Contents lists available at ScienceDirect

Atmospheric Environment

Examination of total mercury inputs by precipitation and litterfall in a remote upland forest of Southwestern China

Jun Zhou [a, b], Xinbin Feng [a, *], Hongyan Liu [b], Hui Zhang [a, c], Xuewu Fu [a], Zhengduo Bao [a, c], Xun Wang [a, c], Yiping Zhang [d]

[a] State Key Laboratory of Environmental Geochemistry, Institute of Geochemistry, Chinese Academy of Sciences, Guiyang 550002, China
[b] College of Resources and Environmental Engineering, Guizhou University, Guiyang 550025, China
[c] Graduate University of the Chinese Academy of Sciences, Beijing 100049, China
[d] Xishuangbanna Tropical Botanical Garden, Chinese Academy of Sciences, Kunming 650223, China

HIGHLIGHTS

- The Hg deposition fluxes at a remote area in Southwestern China were studied.
- Atmospheric deposition fluxes were highly elevated in the studies forest area.
- Litterfall Hg depositions were the major pathway for Hg loading to the forest catchment.
- Forest ecosystem in the study area was a large pool of atmospheric Hg.

ARTICLE INFO

ABSTRACT

China is regarded as one of the largest anthropogenic mercury (Hg) emission source regions over the world. However, wet and dry deposition of atmospheric Hg in China has not been well investigated. In the present study, wet and litterfall depositions of total mercury (THg) were continuously measured from June 2011 to May 2012 at a high-altitude site in Mt. Ailao area, Southwestern China. The annual volume-weighted mean concentration of THg and reactive mercury (RHg) in precipitation was 2.98 and 0.92 ng L^{-1}, respectively. The mean THg concentration in litterfall was 52 ng g^{-1} (dry weight). Atmospheric deposition was highly elevated in forest in the study area, with the annual mean THg deposition fluxes of 76.7 µg m^{-2} yr^{-1}. Litterfall Hg depositions were the major pathway for Hg loading to the forest catchment, which were 71.2 µg m^{-2} yr^{-1} (about 92.8% of total input for THg). Forest ecosystem in the study area was a large pool of atmospheric Hg, and the average storage of Hg in forest soil (0 −80 cm depth) was 191.3 mg m^{-2}.

1. Introduction

Deposition of atmospheric mercury (Hg) to landscapes and watersheds plays an important role in the global biogeochemical cycling of Hg (Driscoll et al., 2007). Unlike other heavy metals, Hg can be released to the atmosphere in vapor form by both natural and anthropogenic sources. The levels of atmospheric mercury in the 1990s were approximately three times higher than those of before the industrial era (Bergan et al., 1999), and many studies indicated that deposition of atmospheric Hg is the major pathway for Hg entering into aquatic environmental and earth surfaces (Buehler and Hites, 2002; Rolfhus et al., 2003), where it is available for transformation to methyl mercury (MeHg) and poses a threat to human beings and wildlife health via food chain (Watras and Bloom, 1994; Feng et al., 2008).

There are three major forms of mercury in the atmosphere, namely gaseous elemental Hg (GEM), reactive gaseous mercury (RGM) and particulate bounded mercury (PBM) (Schroeder and Munthe, 1998; Lindberg and Stratton, 1998). Due to the low solubility of GEM in water, mercury in precipitation mainly comes from the scavenging of PBM and RGM in the atmosphere (Guentzel et al., 2001). Because of the large surface area of receptor sites on foliage, litterfall Hg fluxes represents a large portion of Hg dry deposition to forested landscapes of terrestrial ecosystems (Johnson and Lindberg, 1995; St. Louis et al., 2001). Deposition of PBM, adsorption of RGM onto plant surface, and stomatal uptake of GEM are

* Corresponding author. Tel.: +86 851 5895728.
  E-mail address: fengxinbin@vip.skleg.cn (X. Feng).

included in the Hg dry processes to forests (Lindberg et al., 1991, 1992). However, much higher concentrations of GEM in the atmosphere than other mercury species offset its lower deposition velocity, making it likely that the dry deposition flux of GEM via stomatal uptake constitutes a large component of total dry deposition.

China is the largest developing country worldwide. With the rapid development of economy during the past three decades, the amount of Hg emitted in China has increased significantly. Estimation of anthropogenic emissions of Hg in China was 696 ± 307 t in 2003, and emission of Hg from Southwest China was significant (Wu et al., 2006). Precipitation and litterfall are two major pathways for atmospheric Hg delivery to forest floor (St. Louis et al., 2001; Wang et al., 2009). A large number of studies have been carried out to investigate Hg deposition fluxes in remote areas in North America and Europe (e.g., Hultberg et al., 1995; Bishop and Lee, 1997; Poissant et al., 2005; Bushey et al., 2008). However, only a few long-term monitoring studies of Hg deposition fluxes have been performed in rural, semi-rural and urban/industrial areas of China. Wang et al. (2009) and Guo et al. (2008) have reported that THg concentrations in precipitation and direct wet deposition fluxes to Chinese suburban and semi-remote areas were both much higher than that of remote areas in Mt. Leigong and Mt. Gongga (Fu et al., 2010a,b). The characteristics of atmospheric mercury deposition and its subsequent cycling in forested catchments have been poorly studied and there is still a deficiency to adequately describe temporal and spatial deposition of Hg in China. Hence, it is very important to conduct long-term continuous measurements of Hg deposition fluxes in remote areas of China.

In this study, continuous measurements of THg in precipitation and litterfall at a high-altitude forest site, Southwest China, were monitored from June 2011 to May 2012. The major objectives of the present study are to quantify the atmospheric THg input fluxes by precipitation and litterfall in the summit of Mt. Ailao.

2. Materials and methods

2.1. Sites description

This study was conducted in the Xujiaba region (24°32′N, 101°01′E) – a protected virgin forest section covering 5100 ha on the northern crest of broad-leaved forest in Mts. Ailao (23°35′–24°44′ N, 100°54′–101°01′ E, Fig. 1). This forest has been recognized to be a montane moist evergreen broad-leaved primary forest according to the presence of large and old trees (Yang et al., 2008). The altitude ranges from 2450 to 2650 m a.s.l. The climate is mainly controlled by the Southwest monsoon, especially in the summer season with plenty of rainfall (85% of the total annual rainfall occurred in summer). In contrast, winter is controlled by dry and warm monsoon circulation and the climate is arid. Annual mean air temperature and rainfall in the study area are 11.3 °C and 1947 mm, respectively. Primary lithocarpus forest (PLF), which covers nearly 85% of the Xujiaba region, is the most extensive forest type in the study area (Young et al., 1992). According to a study conducted in 2011, the dominant tree species are Manglietia insignis, Lithocarpus chintungensis, Blueberry, Castanopsiswattii and Lithocarpus xylocarpus. The soil is typically yellow-brown earth and its texture is loam, with an acidic pH (4.2–4.9) (Yang et al., 2008).

The sampling site was located at the Research Station of Ailao Mountain Forest Ecosystems, Chinese Academy of Sciences. It was relatively isolated from large anthropogenic Hg sources and situated about 160 km to the southwest of Kunming, the capital of Yunnan Province. The nearest populated center is Jingdong County (Population: 36,500, 1200 a.s.l.), which is located at 20 km to the southwest.

