 2010). The speciated Hg concentrations showed a pronounced dependence on wind direction at Mt. Waliguan, with most of the high concentrations observed under north-easterly and easterly flow patterns. Urban areas such as Xining and Lanzhou, provincial capitals of Qinghai and Gansu respectively were the most important regional source areas impacting measurements at Mt. Waliguan. In addition, long-range transport of Hg from eastern Gansu, western Shanxi, western Ningxia province as well as northern India was identified to partially influence the observations (Fu et al., 2011b).

Zhang (2011) have performed long-term measurements of speciated mercury at Shangri-La station, which is a regional baseline observatory belonging to China Meteorological Administration. The site (elevation of 3558 m a.s.l) is located on the edge of southwestern Tibet Plateau in Yunnan province approximate to national borders with Myanmar and India. Annual mean concentrations observed were 2.59 ± 1.33 ng m⁻³, 43.5 ± 41.6 pg m⁻³, and 8.2 ± 9.4 pg m⁻³ for TGM, PHg and RGM respectively. Since there was no direct local sources at the study site, the relatively high PHg concentrations plausibly reflect the influence from regional combustion sources. Regional transport of PHg to the site would be promoted since precipitation events responsible for scavenging were sparse during the sampling campaigns.

TGM in Chinese coastal regions also showed a tendency towards higher concentrations. The short-term measurement of TGM performed at a remote site in Pearl River Delta showed a mean value of 2.94 ± 2.02 ng m⁻³ (Li et al., 2011), being elevated compared to coastal site in North America and Europe (1.4 – 1.72 ng m⁻³, Kock et al., 2005; Mao et al., 2008). This is due to the fact that the Pearl River Delta region is one of the most heavily industrialized regions of China with significant anthropogenic Hg emissions. Atmospheric TGM at Chenshantou, the eastern tip of Shandong peninsula showed a comparatively lower concentration of 2.31 ± 0.74 ng m⁻³ than that in Pearl River Delta (Ci et al., 2011b). This marine site was mainly affected by regional sources of eastern China and long-range transport of pollutants from the Korean Peninsula and Japan.

![Seasonal variation of atmospheric TGM in China. Sites were located at Guiyang, Mt. Changbai, Mt. Gongga, and Mt. Leigong.](image-url)
Atmospheric TGM concentrations in marine boundary layer in the northern South China Sea (SCS) showed a clear trend of decreasing concentrations when going from coastal regions in China towards the open sea (Fu et al., 2010b). TGM concentrations generally ranged from 3.0 to 5.0 ng m\(^{-3}\) at sites located closely to mainland China while at sea far from land a constant level of \(-1.5\) ng m\(^{-3}\) was obtained. The spatial distribution of TGM in SCS basin indicated that the Hg emission from coastal areas in southern China is remarkable (Fu et al., 2010b). Atmospheric TGM concentrations in the marine boundary layer of Yellow Sea showed a comparable mean (2.61 \pm 0.50 ng m\(^{-3}\)) with that observed in the South China Sea (Ci et al., 2011a). The Yellow Sea is surrounded by eastern China and Korea Peninsula, which are important Hg source regions in the East Asia. Most of the high-TGM events were observed close to the mainland of East China, indicating the effect of outflow of air masses with elevated level of atmospheric Hg from the mainland.

3.3. Seasonal variation

Pronounced seasonal variations are often evident in the long-term monitoring of TGM in China. Fig. 3 shows the seasonal cycles of atmospheric TGM at an urban site (Guiyang) and at three remote sites. In general, TGM concentrations at the remote sites showed a similar seasonal cycle with high concentrations in cold seasons and low in warm seasons. A similar seasonal trend in TGM was also observed in Guiyang city with concentration descending in the following order: winter, spring, fall and summer (Feng et al., 2004b). PHg in urban areas exhibits a strong maximum during cold seasons. Wang et al. (2006b) found that the mean PHg concentration during winter in the urban and suburban areas of Beijing was about 4 times higher compared to summer. PHg concentrations in the industrial, residential, and suburban districts of Changchun city were also highly elevated during the winter period (Fan et al., 2001). In contrast, atmospheric RGM concentrations in Mt. Gongga area showed a seasonal variation of high concentrations in spring and autumn and low in winter and summer (Fu et al., 2008c).

