Atmospheric wet and litterfall mercury deposition at urban and rural sites in China

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Abstract:

Mercury (Hg) concentrations and deposition fluxes in precipitation and litterfall were measured at multiple sites (six rural sites and an urban site) across a broad geographic area in China. The annual deposition fluxes of Hg in precipitation at rural sites and an urban site were 2.0 to 7.2 μg m^{-2} yr^{-1} and 12.6 ± 6.5 μg m^{-2} yr^{-1}, respectively. Wet deposition fluxes of Hg at rural sites showed a clear regional difference with elevated deposition fluxes in the subtropical zone, followed by the temporal zone and arid/semi-arid zone. Precipitation depth is the primary influencing factor causing the variation of wet deposition. Hg fluxes through litterfall ranged from 22.8 to 62.8 μg m^{-2} yr^{-1}, higher than the wet deposition by a factor of 3.9 to 8.7 fluxes and representing approximately 75% of the total Hg deposition at the forest sites in China. This suggests that uptake of atmospheric Hg by foliage is the dominant pathway to remove atmospheric mercury in forest ecosystems in China. Wet deposition fluxes of Hg at rural sites of China were generally lower compared to those in North America and Europe, possibly due to a combination of lower precipitation depth, lower GOM concentrations in the troposphere and the generally lower cloud base heights at most sites that washout a smaller amount of GOM and PBM during precipitation events.

1 Introduction

Mercury (Hg), especially its methylated form, is a potent neurotoxin to humans and wildlife. Because of its high volatility, mild reactivity, and water solubility, gaseous elemental mercury (GEM) is the dominant(> 75%) form of total Hg in the atmosphere (Sprovieri et al., 2010;Gustin et al., 2015;Shah et al., 2016). While the major of remaining fraction of Hg in the atmosphere are gaseous oxidized mercury (GOM) and particulate bound mercury (PBM), which are operationally defined. GEM has an atmospheric residence time of several months to a year and therefore spreads globally before being converted to other forms and deposited to the earth's surface (Holmes et al., 2010;Driscoll et al., 2013;Amos et al., 2015). On the other hand, GOM and PBM have much higher water solubility and reactivity, and are readily deposited via wet and dry deposition pathways.

Quantifying the relationship between atmospheric Hg depositions and emissions is of prime importance in its global biogeochemical cycle. Hg in the atmosphere could be from anthropogenic and natural emission sources. At a global scale, direct and legacy anthropogenic emissions represent the predominant (~2/3) sources of Hg in the atmosphere (Seigneur et al., 2004;Selin et al., 2007;Pirrone et al., 2010), and account for most of the deposition of Hg to the earth's surface (Selin et al., 2008;Amos et al., 2013). Hg is removed from the atmosphere through wet deposition and dry deposition pathways. Atmospheric Hg deposition networks have been established in the North America (NADP, 1994) and Europe (EMEP). These Networks provided critical databases on the spatial and temporal trends in Hg wet deposition and help to constrain regional and global atmospheric Hg deposition budget (Selin et al., 2007;Zhang et al., 2012a). Dry deposition of atmospheric Hg, including uptake of GEM by vegetation and dry deposition of GOM/PBM, is considered to be more important than wet deposition in the total Hg deposition budget (Dastoor and Larocque, 2004;Selin et al., 2007). However, dry deposition of Hg is difficult to be quantified accurately because of the lack of proper measurement techniques. Recently, increasing amount of studies have suggested that measurements of litterfall (uptake of GEM) and throughfall (washout of GOM

and PBM deposited to foliage surfaces) could be a good indicator of Hg dry deposition to forest ecosystems (Johnson and Lindberg, 1995;St Louis et al., 2001;Gustin, 2012). Also, empirical models have been developed to quantify dry deposition of Hg to various landscapes (Lyman et al., 2007;Zhang et al., 2012a). Although subjected to some bias (Zhang et al., 2009;Gustin et al., 2015), these studies significantly improved our understanding of the role of dry deposition in the global Hg biogeochemical cycle.

China is the largest source region of atmospheric Hg in the world. Recent studies revealed that Hg emissions from anthropogenic and natural sources in China were in the range of 800-1200 tons (Shetty et al., 2008;Wang et al., 2014a;Fu et al., 2015a;Wang et al., 2016), approximately 1/6 of the global Hg emissions to the atmosphere (Pirrone et al., 2010;Song et al., 2015). The large emissions in China not only have an impact on the domestic Hg pollutions (Fu et al., 2015b), but also have a potential to enhance atmospheric Hg concentration and deposition elsewhere (Seigneur et al., 2004;Strode et al., 2008;Durnford et al., 2010). To construct the mass budget of Hg in China, measurements of atmospheric Hg wet and dry deposition are essential. However, studies of wet and dry deposition in China are rather limited. Most of previous studies in China were conducted either at a single site using bulk collectors or mostly at urban areas (Guo et al., 2008;Wang et al., 2009;Fu et al., 2010b;Zhou et al., 2013;Wang et al., 2014b;Xu et al., 2014;Zhu et al., 2014), which are not sufficient to depict the overall pattern of atmospheric Hg deposition in China. In the present study, we conducted comprehensive measurements of wet and litterfall deposition fluxes of Hg at multiple sites (six rural sites and one urban site) across a broad geographic area in mainland China. This study aims to better understand the spatial distributions of Hg deposition and mass balance of atmospheric Hg in China. We also compared these fluxes to previous observations in China and other regions worldwide and studied the potential factors influencing wet and litterfall deposition fluxes of Hg.

2 Materials and methods

2.1 Site description

Precipitation samples were collected at six rural sites and one urban site across a broad geographic area in mainland China: Mt. Changbai (MCB, 128.112° E, 42.403° N, 736 m above sea level), Mt. Damei (MDM, 121.565° E, 29.632° N, 550 m above sea level), Mt. Leigong (MLG, 108.203° E, 26.387° N, 2176 m above sea level), Mt. Ailao (MAL, 101.107° E, 24.533° N, 2450 m above sea level), Mt. Waliguan (MWLG, 100.898° E, 36.287° N, 3816 m above sea level), Bayinbuluk (BYBLK, 83.717° E, 42.893° N, 2500 m above sea level), and Guiyang (GY, 106.724° E, 26.573° N, 1041 m above sea level) (Figure 1). Four sites (i.e., MCB, MDM, MLG, and MAL) are rural upland forest sites, two sites (i.e., MWLG and BYBLK) are alpine grassland sites and the remainder (i.e., GY) is an urban site (Table 1).

The MCB site is located in a temperate broadleaf and Korean pine mixed forest on the north slope of Mt. Changbai, northeastern China, which is one of the largest national forest reserves in China and covers an area of ~1,965 km^2. The forest is dominated by tree species of *Pinus koraiensis*, *Fraxinus mandshurica*, *Tilia amurensis*, *Acer mono* and*Quercus mongolica (Dai et al., 2011)*. There are no nearby sources at the MCB site and the predominant wind directions are from southwest to northwest, which could carry air pollutants from regional industrial and urbanized centers and northern China by long-range atmospheric transport (Fu et al., 2012b).

The MDM site is located at the summit of Mt. Damei in eastern Yangtze River Delta, eastern China and about 15 km away from the East China Sea coast. This site is surrounded by a shrub (*Styrax faberi, Lithocarpus glaber, and Indocalamus tessellates*), broadleaf (*Quercus fabri, Liquidambar formosana, and Platycarya Strobilacea*) and conifer (*Pinus massoniana*) mixed forest. The Yangtze River Delta is one of the largest atmospheric Hg source regions in China (Zhang et al., 2015b). There are no large point sources within 10 km of the sampling site. The predominant wind direction during wet season is from east and the site mainly receives clean marine air masses. During dry season, the predominant wind direction changes to from northwest which enhances the transport of air pollutants from northern and eastern China to the sampling site (Yu et al., 2015).

The MLG site is located on top of an isolated peak (elevated approximately 1000 m

115 relative to the surrounding terrain) of the Miaoling mountain range, southwestern China. The
116 sampling site is surrounded by an upland shrub (*Rhododendron*) and deciduous broadleaf
117 mixed forest (*Cinnamomum, Acer Linn., Chaenomeles, Sinarundinaria Nakai*). It is isolated
118 from industrial sources and populated regions and frequently receives free tropospheric air
119 from central China plain regions and the South China Sea (SCS) (Fu et al., 2010a). The MLG
120 site is partly influenced by boundary layer air transported by plain-to-mountain winds from
121 southwestern China during daytime.