2.2. Precipitation collection

Wet-only precipitation was collected from June 2011 to May 2012 at the wide-open site in the study area. Precipitation samples were collected by a modified automated wet-only precipitation collector with a bulk borosilicate glass bottle which replaced the plastic bucket. It is proven that borosilicate glass bottles have the lowest mercury blanks and don't absorb mercury (Landis and Keeler, 1997). The collector was approximately 1.5 m from the ground surface to avoid contamination from soil particles and far away from any obvious anthropogenic disturbances. A strict cleaning procedure was conducted using trace metal clean protocols. All Teflon bottles were cleaned rigorously by dipping in dilute acid (10% HNO_3), rinsing with ultrapure deionized water (18 MΩ cm), and then triple rinsed with ultrapure deionized water (18 MΩ cm) and finally, doubled-bagged, stored in a plastic boxes
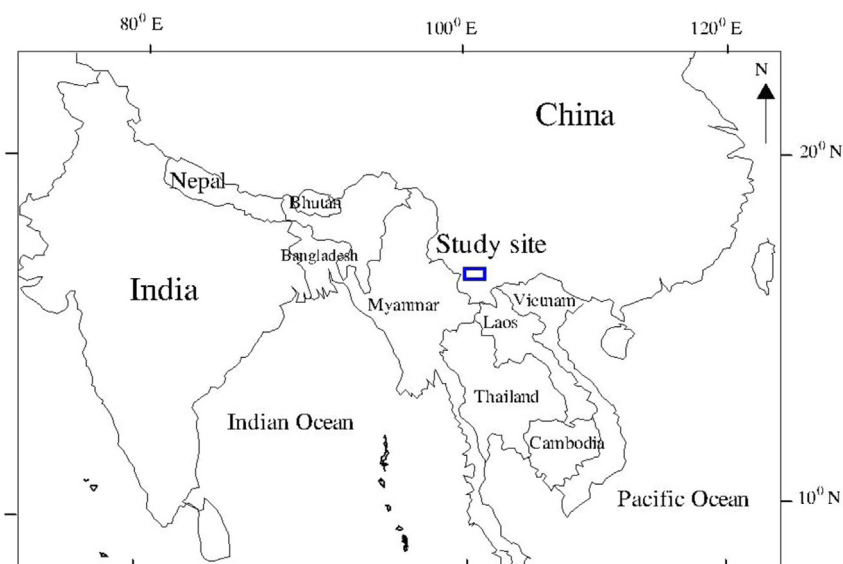

Fig. 1. The location of the study site.

until use. Each precipitation sample collected by the bulk borosilicate glass bottle were poured into pre-cleaned 250-ml Teflon bottles immediately after every rainfall event and divided into two subsamples. One of the subsamples was preserved by adding trace-metal grade HCl (to 0.5% of total sample volume), and the other subsample was used to measure anion concentrations without adding acids. Polyethylene gloves were used throughout the whole sampling campaign. PTEE bottles with samples were individually sealed into double successive polyethylene bags and packed in black plastic bags to avoid light, then shipped to the laboratory and stored in a refrigerator (0–4 °C) until analysis. Before each of the new sampling cycle, the sampling collectors were rinsed with ultrapure deionized water.

THg concentrations in precipitation samples were determined following US EPA Method 1631 (US EPA, 1999). THg concentrations in samples were analyzed via BrCl oxidation followed by $SnCl_2$ reduction, and dual amalgamation combined with CVAFS detection (US EPA, 1999). Reactive mercury (RHg) concentrations in precipitation were determined by directly $SnCl_2$ reduction, and dual amalgamation combined with CVAFS detection (US EPA, 1999). However, it should be noted that RHg is an operationally defined Hg species. Anion concentrations in precipitation samples were determined by Ion Chromatography (Model ICS-90, Dionex Ltd., USA).

The limits of detection for THg and RHg, based on three times the standard deviation of replicate measurements of a blank solution, were 0.03 and 0.01 ng L^{-1}, respectively. It was found that the instrumental blank was obviously under the detection limits in all cases. THg measured in reagent blank (0.08 ng L^{-1}) was subtracted from concentrations measured during the sample analysis. The average relative standard deviation on precision test for the duplicate analysis ($n = 5$) of THg and RHg was 8.5% and 5.4%, respectively, and the spike recoveries for THg and RHg were in the ranges of 87–111% and 93–109%, respectively.

2.3. Litterfall and soil sample collection

Modified litterfall collectors (1 m × 1 m) were developed to collect litterfall in the study areas. The forest canopy type and the density of forest are the two major factors controlling THg deposition fluxes (Witt et al., 2009) and specific surveys in ten tropical trees show that Hg concentration varies up to six folds, depending on the species (Millhollen et al., 2006). As we stated above, this forest is a montane moist evergreen broad-leaved primary forest and the most typical species of the trees in the study area cover nearly 85% of surface area in the region. Hence, five most representatively dominant species: Manglietia insignis (collection 1), Lithocarpus chintungensis (collection 2), Blueberry (collection 3), Castanopsiswattii (collection 4), Lithocarpus xylocarpus (collection 5) and three random species (collection 6, 7, 8) which were located under the other dominant and adjacent tree species and collected the mixtures of litterfall from different tree species, were selected to study the litterfall Hg deposition fluxes.

Litterfall samples were collected monthly at the same period with precipitation, packed into paper bags and air-dried in a clean environment of the Research Station of Ailao Mountain Forest Ecosystems. Monthly litterfall samples from each site were completely ground to a fine powder in a pre-cleaned food blender to analyze Hg concentrations in litterfall and calculate annual mass flux of each species. In addition, litterfall samples of June to August, September to November, December to February and March to May, which are representing summer, autumn, winter, and spring, respectively, were used to characterize the seasonal variation of litterfall deposition of THg. Litterfall samples were divided into leaves, twigs (below 2 cm in diameter), and reproductive material and debris (miscellaneous plant materials).

Table 1
Statistical summary of THg and RHg concentrations and deposition fluxes in precipitation and litterfall.

	Annual rainfall depth (mm)	THg concentration (ng L^{-1})	RHg concentration (ng L^{-1})	THg flux (µg m^{-2} yr^{-1})	RHg flux (µg m^{-2} yr^{-1})
Precipitation	1752	2.98	0.92	5.22	1.61
Litterfall	Litter Mass (g m^{-2} yr^{-1})	THg concentration (ng g^{-1})		THg flux (µg m^{-2} yr^{-1})	
Collector 1	1926	54		103.7	
Collector 2	1077	62		66.3	
Collector 3	899	43		38.4	
Collector 4	1346	57		76.4	
Collector 5	1421	54		77.3	
Collector 6	1471	49		71.4	
Collector 7	1114	54		60.0	
Collector 8	1320	57		75.8	
Annual avg	1322	54		71.2	

THg distributions in soil profile were monitored at two sites in the study area. S1 was located at a meadow zone without litterfall deposition and S2 was located at dark ever-green forest with a litter layer (2–5 cm in thickness) and both sites were selected at random. Samples in the depth profiles were collected every 5 cm from top layer to the depth of 80 cm. Soil samples were collected in polyethylene bags using polyethylene gloves and air-dried in a clean environment in the laboratory. Subsequently the air-dried soil samples were homogenized to a size of 150 meshes per inch with a mortar before chemical analysis.

A Lumex RA-915 + multifunctional mercury analyzer (Lumex Ltd., Russia) equipped with a pyrolysis attachment was used for measuring THg concentrations in plant and soil samples. Samples were thermally decomposed in an atomizer chamber at 800 °C with aided catalytic action, and then Hg^0 was detected by the RA-915 + analyzer (Rodriguez et al., 2007; Shuvaeva et al., 2008). Each sample was measured twice and the mean concentrations were used in this study. Instrument calibration curves covering the appropriate concentrations were confirmed by soil standards (IGGE IRMA China, 290 ng g^{-1}) and citrus leaf standards (GBW10020, 150 ng g^{-1}), and checked by the control standard samples every six samples. Detection limits were 0.5 ng g^{-1} for solid samples. The precisions, obtained from nine duplicated measurements of the standards, were 8.8% for soil and 4.0% for plant. The recoveries of certified standards ranged from 92% to 113% for soil materials and 94%–103% for plant samples. And the C and N of litterfall were determined by PE2400-II Element Analyzer.