Elevated TGM and PHg concentrations in winter are likely related to regional Hg emissions with a strong seasonality. This is particularly true for coal and biofuel consumption increasing during cold seasons owing to the demand for domestic heating. Linear correlation analysis between TGM and atmospheric criteria pollutants found that TGM concentrations were strongly correlated with SO\(_2\) and NO\(_x\) concentrations in winter (Fu et al., 2008b). In addition, decreased oxidation rate of Hg\(_0\) by atmospheric oxidants (e.g., OH\(^-\), Br\(^-\), O\(_3\) etc.), dry deposition flux of Hg\(_0\) to leaf surface, wet scavenging of PHg, and boundary layer depth in winter were also important factors promoting elevated TGM and PHg concentrations during this period of the year (Feng et al., 2004b; Fu et al., 2008b, c, 2010c).

3.4. Comparison between measured data with model predictions

Recently, a number of model studies have attempted to predict the spatial and temporal distributions of atmospheric Hg in China (Bergan et al., 1999; Seigneur et al., 2001, 2004; Li et al., 2007; Selin et al., 2007; Shia et al., 1999; Lin et al., 2010). Accordingly, atmospheric TGM concentrations in China especially in central and eastern China are significantly elevated. However, the predicted TGM concentrations in eastern, central, southern, and southwestern China, generally in the range of 2.0–2.5 ng m\(^{-3}\), tend to be underestimated compared to field observations. This is also the case for model-predicted PHg concentrations in surface air. In contrast, modeled RGM concentration in the atmospheric boundary layer in the range of 100–250 pg m\(^{-3}\) is in general biased high compared to the field observations.

The highly elevated observation results of atmospheric TGM and PHg concentrations in China could be a result of many factors. First of all, many of the previous observations in China were impacted by local and regional emissions, and this might lead to an overestimates of regional backgrounds of atmospheric Hg species. Moreover, there are still a lot of uncertainties related to the Hg emission inventories used in the modeling studies, which might underestimate or miss certain emission sources (Friedli et al., 2004; Jaffe et al., 2005; Li et al., 2007; Weiss-Penzias et al., 2007; Strode et al., 2008; Choi et al., 2009; Lin et al., 2010).

4. Atmospheric Hg deposition

4.1. Atmospheric Hg deposition in urban areas

THg concentrations in precipitation and direct wet deposition fluxes to Chinese urban and suburban areas fall in the range of 55.3–354 ng L\(^{-1}\) and 77.6–152 μg m\(^{-2}\) yr\(^{-1}\), respectively (Table 3), which are much higher than observations made in urban and industrial areas of USA (THg concentrations in precipitation: 10–20 ng L\(^{-1}\); THg wet deposition fluxes: 8–15 μg m\(^{-2}\) yr\(^{-1}\), Guentzel et al., 2001; Landis and Keeler, 1997; Keeler et al., 2006). THg concentrations in precipitation observed in Changchun city, northeastern China are remarkably high. This city is heavily polluted with respect to atmospheric Hg. As shown in sect. 3.2.1, PHg concentrations, one crucial factor controlling wet deposition of Hg (Lee et al.,