122 The MAL site is located at a summit of the northern edge of the Ailao Mountain National
123 Nature Reserve in central Yunnan province, southwestern China. The reserve has an area of
124 677 km^2 and is predominantly (> 80%) covered by evergreen broadleaf primary forests.
125 Canopy species mainly include *Castanopsis wattii*, *Lithocarpus xylocarpus*, *Schima noronhae*,
126 *L. jingdongensis*, and *Hartia sinensi* (Yuan et al., 2009). The MAL site is located 180 km
127 southwest of Kuming City, the capital of Yunnan province and 200 km north of the
128 Indochinese Peninsula. This site is frequently influenced by long-range transport of Hg
129 released from anthropogenic sources and biomass burning in southwestern China,
130 Indochinese Peninsula, and South Asia (Wang et al., 2015).

131 The MWLG site is situated at the summit of Mt. Waliguan at the edge of northeastern
132 part of the Qinghai-Xizang (Tibet) Plateau. The surrounding area is mainly dominated by
133 naturally preserved upland grasslands and arid/semiarid lands and there are no nearby sources.
134 The potential source regions are industrial and urbanized centers in northwestern China and
135 northern India (Fu et al., 2012a).

136 The BYNLK site is located in the Bayinbuluk upland grassland in the southern Tianshan
137 mountains, Xinjiang Uygur Autonomous Region, central Asia and with a total area of ~23,000
138 km^2. The surrounding area is rural and the population density is generally lower than 1 people
139 per km^2.

140 The GY site is located in the downtown of Guiyang city, the capital city of Guizhou
141 province, southwestern China, with a population of ~4.5 million. Guiyang is heavily polluted
142 with respect to atmospheric Hg due to significant industrial emissions (Fu et al., 2011).

143 Surrounding areas of the sampling site are mixed residential and commercial.

144 **2.2 Precipitation collection and analysis**

145 Precipitation samples were collected in 2011-2014 at MCB and MAL, 2012-2014 at
146 MDM and MWLG, 2013-2014 at BYBLK, 2012-2013 at GY, and 2008-2009 at MLG.
147 Wet-only precipitation samples were collected at all the studied sites using wet-only
148 automatic precipitation collectors (HHG9-ZJC-III, Beijing Midwest Great Technology Co.
149 Ltd., Beijing, China) with the exception of the MLG site. Wet-only precipitation samples at
150 the MLG site were collected using a manual method described in Fu et al. (2010a). The
151 HHG9-ZJC-III sampler consists of a 25-cm diameter, borosilicate glass bucket (volume: 15 L)
152 for precipitation collection and a moisture sensor that could move a cover from and back to
153 the bucket at the onset and the end of a precipitation event using a mechanical arm,
154 respectively (Price et al., 2008). The cover consists of a foam pad encased in plastic wrap and
155 forces a tight seal over the borosilicate glass bucket to prevent evaporation of the sample. The
156 borosilicate glass bucket was baked for one hour in a muffle furnace at a temperature of
157 500 °C before field sampling and rigorously rinsed by Milli-Q water before each of the new
158 sampling cycle during the field sampling, and this was aimed to avoid possible contamination
159 during the collection of precipitation Hg (Burke et al., 1995). Immediately after a
160 precipitation event, precipitation sample was transferred carefully to a rigorously acid-cleaned
161 Teflon sample bottle (volume: 250 mL) for Hg analysis. Extra samples during heavy
162 precipitation events (e.g., accumulative precipitation depth of >6 mm) were used for anion
163 and trace element analysis or abandoned in the field. We caution that, due to the adsorptive
164 behavior of Hg to the wall of borosilicate glass bucket, Hg in precipitation might be lost to the
165 bucket during the transfer processes which consequently resulted in an underestimation of Hg
166 concentrations in precipitation. However, the artifact during transferring of precipitation is
167 expected to be low because pervious studies observed minor adsorption of Hg to the clean
168 Teflon and borosilicate glass surface (Vermette et al., 1995;Landis and Keeler, 1997). In the
169 winter at several sampling sites, i.e., MCB, MWLG, and BYBLK, surface fresh snow was

collected from the upper 1 cm of the snowpack at open-air sites using a pre-cleaned Teflon scoop. Snow samples were collected immediately after snow events. The snow samples were transferred carefully to a thoroughly acid-cleaned Teflon sample bottle (volume: 400 mL) and remained sealed at a room temperature of 10-20 °C in the dark to melt snow. After the completion of field sampling, trace metal grade HCl (to 5‰ of total sample volume) was added into the Teflon bottles with rain and snow samples and each sample was individually sealed into three successive polyethylene bags and kept in a refrigerator at 4 °C until Hg analysis. To ensure clean operation, polyethylene gloves were worn throughout the sample collection period.

Hg concentrations in precipitation were analyzed using US EPA Method 1631 (USEPA, 2002). Briefly, Hg in precipitation was purged from solution in a Hg-free nitrogen stream and concentrated on to a gold-coated sand trap after oxidation by BrCl followed by addition $NH_2OH{\cdot}HCl$ to discharge the excess BrCl and reduction of divalent Hg by $SnCl_2$ to Hg^0. The trapped Hg is then thermally desorbed from the gold trap into an inert gas stream and quantified using a dual amalgamation technique followed by cold-vapor atomic fluorescence spectrometry (CVAFS) (USEPA, 2002). The data quality was controlled using laboratory blank tests (< 0.3 ng L^{-1}), field blank tests (Hg conc. = 0.46 ± 0.51 ng L^{-1}, 1SD, n = 21), and the Interlaboratory Comparison Program initiated by the U.S. Geological Survey (USGS, https://bqs.usgs.gov/PCQA/Interlaboratory_Comparison/). The measured Hg values of the standard reference samples prepared by the USGS during 2012-2014 were overall within the quality control acceptance criteria (62% and 90% of measured values were within the 10% and 20% of the most probable values (MPV, the mean results submitted by all the laboratory participants), respectively and showed a mean difference of 0.47 ± 1.55 ng L^{-1} (1SD, n = 50) in Hg concentrations between the measured values and MPV.

2.3 Collection and analysis of litterfall samples

Litterfall samples were collected using litterfall collectors (0.25 m^2 at MLG and 1.0 m^2 at MCB, MDM, and MAL) at the sampling sites (St Louis et al., 2001;Fisher and Wolfe,

2012;Risch et al., 2012). The collectors were constructed using untreated wood frame and Nylon net bottom with a pore size of 1.0 mm × 1.0 mm. At the MCB, MDM, MLG, and MAL sites, four, four, three, and eight collectors were deployed, respectively, which were located under the predominant tree species at the study sites. The litterfall collectors were elevated by approximately 20-40 cm above the ground surfaces. Collections of litterfall at the MDM, MLG, and MAL sites were conducted throughout a full year sampling campaign (Table 2). Collections of litterfall at the MCB site were conducted from September to October 2013, during which the accumulative litterfall biomass fluxes in the four collectors ranged from 214 to 355 g m^{-2} (mean: 281 ± 59 g m^{-2}, 1SD, n = 4), corresponding to approximately 58% of the annual litterfall mass flux at the sampling site (Zhou et al., 2014). We therefore assume that the measured Hg concentrations in litterfall at the MCB site were representative of the annual means. Litterfall samples were collected monthly, packed into paper bags, and air-dried in a clean environment (i.e., low atmospheric total gaseous mercury (TGM) concentration (< 3 ng m^{-3}), relatively dry air condition and isolated from precipitation) near the sampling sites. Litterfall samples were milled using a pre-cleaned food blender and stored in the laboratory until Hg analysis. Polyethylene gloves were worn throughout the sample collections and grinding period and the blenders were cleaned with Mili-Q water and ethanol between samples to prevent potential contaminations.

Hg concentrations in litterfall samples were determined using a Lumex RA-915 b multifunctional mercury analyzer (Lumex Ltd., Russia) equipped with a pyrolysis attachment. The Lumex RA-915 b analyzer was routinely calibrated and the QA/QC of the measurements of litterfall samples were controlled by procedural blanks, certificated reference materials (GBW10020, 150 ng g^{-1}), and duplicated analyses. The recoveries of certified standards ranged from 88-112% (mean: 98 ± 5%, 1SD, n = 60) for plant samples. All samples were measured in duplicates and the mean concentrations were adopted in this study.