The storage of Hg in soil (M_{Hg}) is estimated on the basis of the soil bulk density (d_a) (IMPACTS, 2004) and soil Hg concentrations (c) (Liu et al., 2003). Soil samples were collected by a cutting ring sampler with known volume (v) and then dried in an oven at 105 °C to constant weight (s). Thus, M_{Hg} was calculated according to the following formula (1):

$$M_{Hg} = \sum_{i=1}^{n} c_i \times d_{ai} = \sum_{i=1}^{n} c_i \times s_i/v_i \qquad (1)$$

Total Organic Matter (TOM) content in forest soil was calculated by using the sequential loss on ignition (LOI) (Fu et al., 2010a). A homogenized soil sample (WS) was dried at 105 °C for about 12–24 h to obtain the dry weight of the samples (DW_{105}). The heated dry sample was then burned at 550 °C for 4 h and the weight of the sample after heating at 550 °C was DW_{550}. Thus, the TOM content (LOI_{550}) was calculated according to the following formula (2):

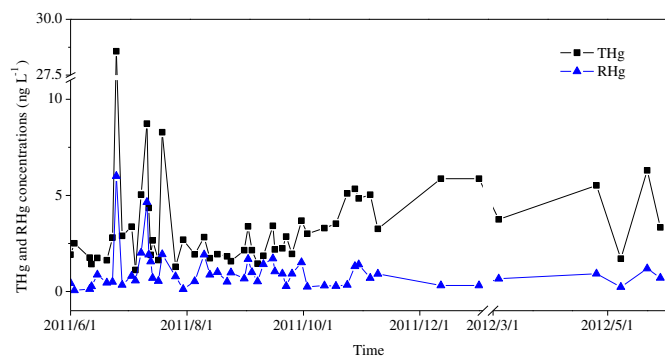$$LOI_{550} = 100(DW_{105} - DW_{550})/WS \qquad (2)$$

Fig. 2. Every event concentrations of THg and RHg in precipitation collected in Mt. Ailao from June 2011 to May 2012.

3. Results and discussion

3.1. Mercury in precipitation

In total we collected 51 individual precipitation event samples from June 2011 to May 2012. No sample was collected in January and February 2012 due to the lack of precipitation events.

3.1.1. THg concentrations in precipitation

The concentrations and deposition fluxes of THg and RHg in precipitation are listed in Table 1. THg concentrations in precipitation ranged from 1.12 to 28.6 ng L^{-1} (Fig. 2), with an overall volume-weighted mean concentration of 2.98 ng L^{-1}. THg concentrations in precipitation in the study site were lower than those reported in North America and Europe (Guentzel et al., 2001; Vanarsdale et al., 2005; Keeler et al., 2006). The levels of THg in the study area were also lower than those observed in other remote areas of China (Guo et al., 2008; Fu et al., 2010a,b; Wang et al., 2009). However, it is interesting that the annual geometric mean of GEM concentrations at the study site was about 2.08 ng m^{-3} (our unpublished data), which are higher than those observed from different remote areas in Europe and North America (generally lower than 2.0 ng m^{-3}, Travnikov, 2005; Kim et al., 2005). This result suggests that atmospheric GEM has a less immediate effect on THg in rainwater compared to PBM and RGM. Our sampling site was relatively isolated from direct anthropogenic Hg emissions sources (e.g., incinerators, non-ferrous metal smelters and power plants), so that the primary sources of RGM and PBM are limited, which resulted in that THg concentrations in precipitation were lower than that in Europe and North America and remote areas of China. These airborne Hg fractions are transients in atmospheric boundary layer and have a limited role in the corresponding long-range transport. The other factor is that GEM is very stable in atmosphere and is able to undergo a long range transport from emission sources and cause elevated levels at the sampling site (Schroeder and Munthe, 1998). This may be the reason that the THg concentration in precipitation was lower and the concentration of GEM was higher in Mt. Ailao.

A clear seasonal pattern of THg concentrations in precipitation was observed, with highly elevated mean volume-weighted THg concentration (4.2 ng L^{-1}) observed in dry season (winter and spring) and relatively lower mean volume-weighted concentration (3.05 ng L^{-1}) in rainy season (summer and fall), which may be attributed to the dilution factor of the higher precipitation in wet season when PBM and RGM are scavenged from the atmosphere. This finding was in contrast to observations in North America which showed relatively higher THg concentration in summer months (Choi et al., 2008).

Table 2
Hg (ng L^{-1}) and anion (mol L^{-1}) concentration in precipitation in Mt. Ailao from June 2011 to May 2012.

	Min–max	Volume-weighted mean	Mean	SD
THg	1.12–28.57	2.98	3.68	3.92
RHg	0.06–4.64	0.92	1.01	0.91
F$^-$	0–0.0104	0.0009	0.0013	0.0018
NO$_2^-$	0–0.0002	0.0001	0.0001	0
Cl$^-$	0.0017–0.0338	0.0062	0.0080	0.0076
NO$_3^-$	0.0008–0.0903	0.0103	0.0142	0.0186
SO$_4^{2-}$	0.0009–0.0899	0.0081	0.0132	0.0190
PO$_4^{3-}$	0–0.0024	0.0004	0.0004	0.0005

3.1.2. RHg and major anion concentrations in precipitation

The statistical summaries of the RHg and anion concentrations of precipitation measured at Mt. Ailao are listed in Table 2. The concentration ratio of RHg to THg ranged from 2% to 76%, with a median value of 32%, which was much lower than that of 62% and 68% reported by Mason et al. (1992) and Lamborg et al. (1999) in North America. Due to low solubility of GEM in precipitation, mercury in rainwater mainly comes from the scavenging of PBM and RGM in the atmosphere (Guentzel et al., 2001). The lower ration of RHg in the rainwater sample could suggest that a large remaining fraction of the Hg in the rainwater sample was particulate bound in this study. And the nearest the source of particulate matter may be derived from the Jingdong County. It is revealed that there was a significantly positive correlation between concentrations of RHg and THg ($r = 0.799$, $p < 0.001$; $n = 51$), which is consistent with the observation that a linear correlation between mean concentrations of THg and RHg in rainwater was observed across large geographic areas in North America areas (Hammerschmidt et al., 2007). It was also suggested that RGM is regarded as the main source for RHg in precipitation while RHg is a pollutant of concern primarily due to its ability to be transformed into MeHg in the environment (Sakata and Marumoto, 2005).

