Mercury fluxes and pools in three subtropical forested catchments, southwest China

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The subtropical forested catchments are important for retention of atmospheric mercury deposition in southwest China.

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ABSTRACT

Forested catchments are an important part of the mercury (Hg) cycle and a link between the atmospheric and the aquatic environments. In this study, Hg input and output fluxes and its retention were investigated at subtropical forested catchments in southwest China. Significantly enhanced atmospheric Hg inputs were observed, and the contribution of litterfall Hg plays a more important role at these subtropical forested catchments. The ratios of Hg output fluxes from stream water to total input were 2.5% and 1.2% for LGS and TSP, which were markedly lower than those reported from Europe and North America. The current annual input Hg only accounted for 0.8 and 1.8 per mille to the Hg stored in the upper 90 cm of soil in LGS and TSP. These suggest that subtropical forested catchments are important for retention of atmospheric mercury deposition in southwest China.

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1. Introduction

Mercury is a well known global contaminant and a potent neurotoxin of significant ecological and public health concern. Neither atmospheric Hg levels nor ambient concentrations of Hg in water constitute a direct public health risk at the levels of exposure usually found in nature, and the risk to humans and wildlife occurs as Hg is transported to water and accumulates in the aquatic food chain (Driscoll et al., 2007). The most widespread concern regarding Hg is human consumption of fish, for instance fish consumption advisories issued in 44 states including more than 53,000 km² of lakes and 1,230,000 km of rivers in the USA in 2004. Although about 90% of the Hg in fish occurs as methylmercury (MeHg), the cycling of total Hg is of great concern because it is the source of MeHg from biological methylation of inorganic Hg in the environment (Boening, 2000).

Large amounts of studies, mostly in Europe and North America, emphasized that forested catchments act as filters between the atmosphere and the hydrosphere (Hultberg et al., 1995; Bishop and Lee, 1997). Long-range atmospheric transport and subsequent deposition of Hg, directly to water and indirectly via deposition to

terrestrial watersheds are the primary sources of Hg in aquatic ecosystem in remote areas (Fitzgerald et al., 1998; Peterson et al., 2007). Between 5% and 25% of atmospheric Hg deposited to forest watersheds and reached the associated lakes, which contributed approximately a half of Hg loading to these lakes (Kang et al., 2000; Galloway and Branfireun, 2004). The amount of Hg and MeHg transported from stream water varies as a function of the characteristics of the catchments, including soil type, hydrology, climate, land use and human activities (Porvari et al., 2003; Munthe and Hultberg, 2004).

Mercury cycles among the atmosphere, terrestrial and aquatic ecosystems undergo a series of complex chemical and physical processes. In recent decade, most studies have been carried out in the temperate and boreal forested catchments in Europe and North America, and yet significant gaps remain in our knowledge of Hg fate within subtropical forested catchments and its effects on aquatic ecosystem functions, and almost none of the data is available on the atmospheric Hg input, output and pool size in subtropical forested catchments especially in those areas with elevated Hg emission. According to the results by Hylander (2001), anthropogenic emissions of Hg still increase in China because of coal-use increasing and industrialization. A recent estimation of 696 ± 307 t Hg was emitted into the atmosphere from the anthropogenic source in 2003, and emission of Hg from southwest China was significant (Wu et al., 2006). Elevated levels of Hg in the

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atmosphere have been shown in several places in China (Wang et al., 2006a, 2007a; Feng et al., 2004). The characteristics of atmospheric mercury deposition and its subsequent cycling in forested catchments have not been previously reported in China. In this study wet-only precipitation (WP), throughfall (TF), litterfall (LF), stream water (SW) and soil profiles were regularly collected to determine Hg concentrations. The objective was to quantify the atmospheric Hg input and output fluxes and the retention in forested catchments, and consequently explore the roles of subtropical forested on the Hg biogeochemistry cycling.