2.4 Deposition fluxes of Hg in precipitation and litterfall

Volume-weighted mean (VWM) Hg concentration at each sampling site was calculated

using Equation (1):

$$VWM = \frac{\sum_i^n Hg_i \times PD_i}{\sum_i^n PD_i} \quad (1)$$

where VWM is the volume-weighted mean of precipitation Hg concentrations in ng L^{-1}, Hg_i and PD_i are the Hg concentration (ng L^{-1}) and precipitation depth (mm) of a single precipitation event i, respectively.

Annual wet deposition flux of Hg at each sampling site was estimated using the VWM Hg concentration calculated from all samples and the mean annual precipitation depth. Mean litterfall deposition flux of Hg at each sampling site was calculated by averaging the litterfall Hg fluxes obtained for all litterfall collectors, which were calculated by multiplying the Hg concentration in litters by the corresponding annual litterfall biomass.

3 Results and discussion

3.1 Precipitation Hg concentrations and deposition fluxes

Concentrations of Hg in precipitation and corresponding precipitation depth are presented in Figure 2. Large variation in precipitation Hg concentrations was observed at all sampling sites with the maximum concentrations up to an order of magnitude higher than the minimum concentration. The VWM Hg concentrations in precipitation at the remote sites varied from 3.7 to 7.7 ng L^{-1} (mean: 5.6 ± 2.0 ng L^{-1}, Table 1), with the highest VWM Hg concentration observed at the BYBLK site and the lowest at the MAL and MDM sites. The VWM Hg concentration in precipitation at the urban site of GY was 11.9 ± 6.1 ng L^{-1}, which was 1.5 to 3.2 times higher than the values at remote sites (Table 1). We acknowledge that, due to the lack of long-term simultaneous observations, the variation in VWM Hg concentrations in precipitation among the sampling sites may have uncertainties. In the present study, precipitation samples at the urban and rural sites were collected throughout one to three years, and there might exist interannual variations in VWM Hg concentrations at each sampling site. For example, the maximum annual VWM Hg concentration (5.1 ng L^{-1}) at MAL was observed during June 2013-May 2014, which was approximately 1.8 times higher

than that (2.9 ng L^{-1}) during June 2011-May 2012. At MCB, annual VWM Hg concentrations showed the highest (8.1 ng L^{-1}) during August 2012-July 2013 and the lowest (6.0 ng L^{-1}) during July 2014- August 2014.

VWM Hg concentrations in precipitation at all sites showed a clear season trend with lower concentrations in summer wet season and higher concentrations in winter dry season (Figure 3). This pattern is consistent with previous observations in rural and urban areas of China (Huang et al., 2012;Huang et al., 2013;Ma et al., 2016). Higher Hg concentrations in precipitation during the winter dry season were potentially due to elevated wintertime atmospheric PBM concentrations in China (Fu et al., 2008;Zhang et al., 2013;Xu et al., 2014;Zhu et al., 2014), which could be incorporated into wet deposition via scavenging processes below cloud. Lower VWM Hg concentrations in precipitation during summer wet season were mostly associated with higher precipitation amounts at the sampling sites, suggesting increasing amounts of precipitation would dilute the Hg concentrations in samples that scavenged from the boundary layer during the onset of the precipitation (Gratz et al., 2009;Yuan et al., 2015).

Annual fluxes of Hg in precipitation at the sampling sites varied from 2.0 to 12.6 μg m^{-2} yr^{-1} (mean: 5.9 ± 3.6 μg m^{-2} yr^{-1}, Table 1). Wet deposition fluxes showed a clear urban-rural difference, with the annual deposition flux at the urban site of GY elevated by a factor of 1.8 to 6.3 compared to the values at rural sites. This could be explained by the elevated VWM Hg concentration in precipitation at the GY site. Wet deposition fluxes at rural sites also showed a clear regional difference. The annual wet deposition fluxes of Hg in the subtropical zones in southwestern and eastern China (i.e., MAL, MLG, and MDM) were relatively higher (by a factor of 1.1 to 1.3) than that at the MCB site in the temperate zone in northeastern China, and much higher (by a factor of 3 to 3.6) than that at the MWLG and BYBLK sites, which were located in the arid/semi-arid zones in northwestern China. This regional variation could not be explained by the difference of VWM Hg concentrations in precipitation because the correlation between annual wet deposition fluxes of Hg and VWM Hg concentrations in precipitation is not significant ($p > 0.05$). Instead, annual wet deposition fluxes of Hg were

positively correlated with annual precipitation depth at the remote sites ($r^2 = 0.86$, $p < 0.01$). This suggests that precipitation depth had a greater influence on the regional variation of wet deposition fluxes of Hg at remote sites of China than VWM Hg concentrations, which is in agreement with previous studies in the North America (Risch et al., 2012;Zhang et al., 2012a).

The VWM Hg concentrations in precipitation at the remote sites of this study were overall consistent with previous observations in China (Figure 4). For example, VWM Hg concentrations in precipitation and wet deposition fluxes of Hg at the Nam Co and SET stations of the Tibetan Plateau and in Mt. Simian, southwestern China ranged from 4.0 to 10.9 ng L^{-1} and from 1.8 to 15.4 μg m^{-2} yr^{-1}, respectively (Huang et al., 2012;Huang et al., 2015;Ma et al., 2016). However, the VWM Hg concentration at the GY site was 1.0-4.4 times lower than the levels (12.3-52.9 ng L^{-1}) observed in other urban areas of China; and the wet deposition fluxes of Hg at the GY site was consequently lower than those (14.0-56.5 μg m^{-2} yr^{-1}) in the urban areas of China, with the exception of the flux observed in Lhasa of the Tibetan Plateau (flux: 8.2 μg m^{-2} yr^{-1}) (Wang et al., 2009;Wang et al., 2012;Huang et al., 2013;Xu et al., 2014;Zhu et al., 2014).

3.2 Litterfall Hg concentrations and deposition fluxes

Average Hg concentrations in litterfall at the MCB, MDM, MLG, and MAL sites were 47.0 ± 19.0, 42.3 ± 5.6, 91.1 ± 29.4, and 56.9 ± 4.4 ng g^{-1}, respectively (mean: 59.3 ± 22.0 ng g^{-1}, Table 2). Concentrations of Hg in litterfall could be affected by many factors including atmospheric Hg concentrations, tree species, "leaf maintenance" period, and environmental factors (Lindberg and Stratton, 1998;Frescholtz et al., 2003;Ericksen and Gustin, 2004;Millhollen et al., 2006;Poissant et al., 2008). The variations of litterfall Hg concentrations observed in different collectors (corresponding to sampling of different tree species) at each sampling site was insignificant (p values for all > 0.05, Table 2). Annual mean atmospheric TGM at the MCB, MDM, MLG, and MAL sites were 1.73 ± 0.48, 3.31 ± 1.44, 2.80 ± 1.51, and 2.09 ± 0.63 ng m^{-3} (Fu et al., 2015b), respectively, which were not significantly correlated with the Hg concentrations in litterfall samples ($p = 0.87$). The lack of

significant correlation might be partly attributed to the fact that mean atmospheric TGM concentration during the "leaf maintenance" period for each tree specie were not entirely equal to the annual means of atmospheric TGM at the sampling sites because atmospheric TGM concentrations generally exhibited clear seasonal trends in rural areas of China (Fu et al., 2015b). In addition, other factors including tree species, "leaf maintenance" period, and regional environmental factors should also play an more important role in litterfall Hg concentrations. Hg concentrations in litterfall at the MDM site were found to increase from July to December (correlation slope = 7.0 ± 0.7 ng g^{-1} mon^{-1}, $r^2 = 0.78$, $p < 0.01$, litterfall were not collected during January-June due to little production of litterfall biomass). In contrast, significant monthly variation in Hg concentrations in litterfall samples at the MCB and MAL sites were not found (p values for both > 0.05).