The major anions in precipitation are sulfate, nitrate and chloride, which constituted 94.8% of total molar concentration. No significant correlations were observed between RHg and sulfate as well as chloride. However, a significantly positive correlation between RHg and nitrate ($r^2 = 0.4313$, $p < 0.001$; $n = 51$) (Fig. 3) was obtained. Nitrate in precipitation is mainly from nitrogen oxide in the atmosphere and lightening discharges, stratosphere–troposphere exchange, soil microbial emissions and biomass burning are considered as the major sources of NO$_x$ to the atmosphere (Prasad et al., 2000). Significantly positive correlations indicate that RHg
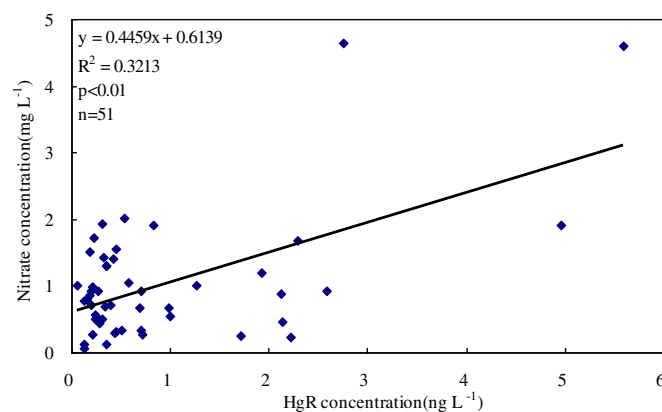

Fig. 3. The correlation between concentrations of RHg and concentrations of nitrate in precipitation in Mt. Ailao from June 2011 to May 2012.
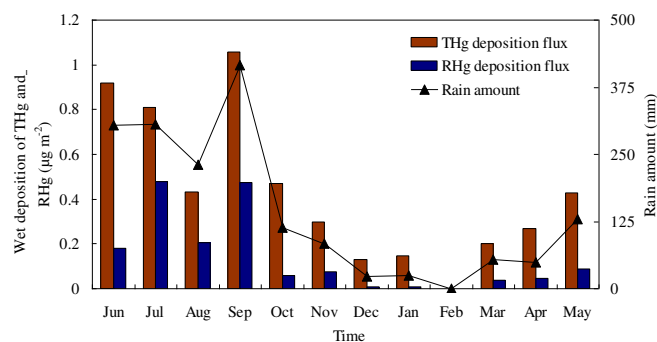
Fig. 4. Monthly wet deposition fluxes of THg, RHg and precipitation amounts in Mt. Ailao from June 2011 to May 2012.

and nitrate in precipitation may have similar sources in Mt. Ailao. However, the other anions in precipitation indicated that other processes are also contributing. In this area, precipitation in summer accounts for 85% of annual precipitation and most of precipitation comes from the southeast monsoon. Therefore, pollutants such as mercury in the precipitation may be originated from Southern Asia. Besides, through Asian winter monsoon, the number and the extent of emission sources from Guizhou province, where high anthropogenic emissions were reported from coal burning (with high Hg content coal), non-ferrous metal smelting activities and artisanal Hg mining activities (Feng and Qiu, 2008; Li et al., 2009), also plays an important role. These results indicated that the air masses with relatively high precipitation amounts that pass over these Hg sources may result in elevated Hg wet deposition.

3.1.3. Wet deposition flux

Wet deposition fluxes of THg and RHg were estimated using the volume-weighted concentrations and depths of precipitation. The annual wet deposition fluxes of THg and RHg was 5.22 and 1.61 $\mu g\ m^{-2}\ yr^{-1}$, respectively. A clear seasonal variation with higher THg and RHg wet deposition fluxes in rainy season was observed (Fig. 4). THg and RHg deposition fluxes in wet seasons (from May to October) were 4.1 and 1.31 $\mu g\ m^{-2}\ yr^{-1}$, accounting for 79.8% and 83.5% of the total annual wet deposition fluxes for THg and RHg, respectively. This is in contrast with the seasonal variations of THg concentrations, indicating variations of wet deposition fluxes were mainly regulated by rainfall.

A comparison of THg wet deposition fluxes between Mt. Ailao and other remote areas is given in Table 4. Wet deposition fluxes of THg in this study were comparable to those observed from forests in some remote areas of North America and Europe, and slightly lower than other remote areas in south China. However, the THg wet deposition flux of 9.3 $\mu g\ m^{-2}\ yr^{-1}$ in all MDN sites (Mercury Deposition Network, 2007), was higher than that found in this study. This is mainly because significantly elevated THg concentrations (9.3 $ng\ L^{-1}$) in precipitation samples were observed at all MDN sites.

3.2. Mercury in litterfall

3.2.1. Litterfall deposition fluxes and variation of Hg concentrations in different tissues of litterfall

The annual litterfall deposition flux in the study areas was 1.32 $mg\ m^{-2}\ yr^{-1}$, with the monthly deposition fluxes varying from 0.043 $g\ m^{-2}$ to 0.235 $g\ m^{-2}$, which were much higher compared to some deciduous forests in North America (Sheehan et al., 2006; Bushey et al., 2008) and tropical forests in Brazil (Silva-Filho et al., 2006; Teixeira et al., 2012). Leaf tissue constitutes 67.1% of total mass of litterfall, and twig tissue and the reproductive material and debris account for 19.8% and 13.1% of the total mass of litterfall, respectively. Generally, THg concentrations in leave tissues were significantly higher than twigs as well as other fractions ($p < 0.01$ and $p < 0.05$, respectively), and the same pattern was observed for the deposition fluxes ($p < 0.05$ for both).

3.2.2. Litterfall Hg concentration and correlation with litter quality

THg concentrations in litterfall were in the ranges of 43–62 $ng\ g^{-1}$ (dry weight, Table 1). Litter quality can be characterized by the C/N ratio representing the balance between potentially limiting energy and nutrient resources for microbial communities (Gower and Son, 1992; Berg and Laskowski, 2006). In this study, we found that the C/N ratios of litterfall showed a significantly negative correlation with litterfall Hg concentrations (Fig. 5). The relative proportion of twig versus leaf tissues in the sample controlled the C/N to Hg correlations. Table 3 showed that THg concentrations in leave tissues were significantly higher than twigs and moreover, W. Liu et al. (2002) found that C/N ratios were much higher in twigs than in leaves in this study area of Mt. Ailao. Thus, the negative correlation can be explained as lower Hg concentrations in samples with higher twig tissue content which have been shown to have lower Hg concentrations and higher C/N ration compared to leaf tissue. Conversely, litterfall samples with higher proportion of leaf tissue to twig tissue have a higher mercury concentration and lower C/N ration.

3.2.3. Seasonal variation of litterfall deposition fluxes

Monthly variations of THg concentrations and deposition fluxes of litterfall in Mt. Ailao are shown in Fig. 6. The mean THg concentration in litterfall was 54 $ng\ g^{-1}$ (monthly means varied from 46 to 63 $ng\ g^{-1}$). Generally, the mean THg concentrations in litterfall in Mt. Ailao were much lower compared to some suburban and rural sites in China, such as Tieshanping in Chongqing city (THg:105 $ng\ g^{-1}$, Wang et al., 2009) and Leigongshan in Guizhou province (THg: 91 $ng\ g^{-1}$, Fu et al., 2010b). This is because the sampling site was isolated from sites near human activities. However, the mean THg concentrations in litterfall in the study area were slightly higher than those observed in North America and European temperate and boreal forests as shown in Table 4. A previous study by Fay and Gustin (2007) reported that plant foliage can accumulate Hg from the air and that the air Hg level was a good indicator of leaf Hg concentration, especially for deciduous plants. It is shown that total gaseous mercury (TGM) concentrations in

Table 3
Seasonal statistical summary of THg weighted concentration and deposition fluxes of different litterfall fractions in Mt. Ailao.

	THg concentration ($ng\ g^{-1}$)			Biomass (g)			THg deposition ($\mu g\ m^{-2}\ yr^{-1}$)		
	Leaves	Twigs	Others	Leaves	Twigs	Others	Leaves	Twigs	Others
Spring	69	27	33	367.05	367.05	72.77	25.3	9.9	2.4
Summer	65	37	43	173.97	173.97	42.15	11.3	6.4	1.8
Autumn	59	43	52	250.99	250.99	54.78	14.8	10.8	2.8
Winter	75	32	65	108.77	108.77	80.52	8.2	3.5	5.2

Table 4
Comparison of precipitation (ng L^{-1}) and litterfall (ng g^{-1}) Hg concentration and Hg deposition fluxes (µg m^{-2} yr^{-1}) in Mt. Ailao with literature data at other remote areas worldwide.