2. Material and method

2.1. Sites description

Research forested catchments are TieShanPing (TSP), LuChongGuan (LCG) and LeiGongShan (LGS) in southwest China, which belong to typical subtropical monsoonal climate. TSP and LCG are located near big cities of Chongqing and Guiyang, respectively. Whereas LGS stands in a national forest park away 140 km of Guiyang, and is more regionally representative and a rural reference site. Mountains surrounding Guiyang and Chongqing cities cause poor air circulation and thus severe air pollution problems. These two areas belong to acid-rain control zone and the largest Hg-producing districts in China. The detailed characteristics about three forested catchments are showed in Table 1.

2.2. Field sampling

At each catchment, a modified automated wet-only precipitation collector (SYC-2, QingdaoLaoshan Ltd.) with Teflon-coated funnel connecting to 5-L polyethylene terephthalate (PET) bottle was set up at open space area close-by the catchment. Two 30 m × 30 m monitoring plots were established in each catchment, and four sets of throughfall samplers were fixed under the canopy of trees at the corners of the plots. The throughfall samplers were self-constructed based on the NILU (Norwegian Institute for Air Research) design, consisting of a 10.6-cm diameter PET funnel connected to a 3-L PET bottle. Four nylon nets (1 m² each) mounted 1 m above the forest floor at corners of the plot, were used for litterfall collection. Stream water was collected in the dam at the V-notch weir at the outlet of forested catchments. Soils were collected at intervals of 10 cm in typical soil profiles (0–90 cm) at each catchment.

The wet-only precipitation (WP), throughfall (TF) and stream water (SW) samples were collected weekly and litterfall (LF) monthly. The collecting bottles were pre-acidified with 7.5 ml of 6 N HCl (MOS, Suprapur) at the beginning of sampling to avoid the losses of Hg for volatilization or absorption on the container walls

(Guentzel et al., 1995; Krabbenhoft et al., 1995). All water samples were preserved by 2% (v/v) HCl to stabilize mercury, and freezing stored in the dark and analyzed for two months. Soil and litterfall samples were natural dried, ground and sieved to 2 mm for analysis.

2.3. Laboratory analysis

To measure Hg concentrations in water samples, liquid samples were oxidized with 0.2 N bromine monochloride (BrCl) for at least 12 h. Excess BrCl was removed by the addition of hydroxylamine hydrochloride (NH₂OH–HCl) prior to analysis. An oxidized sample aliquot was added to bubbler, and Hg²⁺ in sample was reduced to Hg⁰ by stannous chloride (SnCl₂). Then Hg⁰ was carried and measured in the multi-path analytical cell of RA-915+ analyzer (Lumex Scientific-production Company, Analytical Equipment, St. Petersburg, Russia), which is based on Zeeman atomic absorption spectrometry with high frequency modulation of light polarization (ZAASHFM). The detection limit of this method (defined as 3σ of the method blank value) was 0.40 ng L⁻¹ for 20 ml aliquot.

Hg concentrations in soil and LF samples were determined by RA-915+ analyzer attached with PYRO-91 thermal decomposition accessory from Lumex Ltd. The solid samples were directly decomposed in atomizer chamber at 800 °C with the aided catalytic action, and then Hg⁰ was detected by the RA-915+ analyzer. Detection limits were 0.5 µg kg⁻¹ for soil and 0.5 µg kg⁻¹ for litterfall samples. The detailed information was documented by Chen et al. (2001), USEPA (2004) and Huang et al. (2006).

2.4. Validation of analytical and sampling methods

Application RA-915+ analyzer with a multi-path analytical cell attached the RP-91 (cold vapor) and PYRO-91 (pyrolysis) accessories allow to analyze Hg in liquid and solid samples, and some authors have successfully used these methods for measuring Hg concentrations in water (Esbrí and Higuera, 2005; Higuera et al., 2006; Won et al., 2007) and solid (Chen et al., 2001; Oyarzun et al., 2007; Rodriguez et al., 2007; Shuvaeva et al., 2008) samples. In this study, the precision, obtained from eight replicate determinations of certificated reference material, was 3.0% for diluted (10 ng L⁻¹) Hg standard solution (GBW08617), 2.1% for soil (GBW07404) and 3.3% for peach leave (GBW08501). The recoveries were ranged between 92.2% and 105.1% for water samples, and from 96.0% to 102.3% for solid materials. In addition, calibration curves covering the appropriate concentration levels were run each day during analysis and control standard sample checked every tenth sample.