Annual fluxes of Hg through litterfall at the four sampling sites ranged from 22.8 to 62.8 μg m^{-2} yr^{-1} (mean of 37.0 μg m^{-2} yr^{-1},Table 2). The litterfall fluxes of Hg showed a clear regional distribution pattern with the fluxes decreasing with latitude. The highest flux (62.8 μg m^{-2} yr^{-1}) was observed at the MAL site in the south subtropical zone in southwestern China, followed by the MLG site (flux: 39.5 μg m^{-2} yr^{-1}) in the middle subtropical zone, the MDM site (flux: 23.1 μg m^{-2} yr^{-1}) in the north subtropical zone, and the MCB site (flux: 23.1 μg m^{-2} yr^{-1}) in the middle temperate zone (Zheng et al., 2010). The relatively higher litterfall flux of Hg at the MAL and MLG sites could be explained by either higher annual biomass of litterfall or higher Hg concentrations in litterfall samples (Table 2). Deposition fluxes of Hg through litterfall in this study were comparable to those (35.5-42.9 μg m^{-2} yr^{-1}) measured in Mt. Gongga and Mt. Simian, southwestern China (Fu et al., 2010b;Ma et al., 2016), but substantially lower than that (220 μg m^{-2} yr^{-1}) measured at Tieshanping which was close to Chongqing city, southwestern China(Wang et al., 2009).

3.3 Relative contribution of wet and litterfall deposition to total Hg deposition in forests

Ratios of annual mean litterfall deposition flux relative to annual wet deposition flux of Hg at the four sampling sites ranged from 3.9 to 8.7 (mean: 5.8 ± 2.3). The ratios were overall

consistent with the previous observations in China. The ratios at the four remote sampling sites were relatively higher (by a factor of 1.4 to 3.1) than that measured at a rural sites in Mt. Simian, southwestern China (ratio: 2.8) (Ma et al., 2016), but relatively lower (by a factor of 1.2 to 2.0) than that measured at Tieshanping, Chongqing, southwestern China (ratio: 7.6) (Wang et al., 2009), with the exception of the MAL site. On the other hand, the observed ratios in China were much greater than those observed in the North America and Europe. Rich et al. (2012) collected litterfall at twenty-three remote sites in the eastern USA and found that the mean ratio of litterfall Hg deposition to Hg wet deposition was 1.3 (ranged from 0.4 to 2.6), which was 3.0 to 6.7 times lower compared to the ratios observed in China. In Europe, ratios of litterfall Hg deposition to Hg wet deposition were in the range of from 0.4 to 2.6 (mean: 1.2 ± 0.8, n = 5) (Iverfeldt, 1991;Munthe et al., 1995;Lee et al., 2000;Schwesig and Matzner, 2000).

Hg in litterfall biomass has been suggested to be mostly from uptake of atmospheric GEM, and therefore litterfall deposition could be a good indicator of GEM dry deposition to forest ecosystems (Frescholtz et al., 2003;Gustin, 2012;Zhang et al., 2012a). In addition to litterfall and wet deposition, dry deposition of PBM and GOM to forest floor and other surfaces could also contribute to the total Hg deposition to a forest. Given the measured atmospheric PBM and GOM concentrations (Fu et al., 2015b;Yu et al., 2015), the dry deposition fluxes of PBM and GOM at the MCB, MDM, and MAL sites were estimated to be 3.0, 9.6, and 2.3 μg m^{-2} yr^{-1}, respectively, using the average dry deposition velocities of PBM and GOM over forests modeled by Zhang et al. (2012a). At the MLG sites, annual dry deposition flux of PBM and GOM was estimated to be 4.4 μg m^{-2} yr^{-1} using the comparison of precipitation and throughfall data collected side by side (St Louis et al., 2001;Fu et al., 2010a;Gustin, 2012). The importance of litterfall in the total deposition of Hg has been highlighted by many previous studies (St Louis et al., 2001;Lindberg et al., 2007;Risch et al., 2012;Zhang et al., 2012a). In this study, we estimate that litterfall deposition represented 60-87% (mean: 74.5 ± 11.4%) of total Hg deposition to the four studied forests, which were much higher compared to those (mean: 46.2 ± 12.5%) over rural forests in the North America

and Europe (Munthe et al., 1995;Rea et al., 1996;Grigal et al., 2000;Lee et al., 2000;St Louis et al., 2001;Zhang et al., 2012a); whereas the wet deposition played a minor role (mean: 13.9 ± 3.5%) in the total Hg deposition budget. Therefore, Hg deposition through litterfall played a predominant role in the total Hg deposition budget in forest ecosystems in China.

3.4 Comparison with observations in other regions worldwide

Figure 5 shows the comparison of wet deposition and litterfall fluxes of Hg as well as TGM/GEM, PBM, and GOM concentrations in China, North America, and Europe. The mean wet deposition flux of Hg at remote sites in China was 5.6 ± 4.2 μg m^{-2} yr^{-1} (Figure 5A, data from this study and the literature (Huang et al., 2012;Huang et al., 2015;Ma et al., 2016)), which is 4.4 times lower than the mean (24.8 ± 17.8 μg m^{-2} yr^{-1}) at urban sites of China (Figure 5A, data are from this study and the literature (Wang et al., 2009;Wang et al., 2012;Huang et al., 2013;Xu et al., 2014;Zhu et al., 2014)). The mean wet deposition fluxes of Hg in the North America and Europe were 9.5 ± 4.2 and 6.8 ± 3.2 μg m^{-2} yr^{-1} (Figure 5A), respectively (EMEP;Prestbo and Gay, 2009). In contrast to the observations in China, the urban-rural variation in the wet deposition fluxes of Hg was insignificant in the North America (Zhang et al., 2012a).

The observations from this study and the literature suggested that wet deposition fluxes of Hg in urban areas of China were highly elevated (by a factor of 2.6 to 3.6) compared to the North America and Europe. In China, wet deposition fluxes of Hg were significantly correlated with VWM Hg concentrations in precipitation ($r^2 = 0.87$, $p < 0.01$, Figure 6A), whereas no significant correlation existed between wet deposition fluxes of Hg and annual precipitation depth ($r^2 = 0.02$, $p = 0.65$). Elevated wet deposition fluxes of Hg at urban sites of China were associated with the elevated VWM Hg concentrations in precipitation (Figure 6A). Wet deposition fluxes of Hg in China were also positively correlated with ground-level TGM/GEM, PBM, and GOM concentrations (Figure 6B, C, and D). Wet deposition of Hg has been suggested to result from the scavenging of PBM and GOM in cloud (i.e., rainout) and below cloud (i.e., washout) (Seigneur et al., 2004;Lin et al., 2006). In North America, a

387 modeling study suggest that scavenging of GOM in and below cloud contributed mostly
388 (~89%) of wet deposition of Hg, with ~41% contributed by washout (Selin and Jacob, 2008).
389 In China, ground based measurements of GOM in urban areas found that the mean GOM
390 concentrations (means: 47.9 pg m^{-3}, Figure 5E, (Fu et al., 2011;Xu et al., 2015)) were 5.4
391 times greater than the mean (8.9 pg m^{-3}) in the North America (Swartzendruber et al.,
392 2006;Yatavelli et al., 2006;Valente et al., 2007;Fain et al., 2009;Peterson et al., 2009;Song et
393 al., 2009;Engle et al., 2010;Zhang et al., 2012a). Scavenging of GOM in the continental
394 boundary layer (i.e., washout) would therefore contribute to the elevated wet deposition
395 fluxes of Hg at urban sites of China. It should be noted that PBM concentrations were also
396 highly elevated (mean: 239 ± 102 pg m^{-3}) in the urban areas of China, which were 10-20
397 times greater than the levels observed in North America and Europe; and approximately 5
398 times greater than the mean GOM concentrations at the same locations (Figure 5D and 5E).
399 Lee et al. (2001) estimated that washout of PBM contributed approximately 1.0 μg m^{-2} yr^{-1} to
400 the total Hg deposition in the United Kingdom at a background PBM concentration of 10 pg
401 m^{-3}. Given the mean PBM concentration in urban areas of China, the mean flux of washout of
402 PBM below cloud is roughly estimated to be 24 μg m^{-2} yr^{-1}, which explains > 90% of the wet
403 deposition flux of Hg in the urban areas of China. Since scavenging of PBM below cloud also
404 depends on other factors including the vertical distribution of PBM, intensity of precipitation,
405 and cloud base height (Tanner et al., 1997;Hicks, 2005;Brooks et al., 2014), the estimate may
406 have large uncertainties. Nevertheless, the estimate is in agreement with the measured fraction
407 of particulate mercury (Hg_p) in wet deposition flux of Hg at an urban site in China. Huang et
408 al. (2013) found that ~86% of the annual wet deposition of Hg in Lhasa of the Tibetan Plateau
409 was associated with Hg_p, much higher than that at a rural site in the Tibetan Plateau (55%,
410 (Huang et al., 2015)) as well as at rural and urban sites in the North America (26-63%, (Burke
411 et al., 1995;Lamborg et al., 1995;Poissant and Pilote, 1998)). These suggest the scavenging of
412 PBM below cloud was an important contributor of the elevated wet deposition fluxes of Hg at
413 urban sites of China.