Precipitation site	Period		Concentration	Flux	Reference
Hokkaido, Japan	2002.12–2003.11	Precipitation	5.2	5.8	Sakata and Marumoto, 2005
Britain	2006	Precipitation	1.0	4.3	Rowland et al., 2010
Germany	1998.4–1999.4	Precipitation	18	35	Schwesig and Matzner, 2000
Ontario, Canada	1992–1996	Precipitation	5.7	3.6	Graydon et al., 2009a,b
MDN	2006	Precipitation	9.2	9.3	NADP, 2007
Mt. Leigong, China	2008.5–2009.5	Precipitation	4	6.1	Fu et al., 2010a
Mt. Gongga, China	2005.5–2007.4	Precipitation	14.3	26.1	Fu et al., 2010b
Mt. Ailao, China	2011.6–2012.5	Precipitation	2.98	5.4	This work
Guyana, French	1999–2001	Litterfall	64	45	Melieres et al., 2003
Bavaria, Germany	1998.9–1999.9	Litterfall	70	15.1	Schwesig and Matzner, 2000
Ontario, Canada	1995–1996	Litterfall	32	88	St. Louis et al., 2001
USA (23 sites)	2007–2009	Litterfall	41.1	12.3	Risch et al., 2011
Atlantic coast, Brazil	1997.1–12	Litterfall	131	122	Silva-Filho et al., 2006
Mt. Leigong, China	2008.5–2009.5	Litterfall	91	39.5	Fu et al., 2010a
Mt. Gongga, China	2005.5–2007.4	Litterfall	35.7	35.5	Fu et al., 2010b
Mt. Ailao, China	2011.6–2012.5	Litterfall	54	71.2	This work

ambient air at remote areas in North America and Europe are slightly lower than that of in this study, suggesting that TGM concentrations in ambient air might be responsible for the variation in foliar Hg concentrations. However, some other factors such as vegetation types may also correspond to variation of foliar THg concentrations at different areas.

The annual litterfall deposition flux of THg was 71.2 µg m^{-2} yr^{-1}. The Litterfall deposition fluxes of THg in the study area were much higher than the values observed from forests in North America and Europe (Lee et al., 2000; Schwesig and Matzner, 2000; Larssen et al., 2008; Graydon et al., 2009a; Risch et al., 2011). The highly elevated deposition via litterfall constituted a very important source of Hg in forest ecosystem. Previous study suggested that uptake of Hg by foliage via stomatal and nonstomatal pathways could be enhanced by elevated atmospheric Hg concentrations (Stamenkovic and Gustin, 2009) and atmospheric Hg^0 is almost the exclusive source of Hg in vegetation leaf (Zhang et al., 2006). Leaves may scavenge Hg from the atmosphere as PBM (Iverfeldt, 1991) or RGM species (Lindberg, 1996). However, lower THg concentrations in precipitation compared to other areas (Table 4) may indicate that PBM and RGM were lower in Mt. Ailao. Therefore, we attributed the elevated litterfall Hg deposition fluxes to the elevated GEM concentrations in ambient air in the study area. Rea et al. (2000) suggested that all or a large portion of the PBM and RGM scavenged by leaves is believed to wash off foliage surfaces in throughfall,

indicating that throughfall is another primary deposition pathway of Hg to forested ecosystems. Some studies reported that mercury dry deposition caused by throughfall was about 2 times higher than precipitation in Southwestern China (Fu et al., 2010a,b). Assuming the throughfall deposition fluxes of this research similar to previous studies, dry deposition by throughfall was approximately accounted for 10% of the total deposition. Hence, more research is urgently needed to examine mercury deposition by throughfall in this study area.

Therefore, highly elevated litterfall deposition fluxes may suggest that in remote forest areas of China, deposition of atmospheric Hg^0 via uptake by vegetation leaf was the major pathway for the depletion of atmospheric Hg. Besides, these fluxes were also higher than some polluted areas of China (Wang et al., 2009; Fu et al., 2010a, b). This is in contrast to litterfall THg concentrations and the elevation of litterfall THg deposition fluxes were mainly resulted from the highly elevated annual litter deposition fluxes.

As shown in Fig. 6, a seasonal pattern in deposition fluxes of THg in litterfall was observed. Generally, the highest litterfall deposition fluxes of THg was observed in spring (March, April, May) (30.5 µg m^{-2}), followed by fall (September, October, November) (18.7 µg m^{-2}), summer (June, July, August) (12.3 µg m^{-2}) and winter (December, January, February) (9.7 µg m^{-2}). This is consistent with tropical seasonal forest in Southwestern China where litterfall production peak occurred during the hot and dry season (January–February) (Tang et al., 2010). However, this is in contrast with North America where Maximum monthly rates of leaf litterfall occurred in the fall (October to December) (Cormier et al., 2013), indicating the variations of litterfall deposition flux were regulated by different ecosystems and climates. Significant correlations between biomass fluxes and litterfall deposition fluxes of THg was observed ($r_{THg} = 0.77$, $p < 0.01$), indicating that litterfall deposition fluxes was mainly controlled by the biomass flux of litter in the study area. Previous studies have demonstrated that differences in vegetation/canopy types may influence the capture of deposited mercury (Demurs et al., 2007; Witt et al., 2009). However, our observation suggested that Hg flux via litterfall was driven primarily by the biomass flux of litter, despite of differences in Hg concentrations among different litter types in remote forest areas of China.

3.3. Mercury in soil profile

THg and TOC concentrations in the two soil profiles in the forest of Mt. Ailao are shown in Fig. 7. The highest THg concentrations were observed at the topsoil layer and THg concentrations
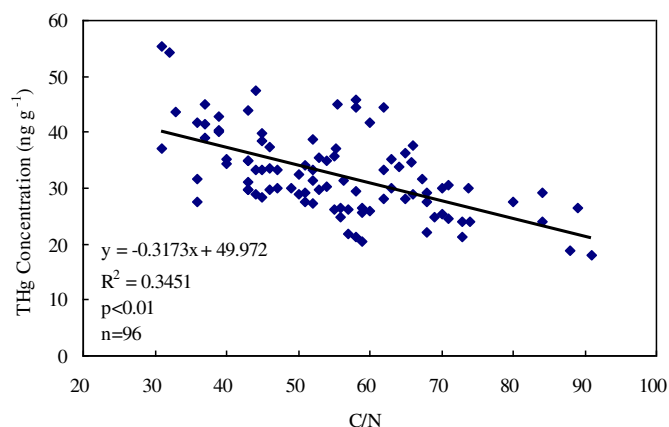
Fig. 5. The correlation between litterfall C/N ratio and THg concentration in litterfall in the forest of Mt. Ailao from June 2011 to May 2012.
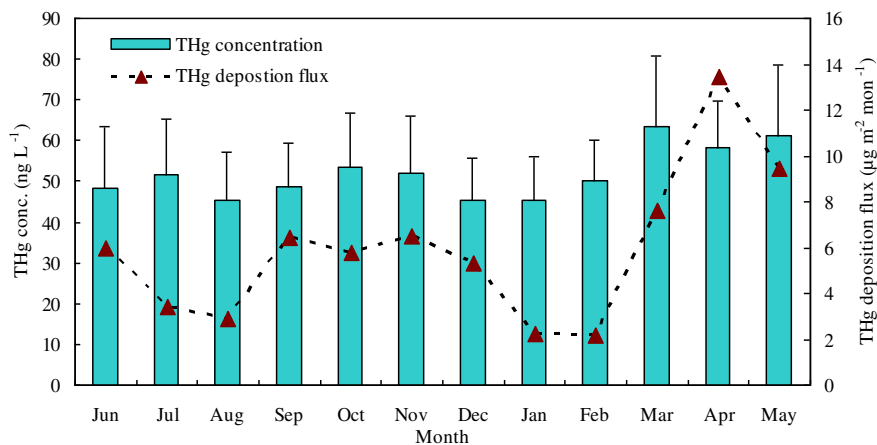
y = -0.3173x + 49.972
R^2 = 0.3451
p<0.01
n=96