Table 3

<table>
<thead>
<tr>
<th>Locations</th>
<th>Time</th>
<th>Classification</th>
<th>Concentration (ng L(^{-1})) or ng g(^{-1})</th>
<th>Flux (μg m(^{-2}) yr(^{-1}))</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Changchun</td>
<td>Jul 1999 to Jul 2000</td>
<td>Urban Precipitation</td>
<td>354</td>
<td>152</td>
<td>Fang et al., 2004</td>
</tr>
<tr>
<td>Chongqing</td>
<td>March 2003 to Feb 2006</td>
<td>Suburban Precipitation</td>
<td>55.3</td>
<td>77.6</td>
<td>Wang et al., 2008</td>
</tr>
<tr>
<td>Luchongguan, Guizhou</td>
<td>Jan to Dec 2005</td>
<td>Suburban Throughfall</td>
<td>105</td>
<td>113</td>
<td>Fu et al., 2008b</td>
</tr>
<tr>
<td>Wujiang, Guizhou</td>
<td>Jan to Dec 2006</td>
<td>Semi-remote Precipitation</td>
<td>8.9</td>
<td>220</td>
<td>Fu et al., 2010c</td>
</tr>
<tr>
<td>Mt.Leigong, Guizhou</td>
<td>May 2008 to May 2009</td>
<td>Remote Throughfall</td>
<td>36.0</td>
<td>19.1</td>
<td>Guo et al., 2008</td>
</tr>
<tr>
<td>Mt.Gongga*, Sichuan</td>
<td>Jan to Dec 2006</td>
<td>Remote Precipitation</td>
<td>9.9</td>
<td>11.1</td>
<td>Fu et al., 2008c</td>
</tr>
<tr>
<td>Mt.Gongga**, Sichuan</td>
<td>May 2005 to Apr 2007</td>
<td>Remote Throughfall</td>
<td>14.3</td>
<td>26.1</td>
<td>Fu et al., 2010d</td>
</tr>
<tr>
<td>Mt.Changbai, Jilin</td>
<td>Aug 2005 to Jul 2006</td>
<td>Remote Precipitation</td>
<td>40.6</td>
<td>57.9</td>
<td>Fu et al., 2010d</td>
</tr>
</tbody>
</table>

* Elevation of the sampling site was 1600 m above sea level.
** Elevation of the sampling site was 3000 m above sea level.
1995, were highly elevated in Changchun, which could partially explain the high precipitation THg concentrations.

4.2. Atmospheric Hg deposition in remote areas

THg concentrations in precipitation and wet deposition fluxes in semi-remote and remote areas of China are significantly lower than in the urban environment and fall in the range of 4.0–36 ng L\(^{-1}\) and 6.0–34.7 ng m\(^{-2}\) yr\(^{-1}\), respectively. In general, concentrations and deposition fluxes of THg in remote and semi-remote areas of China were both higher compared to the results from analogous studies in North America, which are generally less than 10 ng L\(^{-1}\) for THg concentrations in precipitation and 10 ng m\(^{-2}\) yr\(^{-1}\) for wet deposition fluxes, respectively (Graydon et al., 2001; St. Louis et al., 2001; Choi et al., 2008b; Graydon et al., 2009; Lindberg et al., 2007 and references therein).