414 On the other hand, mean wet deposition flux of Hg (5.6 ± 4.2 μg m^{-2} yr^{-1}) at the rural

415 sites of China was relatively lower (by a factor of 1.2 to 1.7) compared to those measured in
416 the North America (9.5 ± 4.2) and Europe (6.8 ± 3.2) (EMEP;Prestbo and Gay, 2009). This
417 regional pattern is different from model results that predicted higher wet deposition in China
418 because of large anthropogenic Hg emissions (Bergan et al., 1999;Dastoor and Larocque,
419 2004). There are several possible explanations for the lower wet deposition fluxes of Hg
420 observed in the rural areas of China. Wet deposition fluxes of Hg at the rural sites of China
421 were mostly observed in arid, semi-arid, and sub-humid climate zones in northwestern and
422 northeastern China (i.e., MWLG, MCB, and BYBLK in this study, Nam Co, and SET
423 stations), where the precipitation depth is generally low (260-975 mm, data from this study
424 and the literature (Huang et al., 2012;Huang et al., 2015)) and anthropogenic Hg sources are
425 scarce (Zhang et al., 2015b). The remaining four rural sites (i.e., MDM, MLG, MAL in this
426 study and Mt. Simian (Ma et al., 2016)) were all located in mountaintop forests. Although the
427 observations at some of these sites showed elevated PBM concentrations (mean: 31-154 pg
428 m^{-3}) (Fu et al., 2015b;Yu et al., 2015), washout of PBM below cloud was not expected to
429 contribute significantly to Hg in precipitation because of low cloud base heights (Ray et al.,
430 2006). In addition, observations of GOM at high-altitude sites in China (i.e., MWLG, MAL,
431 and Shangri-La) showed mean concentrations of 2-8 pg m^{-3} (Fu et al., 2012a;Fu et al.,
432 2015b;Zhang et al., 2015a), significantly lower than those (20-87 pg m^{-3}) measured at
433 high-altitude sites in the North America and Europe (Swartzendruber et al., 2006;Fain et al.,
434 2009;Weiss-Penzias et al., 2009;Fu et al., 2016). Relatively lower GOM concentrations at the
435 high-altitude sites in China were possibly due to the elevated atmospheric particulate matters
436 in China that facilitates the partitioning of GOM to the particulate phase (Slemr et al.,
437 2009;Swartzendruber et al., 2009;van Donkelaar et al., 2010;Amos et al., 2012;Zhang et al.,
438 2013). Since the scavenging of GOM in the free troposphere and continental boundary layer
439 is an important source of wet deposition of Hg (Selin and Jacob, 2008), the lower GOM
440 concentrations in the rural areas of China could be responsible for the lower wet deposition
441 fluxes of Hg observed in the rural areas of China.

442 Annual fluxes of Hg through litterfall at the rural sites in this and previous studies in

China ranged from 22.8 to 62.8 μg m^{-2} yr^{-1} (mean: 37.8 ± 14.8 μg m^{-2} yr^{-1}, n = 6, Figure 5B, data are from this study and the literature (Fu et al., 2010b;Ma et al., 2016)). Hg fluxes through litterfall in the rural areas of China were 1.4-4.7 times higher than the means observed in North America (13.3 ± 5.8 μg m^{-2} yr^{-1}) and Europe (16.5 ± 8.7 μg m^{-2} yr^{-1}) (Munthe et al., 1995;Rea et al., 1996;Lee et al., 2000;Schwesig and Matzner, 2000;St Louis et al., 2001;Lindberg et al., 2007;Larssen et al., 2008;Fisher and Wolfe, 2012;Juillerat et al., 2012;Risch et al., 2012), but approximately 2.2 times lower than those (mean: 84.4 ± 49.0 μg m^{-2} yr^{-1}) measured in South America (Roulet et al., 1998;Fostier et al., 2003;Mélières et al., 2003;Magarelli and Fostier, 2005;Silva-Filho et al., 2006;Teixeira et al., 2012). Global Hg fluxes through litterfall were positively correlated with both Hg concentrations in litterfall (r^2 = 0.69, $p < 0.01$) and litterfall biomass production (r^2 = 0.70, $p < 0.01$) (Figure 7). Forward stepwise multiple regression analysis suggests that litterfall biomasses and Hg concentrations in litterfall explained 69.2% and 25.4% of the regional variations in litterfall Hg fluxes, respectively. Production of litterfall biomasses at the rural sites of China ranged from 434 to 1100 g m^{-2} yr^{-1} (mean: 661 ± 307 g m^{-2} yr^{-1}, n = 6) and was approximately 2 times higher than that in North America and Europe (Munthe et al., 1995;Rea et al., 1996;Lee et al., 2000;Schwesig and Matzner, 2000;St Louis et al., 2001;Lindberg et al., 2007;Larssen et al., 2008;Fisher and Wolfe, 2012;Juillerat et al., 2012;Risch et al., 2012), which is the dominant factor causing the difference in litterfall Hg fluxes between China and North America/Europe. It is worth noting that most (5 out of the 6) of observations at the rural sites of China were made in subtropical moist forests, where the litterfall biomass productions are larger than those in the temperate and boreal forests in North America and Europe (Xiong and Nilsson, 1997;Running et al., 2004;Wang et al., 2008). Additionally, mean Hg concentrations in litterfall at the rural sites of China (63.3 ± 29.0 ng g^{-1}) were elevated by a factor of 1.4 compared to that (44.0 ± 10.4 ng g^{-1}) in the North America and Europe (Munthe et al., 1995;Rea et al., 1996;Lee et al., 2000;Schwesig and Matzner, 2000;St Louis et al., 2001;Lindberg et al., 2007;Larssen et al., 2008;Fisher and Wolfe, 2012;Juillerat et al., 2012;Risch et al., 2012). This could be partly attributed to the elevated TGM concentrations

(Figure 5C) and longer "leaf maintenance" period at most rural sites in China (Fescholtz et al., 2003;Poissant et al., 2008;Gustin, 2012;Fu et al., 2015b).

4 Conclusions

In this study, we analyzed the concentrations and deposition fluxes of Hg through precipitation and litterfall at multiple sites across a broad geographic area in mainland China. The VWM Hg concentrations and deposition fluxes of Hg in precipitation at rural sites were 3.7 to 7.7 ng L^{-1} (mean: 5.6 ± 2.0 ng L^{-1}) and 2.0 to 7.2 μg m^{-2} yr^{-1} (mean: 4.8 ± 2.2 μg m^{-2} yr^{-1}), respectively, lower than the VWM Hg concentration (11.9 ± 6.1 ng L^{-1}) and deposition fluxes of Hg (12.6 ± 6.5 μg m^{-2} yr^{-1}) at the urban GY site. Elevated wet deposition fluxes of Hg in urban areas of China corresponded to the elevated ground-level atmospheric Hg concentrations, indicating Hg scavenging below cloud was an important source of wet deposition of Hg in the urban areas of China. The wet deposition fluxes of Hg at the rural sites in this study, however, were lower than the observations in the North America and Europe, and different from previous model results, possibly due to a combination of low precipitation depth and low cloud base heights and the low GOM concentrations in the troposphere in China. Hg fluxes through litterfall at the sampling sites ranged from 22.8 to 62.8 μg m^{-2}, significantly higher (by a factor of 3.9 to 8.7) than the wet deposition fluxes in China and the litterfall deposition fluxes in the North America and Europe, indicating that litterfall deposition contributes significantly to the total Hg deposition budget in forest ecosystems in China.