Fig. 6. Monthly mass-weighted mean concentrations of THg and deposition fluxes of litterfall in the forest of Mt. Ailao from June 2005 to May 2007.

in soil decreased significantly with depth. Below the depth of 20 cm, THg concentration did not change obviously. THg concentrations in soil profile of S1 in the meadow range from 0.016 to 0.054 mg kg^{-1}. In contrast, THg concentrations in the soil profile of S2 in the forest range from 0.118 to 0.279 mg kg^{-1}. We observed significant correlations between THg and TOC concentrations in soil at both sampling sites (r^2 = 0.74 and 0.69 at S1 and S2, respectively, $p < 0.01$ for both), indicating that Hg was absorbed by the organic matters. It's interesting to note that mercury concentration in the upland soil of S2 was much higher than that of S1, and the maximum concentration occurred at 0–5 cm depth, then the second peak appeared at 20–25 cm (Fig. 7). The first peak might be caused by the adsorption of Hg by the organic matters and the second peak might be caused by the eluviation of the Hg, which is consistent with the previous studies of Liu et al. (2003) in Xiaoxing'an Mountain, northeastern China and Selvendiran et al. (2008) in two northern temperate forest wetlands, USA. The mercury concentrations at 0–20 cm surface soil of S1 were much higher than that in the deeper soil, indicating that mercury

concentrations in soil have affected by atmospheric wet deposition. Wiener et al. (2006) suggested that highly elevated THg in forest surface soil was mostly likely attributed to atmospheric depositions via litterfall and throughfall, whereas the contribution from geological sources was relatively smaller. We observed significant and consistent difference in soil THg concentrations and TOC content at the two sites ($p < 0.01$), with THg concentrations in forest soil profile 5 folds higher than those at the open air site. This may indicate that the upland forest soils act as sinks for Hg and that dry deposition of Hg was the major pathway for Hg loading to the forest catchment.

THg concentrations in topsoil in the studied forest were comparable to the data reported from other forest sites in China (S.L. Liu et al., 2002; Wang et al., 2009), but higher than those measured in North America (St. Louis et al., 2001; Biswas et al., 2008). This is in a good agreement with the elevated loading of THg in the study area. The storage of Hg in soil (0–80 cm depth), calculated by the above Formula (1), was 27.9 mg m^{-2} for meadow (S2), which was much less than that for the forest (S1) with a value of 191.3 mg m^{-2}.
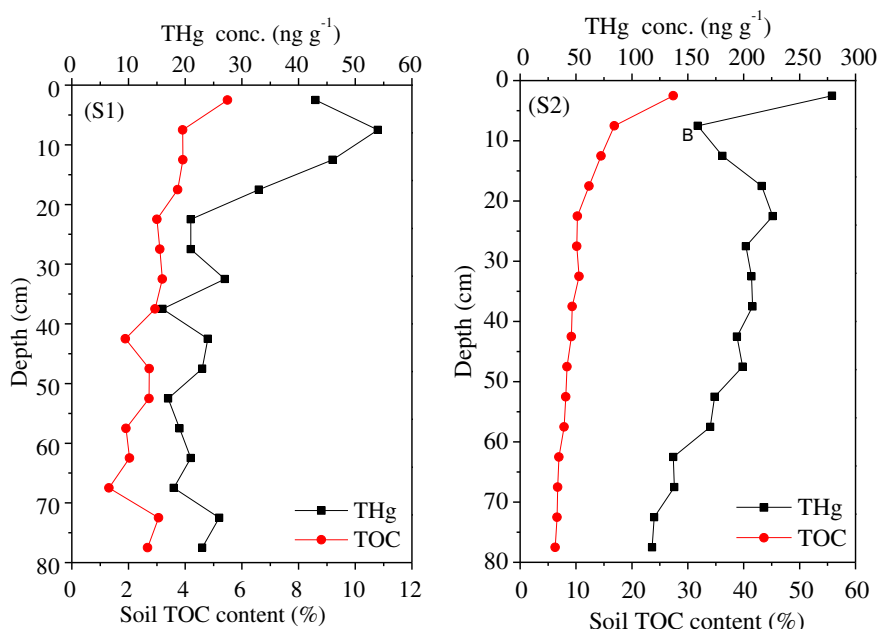

Fig. 7. Hg distribution in soil profile at meadow (S1) and forest (S2) sites in the Mt. Ailao.

4. Conclusion

The purpose of this study was to estimate the wet and litterfall deposition of THg to the soils of the Mt. Ailao broad-leaved and old-growth subtropical forest, respectively, in Southwest China during the period from June 2011 to May 2012. This canopy contribution originates mainly from the atmospheric background load of Hg. Annual volume-weighted mean THg and RHg concentrations in precipitation were 2.98 and 0.92 ng L^{-1}, respectively. We observed a significantly positive correlation between the concentrations of RHg and nitrate in precipitation. Annual litterfall mass-weighted mean THg concentrations were 52 ng g^{-1}, which varied a little among different vegetation species in the forests. The C/N of litterfall showed a significantly negative correlation with litterfall THg concentration.

The annual total deposition fluxes of THg reached 76.7 μg m^{-2} yr^{-1}. Litterfall deposition was the major pathway for Hg loading to the upland forest catchment, which were 71.2 μg m^{-2} yr^{-1} (about 92.8% of total input for THg). The pools of THg in the forest soils was much higher than those reported in remote forest catchments in Europe and North America.

Acknowledgments

References

Berg, B., Laskowski, R., 2006. Litter decomposition: a guide to carbon and nutrient turnover. Adv. Ecol. Res. 38, 20—71.

Bergan, T.L., Gallardo, L., Rodhe, H., 1999. Mercury in the global troposphere: a three-dimensional model study. Atmos. Environ. 33, 1575—1585.

Bishop, K., Lee, Y., 1997. Catchments as a source of mercury/methylmercury in boreal surface waters. Metal Ions Biol. Syst. 34, 113—130.

Biswas, A., Blum, J.D., Keeler, G.J., 2008. Mercury storage in surface soil in a central Washington forest and estimated release during the 2001 Rex Creek fire. Sci. Total Environ. 404, 129—138.

Buehler, S.S., Hites, R.A., 2002. The Great Lakes' integrated atmospheric deposition network. Environ. Sci. Technol. 36, 354—359.

Bushey, J.T., Nallana, A.G., Montesdeoca, M.R., Driscoll, C.T., 2008. Mercury dynamics of a northern hardwood canopy. Atmo. Environ. 42, 6905—6914.

Choi, H.D., Sharac, T.J., Holsen, T.M., 2008. Mercury deposition in the Adirondacks: a comparison between precipitation and throughfall. Atmos. Environ. 42, 1818—1827.

Cormier, N., Krauss, K.W., Conner, W.H., 2013. Periodicity in stem growth and litterfall in tidal freshwater forested wetlands: influence of salinity and drought on nitrogen recycling. Estuaries Coasts 36, 533—546.

Demurs, J.D., Driscoll, C.T., Fahey, T.J., Yavitt, J.B., 2007. Mercury cycling in litter and, soil in different forest types in the Adirondack region, New York, USA. Ecol. Appl. 17, 1341—1351.

Driscoll, C., Han, Y., Chen, C., Evers, D., Lambert, K., Holsen, T., Kamman, N., Munson, R., 2007. Mercury contamination in forest and freshwater ecosystems in the Northeastern United States. Bioscience 57, 17—28.