Studies of MeHg in precipitation have only been carried out in semi-remote and remote areas in China (Table 3). The highest mean MeHg concentration of 0.18 ng L\(^{-1}\) was observed at several semi-remote sites in Wujiang river basin, Guizhou Province (Guo et al., 2008), which was probably impacted by regional anthropogenic activities. The lowest mean MeHg concentration was observed at the summit of Mt. Leigong, which is as mentioned before relatively isolated from anthropogenic activities. In general, source of MeHg in precipitation includes capture of MeHg in atmosphere and oxidation of DMHg to MeHg in both gaseous and aqueous phases in the atmosphere (Sommar et al., 1996; Black et al., 2009). Oxidation of DMHg to MeHg by radical species (e.g. OH\(\cdot\), Cl\(\cdot\)) has been suggested to be a major source of MeHg in precipitation (St. Louis et al., 2007). Terrestrial sources of atmospheric DMHg and MMHg are generally confined to the anthroposphere (sewage sludge, landfills, contaminated sediments, geologically enriched areas, etc.). Several studies have reported highly elevated DMHg concentrations up to several ng m\(^{-2}\) yr\(^{-1}\) in exhaust gas from landfills (Lindberg et al., 2001; Feng et al., 2004c). The MeHg concentrations in atmosphere are generally (much) lower than 20 pg m\(^{-3}\) (Bloom and Fitzgerald, 1988; Brosset and Lord, 1995; Lee et al., 2003). Up to now, there is no evidence for strong terrestrial sources of MeHg to the atmosphere, but diffuse emissions from anthropogenic sources may have a certain impact on local and regional atmospheric MeHg levels. Feng et al. (2004c) reported a mean MeHg concentration of 2.06 ng m\(^{-3}\) in vent gas from a landfill in Guyang, Guizhou province, which is about 2–3 orders of magnitude higher than in ambient air. Due to the fact that landfilling rather than waste incineration is the primary way of disposal of municipal waste (Feng et al., 2004c; Li et al., 2010b) and in addition that flaring or cleaning of landfill gas is normally not implemented, it is likely that the emissions of volatile and semi-volatile organic mercury compounds to air are comparatively high from such facilities in China.

4.3. Estimates of dry depositions

Numerous studies have highlighted the importance of dry deposition of Hg in the biogeochemical cycle of Hg, especially to vegetation covered areas (Graydon et al., 2009; Hartman et al., 2009; Zhang et al., 2009). Several studies in pristine forests of Chongqing, Mt. Gongga and Mt. Leigong revealed that deposition fluxes of THg and MeHg in throughfall and litterfall were profoundly higher than direct wet deposition fluxes. Dry (throughfall + litterfall − wet) deposition fluxes of THg to forest in Chongqing, Mt. Gongga, and Mt. Leigong were estimated to be 256, 66.4, and 44.4 ng m\(^{-2}\) yr\(^{-1}\), respectively (Fu et al., 2010c, 2010d; Wang et al., 2008). The dry deposition fluxes of Hg in forest areas were highly elevated (by the factor of 2.54 − 7.3) compared to the direct wet deposition fluxes. Measured dry deposition fluxes of Hg in China were much higher compared to the measurement data in North America and Europe (Scherbatakov et al., 1998; Lee et al., 2000; Schwesig and Matzner, 2000; St. Louis et al., 2001). This is probably owing to the elevated atmospheric TGM, RGM, and PHg concentrations, which were mainly driven from the large emissions of Hg from anthropogenic activities and natural surfaces in China.

5. Conclusion

China is the largest single emitter of Hg worldwide. Although associated with large uncertainties, the magnitude of atmospheric Hg emissions from human activities in China are in the range of 500–700 tons/y, whereby contributing to about 25–30% of the total global anthropogenic emissions. Although poorly quantified, emissions of Hg from natural sources in China constitute a significant fraction of its total Hg emissions to the atmosphere. This is true especially for GEM emissions. In general, Hg emission fluxes from bare soil, vegetation covered soil, and water surfaces of China are higher in magnitude than fluxes measured in analogous studies in North America and Europe, being partially attributed to the re-emission of anthropogenic “legacy” Hg previously deposited to the Earth surface.

In an international perspective, TGM and PHg concentrations in urban air of Chinese cities turn out to be elevated, often considerably higher compared to the measurement data in Europe and North America. TGM and PHg are frequently both elevated in remote areas of China; whereas RGM concentrations are comparable and/or slightly higher than those observed from North America and Europe. Relatively high levels of Hg in remote areas can be attributed to regional and local emissions, with an additional contribution from long-range atmospheric transport. The magnitude of atmospheric Hg deposition reported in Chinese studies is elevated compared to studies performed in the developed world. Depositions fluxes of Hg in remote areas of China were 1–2 times higher compared to data observed in North America and Europe; whereas in urban areas of China, the depositional load was 1–2 orders of magnitude higher compared to North America and Europe.

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