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499 Figure1 Locations of the sampling sites in this study.

500 Figure 2 Mercury concentration and rain depth in single precipitation event at Mt. Ailao
501 (MAL), Mt. Changbai (MCB), Mt. Damei (MDM), Mt. Waliguan (MWLG), Bayinbuluk
502 (BYBLK), Mt. Leigong (MLG) and urban Guiyang (GY) in this study.

503 Figure 3 Monthly variations in volume-weighted mean (VWM) Hg concentrations at rural
504 and urban sites in China.

505 Figure 4 Annual VWM Hg concentrations in precipitation and wet deposition fluxes of Hg in
506 this study and literatures (Huang et al., 2012;Huang et al., 2013;Xu et al., 2014;Zhu et al.,
507 2014;Huang et al., 2015;Ma et al., 2016). Black and red values above columns
508 correspond to VWM concentration and wet deposition of Hg, respectively.

509 Figure 5 Comparison of (A) wet deposition flux of Hg; (B) Litterfall deposition of Hg; (C)
510 atmospheric total gaseous mercury (TGM)/gaseous elemental mercury (GEM)
511 concentrations; (D) atmospheric particulate bound mercury (PBM) concentrations; and
512 (E) atmospheric gaseous oxidized mercury (GOM) concentrations between China and
513 North America and Europe. Note that atmospheric PBM in Europe is referred to total
514 particulate bound mercury and in remaining regions is referred to particulate bound
515 mercury on particles with an aerodynamic diameter < 2.5 μm. Data are from this study,
516 the literature and reference therein (EMEP;Munthe et al., 1995;Lee et al., 2000;Schwesig
517 and Matzner, 2000;St Louis et al., 2001;Munthe et al., 2003;Pirrone et al.,
518 2003;Swartzendruber et al., 2006;Wang, 2006;Yatavelli et al., 2006;Demers et al.,
519 2007;Lindberg et al., 2007;Valente et al., 2007;Bushey et al., 2008;Choi et al.,
520 2008;Larssen et al., 2008;Li et al., 2008;Fain et al., 2009;Peterson et al., 2009;Prestbo
521 and Gay, 2009;Song et al., 2009;Wang et al., 2009;Engle et al., 2010;Sprovieri et al.,
522 2010;Liu et al., 2011;Fisher and Wolfe, 2012;Fu et al., 2012b;Juillerat et al., 2012;Risch
523 et al., 2012;Zhang et al., 2012b;Chen et al., 2013;Zhu et al., 2014;Fu et al., 2015b;Fu et
524 al., 2016;Ma et al., 2016).

525 Figure 6 Correlations between wet deposition fluxes of Hg and (A) Volume-weighted mean
526 (VWM) Hg concentrations in precipitation, (B) atmospheric total gaseous mercury

(TGM)/gaseous elemental mercury (GEM) concentrations, (C) atmospheric particulate bound mercury (PBM) concentrations, and (D) atmospheric gaseous oxidized mercury (GOM) concentrations in China. Data are from this study and the literature (Wang, 2006;Fu et al., 2011;Huang et al., 2012;Wang et al., 2012;Zhu et al., 2012;Huang et al., 2013;Xu et al., 2014;Zhu et al., 2014;Fu et al., 2015b;Xu et al., 2015).

Figure 7 Scatterplot of (A) Hg concentrations in litterfall and litterfall fluxes of Hg; and (B) litterfall biomasses and litterfall fluxes of Hg for the global observations. Data are from this study and the literature (Iverfeldt, 1991;Rea et al., 1996;Roulet et al., 1998;Fostier et al., 2000;Grigal et al., 2000;Schwesig and Matzner, 2000;St Louis et al., 2001;Mélières et al., 2003;Magarelli and Fostier, 2005;Sheehan et al., 2006;Silva-Filho et al., 2006;Demers et al., 2007;Wangberg et al., 2007;Bushey et al., 2008;Larssen et al., 2008;Wang et al., 2009;Fu et al., 2010b;Fisher and Wolfe, 2012;Juillerat et al., 2012;Risch et al., 2012;Teixeira et al., 2012;Benoit et al., 2013;Ma et al., 2016).

541 Figure 1
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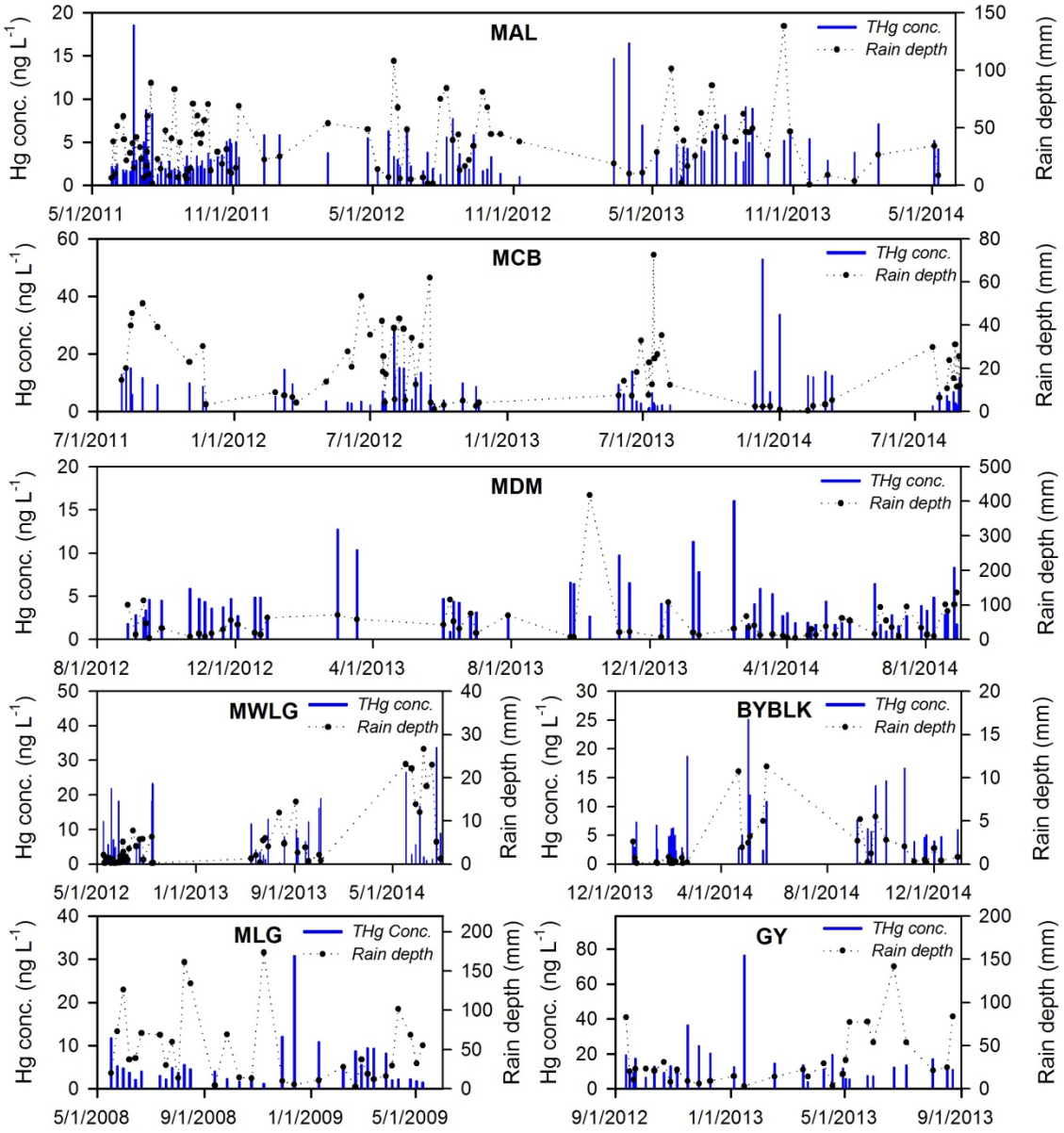
Figure 2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THg conc.
Rain depth
Hg conc. (ng L-1)
Rain depth (mm)
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11/1/2011
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11/1/2013
5/1/2014
MCB
7/1/2011
1/1/2012
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7/1/2013
1/1/2014
7/1/2014
MDM
8/1/2012
12/1/2012
4/1/2013
8/1/2013
12/1/2013
4/1/2014
8/1/2014
MWLG
5/1/2012
1/1/2013
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BYBLK
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4/1/2014
8/1/2014
12/1/2014
MLG
THg Conc.
5/1/2008
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550 Figure 3
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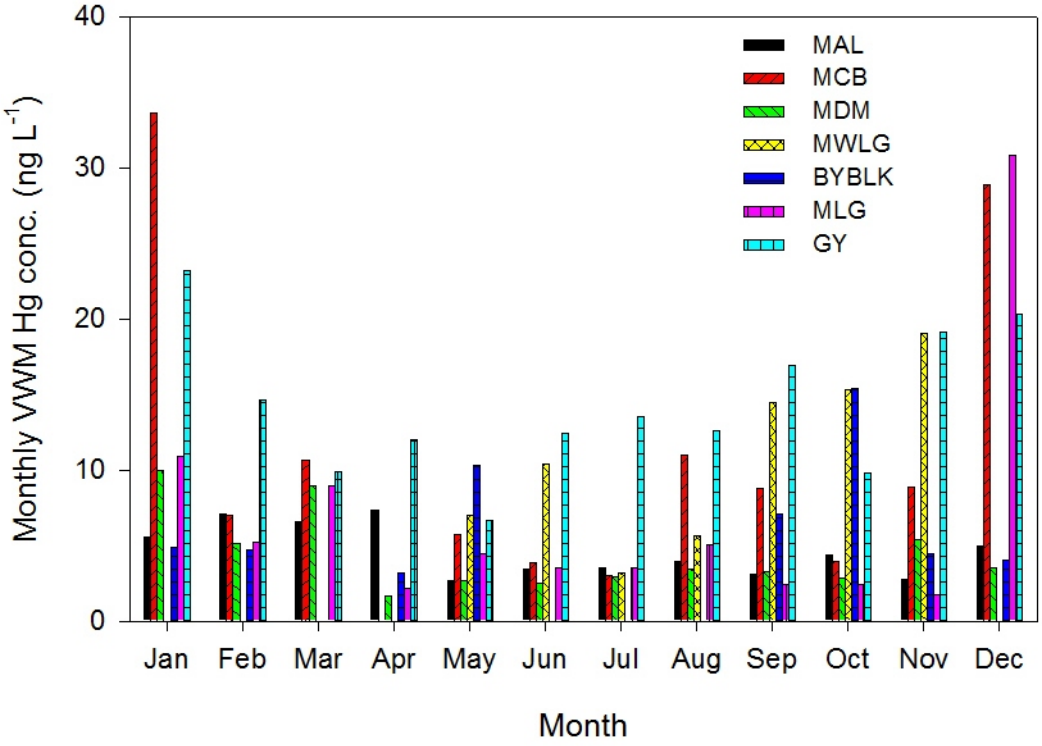