Fay, L., Gustin, M., 2007. Assessing the influence of different atmospheric and soil mercury concentrations on foliar mercury concentrations in a controlled environment. Water Air Soil Pollut. 181, 373—384.

Feng, X., Li, P., Qiu, G., Wang, S., Li, G., Shang, L., Meng, B., Jiang, H., Bai, W., Li, Z., Fu, X., 2008. Methylmercury exposure through rice intake to inhabitants in Wanshan mercury mining area in Guizhou, China. Environ. Sci. Technol. 42, 326—332.

Feng, X., Qiu, G.L., 2008. Mercury pollution in Guizhou, China — an overview. Sci. Total Environ. 400, 227—237.

Fu, X.W., Feng, X.B., Dong, Z.Q., Yin, R.S., Wang, J.X., Yang, Z.R., et al., 2010b. Atmospheric gaseous elemental mercury (GEM) concentrations and wet and dry deposition of mercury at a high-altitude mountain peak in south China. Atmos. Chem. Phys. 10, 2425—2437.

Fu, X.W., Feng, X.B., Zhu, W.Z., Rothenberg, S., Yao, H., Liu, N., et al., 2010a. Elevated atmospheric deposition and dynamics of mercury in a remote upland forest of Southwestern China. Environ. Pollut. 158, 2324—2433.

Gower, S.T., Son, Y., 1992. Differences in soil and leaf litterfall nitrogen dynamics for five forest plantations. Soil Sci. Soc. Am. J. 56, 1959—1966.

Graydon, J.A., Emmerton, C.A., Lesack, L.F.W., Kelly, E.N., 2009a. Mercury in the Mackenzie River delta and estuary: concentrations and fluxes during open-water conditions. Sci. Total Environ. 407, 2980—2988. http://dx.doi.org/10.1016/J.SCITOTENV.2008.12.060.

Graydon, J.A., St. Louis, V.L., Hintelmann, H., Lindberg, S.E., Sandilands, K.A., Rudd, J.W.M., et al., 2009b. Long-term wet and dry deposition of total and methyl mercury in the remote boreal ecoregion of Canada. Environ. Sci. Technol. 42, 8345—8351.

Guentzel, J.L., Landing, W.M., Gill, G.A., Poliman, C.D., 2001. Processes influencing rainfall deposition of mercury in Florida. Environ. Sci. Technol. 35, 863—873.

Guo, Y.N., Feng, X.B., Li, Z.G., He, T.R., Yan, H.Y., Meng, B., et al., 2008. Distribution and wet deposition fluxes of total and methyl mercury in Wujiang reservoir Basin, Guizhou, China. Atmos. Environ. 42, 7096—7103.

Hammerschmidt, C.R., Lamborg, C.H., Fitzgerald, W.F., 2007. Aqueous phase methylation as a potential source of methylmercury in wet deposition. Atmos. Environ. 41, 1663—1668. http://dx.doi.org/10.1016/j.atmosenv.2006.10.032.

Hultberg, H., Munthe, J., Iverfeldt, A., 1995. Cycling of methyl mercury and mercury responses in the forest root catchment to three years of decreased atmospheric deposition. Water Air Soil Pollut. 80, 415—424.

IMPACTS, 2004. Integrated monitoring program on Acidification of Chinese Terrestrial Systems, Annual Report, Results 2003. In: Larssen, T., Tang, D. (Eds.), NIVA Report No. 4905-2004, ISBN 82-577-4594-4.

Iverfeldt, A., 1991. Mercury in forest canopy throughfall water and its relation to atmospheric deposition. Water Air Soil Pollut 56, 553—564.

Johnson, D.W., Lindberg, S.E., 1995. The biogeochemical cycling of Hg in forests: alternative methods for quantifying total deposition and soil emission. Water Air Soil Pollut. 80, 1069—1077.

Keeler, G.J., Landis, M.S., Norris, G.A., Christianson, E.M., Dvonch, J.T., 2006. Sources of mercury wet deposition in eastern Ohio, USA. Environ. Sci. Technol. 40, 5874—5881.

Kim, K.H., Ebinghaus, R., Schroeder, R., Blanchard, P., Kock, H.H., Steffen, A., Froude, F.A., Kim, M.Y., Hong, S.M., Kim, J.H., 2005. Atmospheric mercury concentrations from several observatory sites in the Northern Hemisphere. J. Atmos. Chem. 50, 1—24.

Lamborg, C.H., Rolfhus, K.R., Fitzgerald, W.F., Kim, G., 1999. The atmospheric cycling and air-sea exchange of mercury species in the South and equatorial Atlantic Ocean. Deep Sea Res. Part II Top. Stud. Oceanogr. 46, 957—977.

Landis, M.S., Keeler, G.J., 1997. Critical evaluation of a modified automatic wet-only precipitation collector for mercury and trace element determinations. Environ. Sci. Technol. 31, 2610—2615.

Larssen, T., de Wit, H., Wiker, M., Halse, K., 2008. Mercury budget of a small forested boreal catchment in southeast Norway. Sci. Total Environ. 404, 290—296. http://dx.doi.org/10.1016/j.scitotenv.2008.03.013.

Lee, Y., Bishop, K., Munthe, J., 2000. Do concepts about cycling of methylmercury and mercury in boreal catchments stand the test of time? Six years of atmospheric inputs and runoff exports at Svartberget, northern Sweden. Sci. Total Environ. 260, 11—20.

Li, P., Feng, X.B., Shang, L.H., Qiu, G.L., Meng, B., Liang, P., Zhang, H., 2009. Mercury pollution from artisanal mercury mining in Tongren, Guizhou, China. Atmos. Environ. 23, 2055—2064.

Lindberg, S.E., 1996. Forests and the global biogeochemical cycle of mercury. In: Baeyens, W., Ebinghaus, R., Vasiliev, O. (Eds.), Global and Regional Mercury Cycles: Sources, Fluxes and Mass Balances, NATO-ASI Series, vol. 21. Kluwer Academic Publishers, Dordrecht, The Netherlands, pp. 359—380.

Lindberg, S.E., Meyers, T.P., Taylor, G.E., Turner, R.R., Schroeder, W.H., 1992. Atmosphere surface exchange of mercury in a forest: results of modeling and gradient approaches. J. Geophys. Res. 97, 2519—2528.

Lindberg, S.E., Stratton, W.J., 1998. Atmospheric speciation concentrations and behavior of reactive gaseous mercury in ambient air. Environ. Sci. Technol. 32, 49—57.

Lindberg, S.E., Turner, R.R., Meyers, T.P., Taylor, G.E., Schroeder, W.H., 1991. Atmospheric concentrations and deposition of Hg to a deciduous forest at Walker Branch Watershed, Tennessee, USA. Water Air Soil Pollut. 56, 577—594.

Liu, R., Wang, Q., Lu, X., Fang, F., Wang, Y., 2003. Distribution and speciation of mercury in the peat bog of Xiaoxing'an Mountain, northeastern China. Environ. Pollut. 124, 39—46.

Liu, S.L., Nadim, F., Perkins, C., Carley, R.J., Hoag, G.E., Lin, Y.H., et al., 2002. Atmospheric mercury monitoring survey in Beijing, China. Chemosphere 48, 97—107.

Liu, W., Fox, J.E.D., Xu, Z., 2002. Litterfall and nutrient dynamics in a montane moist evergreen broad-leaved forest in Ailao Mountains, SW China. Plant Ecol. 164, 157—170.

Mason, R.P., Fitzgerald, W.F., Vandal, G.M., 1992. The sources and composition of mercury in Pacific Ocean rain. J. Atmos. Chem. 14, 489—500.