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556 Figure 4

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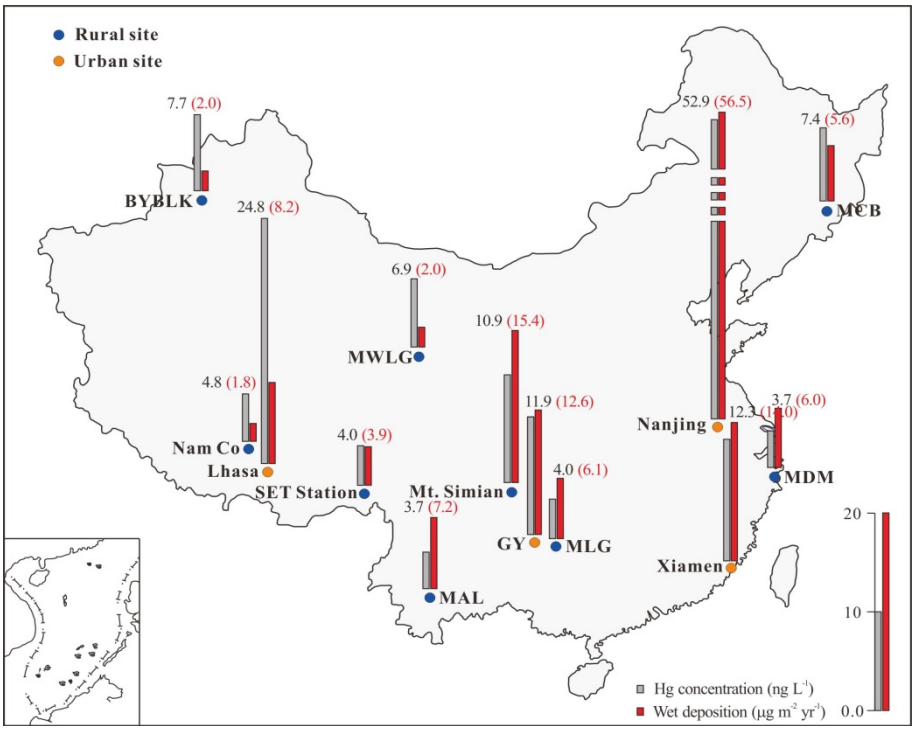

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562 Figure 5
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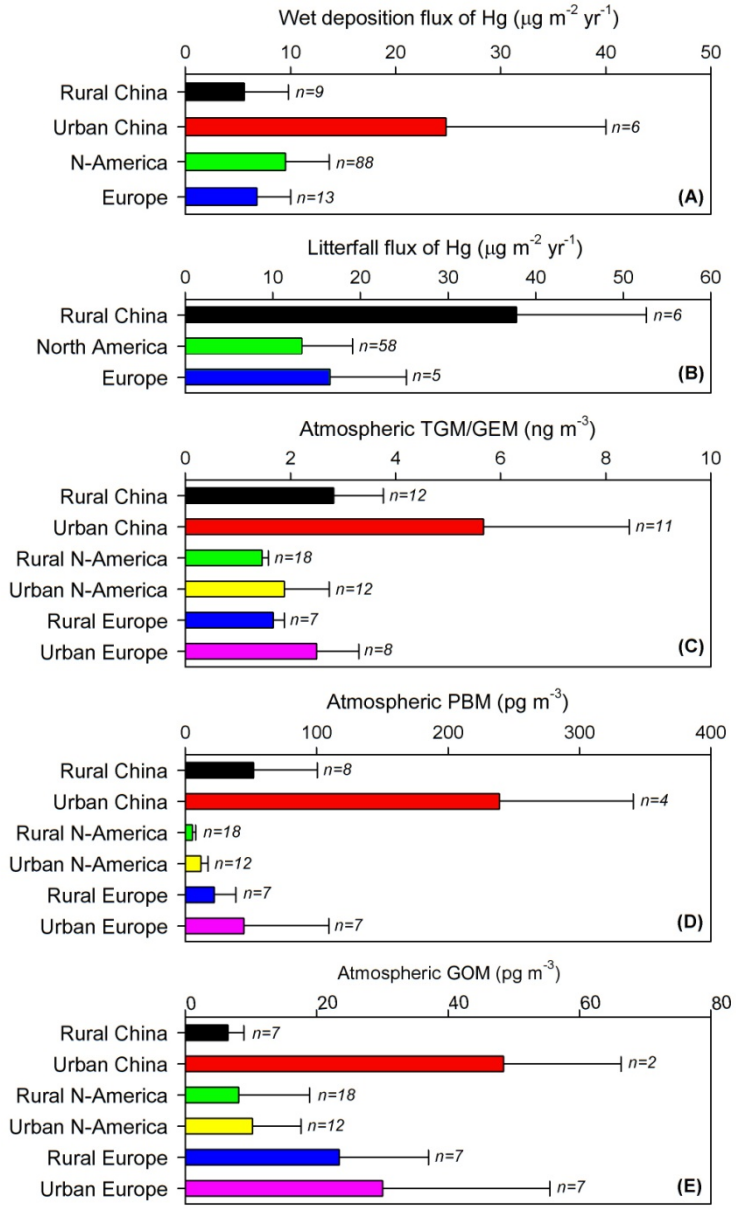