Melieres, M.A., Pourchet, M., Charles-Dominique, P., Gaucher, P., 2003. Mercury in canopy leaves of French Guiana in remote areas. Sci. Total Environ. 311, 261—267.

Millhollen, A.G., Gustin, M.S., Obrist, D., 2006. Foliar mercury accumulation and exchange for three tree species. Environ. Sci. Technol. 40, 6001—6006.

National Atmospheric Deposition Program, 2007. 2006 Annual Summary. Mercury Deposition Network, pp. 11—14. http://nadp.sws.uiuc.edu/lib/data/2006as.pdf.

Poissant, L., Pilote, M., Beauvais, C., Constant, P., Zhang, H.H., 2005. A year of continuous measurements of three atmospheric mercury species (GEM, RGM and Hg-P) in southern Québec, Canada. Atmos. Environ. 39, 1275—1287.

Prasad, V.K., Gupta, P.K., Sharma, C., Sarkar, A., Kant, Y., Badarinath, K.V.S., Rajagopal, T., Mitra, A.P., 2000. NOₓ emissions from biomass burning of shifting cultivation areas from tropical deciduous forests of India — estimates from ground-based measurements. Atmos. Environ. 34 (20), 3271–3280.

Rea, A.W., Lindberg, S.E., Keeler, G.J., 2000. Assessment of dry deposition and foliar leaching of mercury and selected trace elements based on washed foliar and surrogate surfaces. Environ. Sci. Technol. 34, 2418–2425.

Risch, M.R., Gay, D.A., Fowler, K.K., Keeler, G.J., Backus, S.M., Blanchard, P., Barres, J.A., Dvonch, J.T., 2011. Spatial patterns and temporal trends in mercury concentrations, precipitation depths, and mercury wet deposition in the North American Great Lakes Region, 2002–2008. Environ. Pollut. 161, 261–271.

Rodriguez, L., Rincon, J., Asencio, I., Rodriguez, C., 2007. Capability of selected crop plants for shoot mercury accumulation from polluted soils: phytoremediation perspectives. Int. J. Phytoremediation 9, 1–13.

Rolfhus, K.R., Sakamoto, H.E., Cleckner, L.B., et al., 2003. Distribution and fluxes of total and methylmercury in Lake Superior. Environ. Sci. Technol. 37, 865–872.

Rowland, A.P., Lawlor, A.J., Guyatt, H.J., Wadsworth, R., 2010. A Background wet deposition of mercury in Great Britain. Atmos. Environ. 12, 1747–1755.

Sakata, M., Marumoto, K., 2005. Wet and dry deposition fluxes of mercury in Japan. Atmos. Environ. 39, 3139–3146.

Schroeder, W.H., Munthe, J., 1998. Atmospheric mercury: an overview. Atmos. Environ. 32, 809–822.

Schwesig, D., Matzner, E., 2000. Pools and fluxes of mercury and methylmercury in two forested catchments in Germany. Sci. Total Environ. 260, 213–223.

Selvendiran, P., Driscoll, C.T., Montesdeoca, M.R., Bushey, J.T., 2008. Inputs, storage, and transport of total and methyl mercury in two temperate forest wetlands. J. Geophys. Res. 113, G00C01. http://dx.doi.org/10.1029/2008JG000739.

Sheehan, K.D., Feranadez, I.J., Kahl, J.S., Amirbahman, A., 2006. Litterfall mercury in two forested watersheds at Acadia National Park, Maine, USA. Water Air Soil Pollut. 170, 249–264.

Shuvaeva, O., Gustaytis, M., Anoshin, G., 2008. Mercury speciation in environmental solid samples using thermal release technique with atomic absorption detection. Anal. Chim. Acta 621, 148–154.

Silva-Filho, E.V., Machado, W., Oliveira, R.R., Sella, S.M., Lacerda, L.D., 2006. Mercury deposition through litterfall in an Atlantic Forest at Ilha Grande, Southeast Brazil. Chemosphere 65, 2477–2484.

St. Louis, V.L., Rudd, J.W.M., Kelly, C.A., Hall, B.D., Rolfhus, K.R., Scott, K.J., Lindberg, S.E., Dong, W., 2001. Importance of the forest canopy to fluxes of methyl mercury and total mercury to boreal ecosystems. Environ. Sci. Technol. 35, 3089–3098.

Stamenkovic, J., Gustin, M.S., 2009. Nonstomatal and stomatal uptake of atmospheric mercury. Environ. Sci. Technol. 43, 1367–1372.

Tang, J.W., Cao, M., Zhang, J.H., Li, M.H., 2010. Litterfall production, decomposition and nutrient use efficiency varies with tropical forest types in Xishuangbanna, SW China: a 10-year study. Plant Soil 335, 271–288.

Teixeira, D.C., Montezuma, R.C., Oliveira, R.R., Silva-Filho, E.V., 2012. Litterfall mercury deposition in Atlantic forest ecosystem from SE-Brazil. Environ. Pollut. 164, 11–15.

Travnikov, O., 2005. Contribution of the intercontinental atmospheric transport to mercury pollution in the Northern Hemisphere. Atmos. Environ. 39, 7541–7548.

US EPA, Method, 1631, 1999. Revision B: Mercury in Water by Oxidation, Purge and Trap, and Cold Vapor Atomic Fluorescence Spectrometry. United States Environmental Protection Agency, pp. 1–33.

Vanarsdale, A., Weiss, J., Keeler, G., Miller, E., Boulet, G., Brulotte, R., Poissant, L., 2005. Patterns of mercury deposition and concentration in northeastern North America. Ecotoxicology 14, 37–52.

Wang, Z.W., Zhang, X.S., Xiao, J.S., Ci, Z.J., Yu, P.Z., 2009. Mercury fluxes and pools in three subtropical forested catchments, southwest China. Environ. Pollut. 157, 801–808.

Watras, C.J., Bloom, N.S., 1994. The vertical distribution of mercury species in Wisconsin lakes: accumulation in plankton layers. In: Watras, C.J., Huckabee, J.W. (Eds.), Mercury Pollution: Integration and Synthesis. Lewis, pp. 137–152.

Wiener, J.G., Knights, B.C., Sandheinrich, M.B., Jeremiason, J.D., Brigham, M.E., et al., 2006. Mercury in soils, lakes and fish in Voyageurs National Park (Minnesota): importance of atmospheric deposition and ecosystem factors. Environ. Sci. Technol. 40, 6261–6268.

Witt, E.L., Kolka, R.K., Nater, E.A., Wickman, T.R., 2009. Influence of the forest canopy on total and methyl mercury deposition in the boreal forest. Water Air Soil Pollut. 199, 3–11.

Wu, Y., Wang, S.X., Streets, D.G., Hao, F.M., Chan, M., Jiang, J.K., 2006. Trends in anthropogenic mercury emissions in China from 1995 to 2003. Environ. Sci. Technol. 40, 5312–5318.

Yang, L.P., Liu, W.Y., Ma, W.Z., 2008. Woody debris stocks in different secondary and primary forests in the subtropical Ailao Mountains, southwest China. Ecol. Res. 23, 805–812. http://dx.doi.org/10.1007/s11284-007-0442-4.

Young, S.S., Carpenter, C., Wang, Z.J., 1992. A study of the structure and composition of an old growth and secondary broad-leaved forest in the Ailao Mountains of Yunnan, China. Mt. Res. Dev. 12, 269–284.

Zhang, X.M., Luo, K.L., Sun, X.Z., Tan, J.A., Lu, Y.L., 2006. Mercury in the topsoil and dust of Beijing city. Sci. Total Environ. 368, 713–722.