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566 Figure 6
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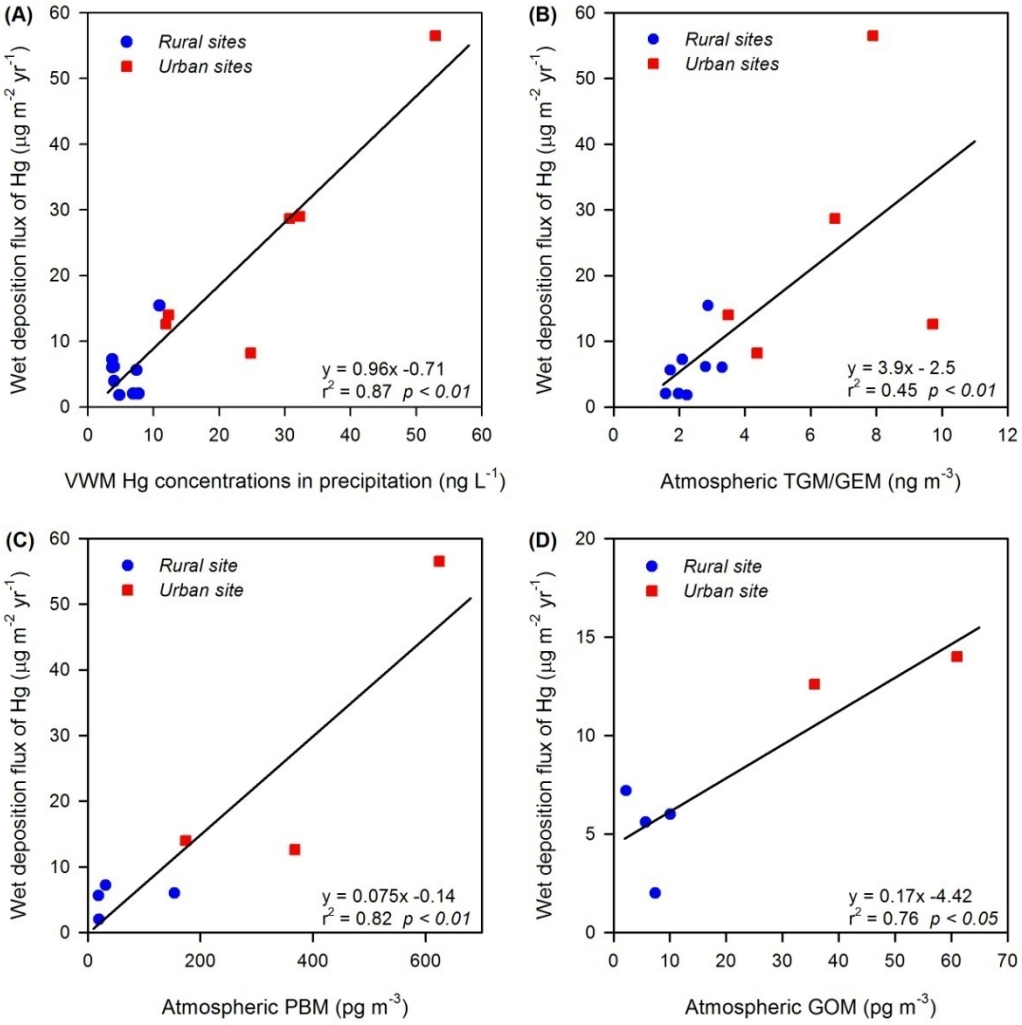

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570 Figure 7
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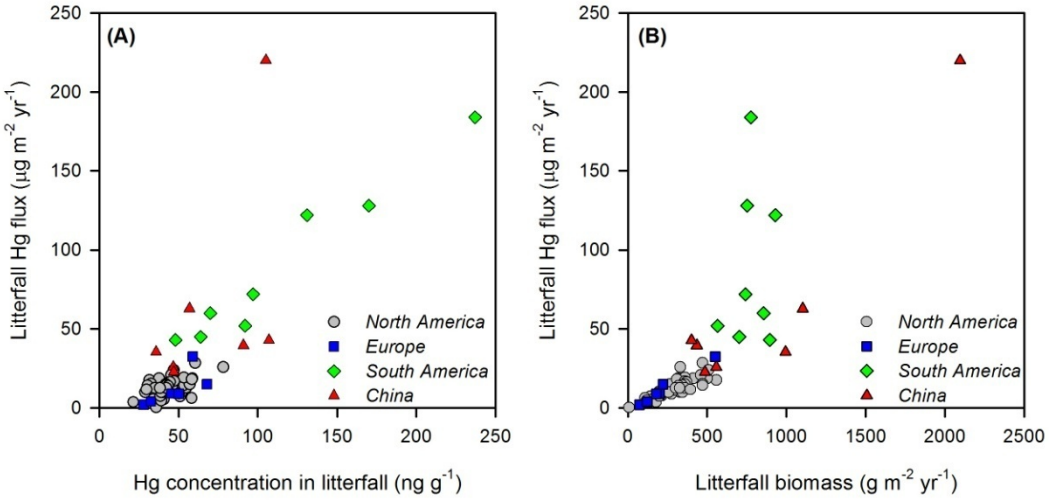

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Table 1 Sampling site information, volume-weighted mean (VWM) concentrations and annual wet deposition fluxes of Hg in precipitation at remote sites and a urban site of China

Site name	Site ID	Long (°)	Lat (°)	Elevation (m a.s.l)	Site category	Data coverage	Annual Rainfall (mm)	VWM Hg concentration (ng L^{-1})	Annual wet Hg deposition flux (μg m^{-2})
Mt. Changbai	MCB	128.112	42.403	736	Rural forest	Aug 2011-Aug 2014	751	7.4±6.1 (n=68)	5.6±4.6
Mt. Damei	MDM	121.565	29.632	550	Rural forest	Aug 2012-Aug 2014	1621	3.7±2.8 (n=63)	6.0±4.6
Mt. Leigong	MLG	108.203	26.387	2176	Rural forest	May 2008-May 2009	1533	4.0±2.8 (n=31)	6.1±4.2
Mt. Ailao	MAL	101.107	24.533	2450	Rural forest	May 2011-May 2014	1931	3.7±2.4 (n=103)	7.2±4.7
Mt. Waliguan	MWLG	100.898	36.287	3816	Rural grassland	May 2012-Aug 2014	290	6.9±8.6 (n=65)	2.0±2.4
Bayinbuluk	BYBLK	83.717	42.893	2500	Rural grassland	Dec 2013-Dec 2014	266	7.7±6.0 (n=36)	2.0±1.6
Guiyang city	GY	106.724	26.573	1041	Urban	Sep 2012-Aug 2013	1057	11.9±6.1 (n=29)	12.6±6.5

Table 2 Sampling site information, concentrations and annual litterfall deposition fluxes of Hg in litterfall at remote forest sites of China

Site name	Site ID	Sampling period	Collector	Concentration (ng g^{-1})	Annual litterfall mass flux (g m^{-2})	Annual litterfall Hg flux (μg m^{-2})
Mt. Changbai	MCB	Sep-Oct 2013	MCB-1	74.6	486*	36.2
			MCB-2	41.4	486*	20.1
			MCB-3	31.1	486*	15.1
			MCB-4	40.9	486*	19.9
			Average ± SD	47.0±19.0	486	22.8±9.2
Mt. Damei	MDM	Aug 2012 - Jul 2013	MDM-1	42.8	381	16.3
			MDM-2	52.	614	31.9
			MDM-3	52.3	557	29.1
			MDM-4	42.0	354	14.9
			Average ± SD	42.3±5.6	476±128	23.1±8.7
Mt. Leigong	MLG	May 2008 – Apr 2009	MLG-1	57.2	308	17.6
			MLG-2	106.2	287	30.5
			MLG-3	109.8	642	70.4
			Average ± SD	91.1±29.4	412±199	39.5±275
Mt. Ailao	MAL	Jun 2012 – May 2013	MAL-1	58.0	1131	65.6
			MAL-2	55.8	1053	58.8
			MAL-3	62.7	948	59.4
			MAL-4	48.4	1710	82.8
			MAL-5	60.6	1001	60.7
			MAL-6	53.7	760	40.8
			MAL-7	59.0	1232	72.7
			MAL-8	56.8	1080	61.3
			Average ± SD	56.9±4.4	1114±278	62.8±12.1

(Annual litterfall mass flux*: values were cited from Zhou et al. (2014))

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