In this study, all containers for sampling and storage were made of polyethylene terephthalate (PET). Documents have validated that

Table 1
Characteristics of three research forested catchments.

Site	TSP (TieShanPing)	LGS (LeiGongShan)	LCG (LuChongGuan)
Latitude and longitude	104°41'E, 29°38'N	108°11'E, 26°22'N	106°43'E, 26°38'N
Elevation (a.s.l., m)	450–500	1630–1735	1320–1400
Catchment area (ha)	16.3	6.0	7.0
Annual mean temperature (°C) ^a	18.2	15.7	15.3
Annual precipitation (mm) ^b	1230	1615	1120
Mean daily RH (%) ^c	95	92	82
Soil classification	Haplic Acrisol/alisol	Haplic Acrisol/alisol	Haplic Acrisol/alisol
Bed rock	Sandstone	Shale	Sandstone
Vegetation information and ecosystem type	Massone Pine dominated, coniferous-broad leave mixed subtropical forest	<i>Pinus armandii</i> dominated, coniferous-broad leave mixed subtropical forest	Massone pine and <i>Cunninghamia lanceolata</i> dominated, broad leave-coniferous mixed subtropical forest

^{a,b,c} Data from nearby meteorological stations.

Table 2

Hg concentrations in wet-only precipitation and throughfall.

Site	Types	Rainfall (mm)	Concentration of Hg (ng L^{-1})				
			Mean	Median	Minimum	Maximum	V.W.C. ^a
LGS	WP	1341	19.5	14.9	3.5	71.0	12.9
	TF	938	54.2	40.2	8.1	285.9	36.7
TSP	WP	1403	55.3	39.0	9.1	232.7	32.3
	TF	1145	98.9	74.4	11.7	582.9	69.7
LCG ^b	TF	929	83.6	47.5	3.5	493.3	43.6

^a V.W.C. – annual volume-weighted mean concentration.^b No WP samples were collected at LCG catchment due to the absence of equipment.

the PET containers are fit for sampling and storage of natural water for Hg analysis (Copeland et al., 1996; Fatemian et al., 1999; Fadini and Jardim, 2000), and more and more studies performed based on it (Paraquetti et al., 2004; Silva et al., 2006; Wang et al., 2005). In our laboratory, no significant difference was found for the storage samples in Teflon and PET bottles (Paired *t*-test, $p = 0.028$) for three months. The procedures of field blanks for WP and TF were referred to the description of Guentzel et al. (1995) and Glass and Sorensen (1999). The reagents for preserving and analyzing of samples were of ultra pure quality grade.

3. Results and discussions

3.1. Wet-only precipitation and throughfall

Hg concentrations in WP and TF are presented in Table 2. Hg annual volume-weighted mean concentrations in TF were 69.7 ng L^{-1} , 36.7 ng L^{-1} and 43.6 ng L^{-1} for TSP, LGS and LCG respectively, which were about two times higher than those in WP during this study and approximately three times higher than those in TF reported from temperate and boreal forested catchments in

Europe and North America (Rea et al., 2000; St. Louis et al., 2001; Larssen et al., 2008). Hg concentrations in deposition decreased in the order of $TSP > LCG > LGS$ with significant difference among the sites (Kruskal–Wallis test, $P_{TF} = 0.0001$). This is because that LGS is located in a remote area relatively isolated from industrial and human activities, while other two sites are located in suburban areas and are more strongly interfered by anthropogenic activities.

Fig. 1 shows weekly and seasonal Hg deposition fluxes from WP and TF during the campaign. The mean weekly fluxes of WP were $0.43 \pm 0.40 \mu\text{g m}^{-2} \text{ week}^{-1}$ and $1.29 \pm 0.50 \mu\text{g m}^{-2} \text{ week}^{-1}$ at LGS and TSP, and $0.89 \pm 0.60 \mu\text{g m}^{-2} \text{ week}^{-1}$, $2.69 \pm 0.92 \mu\text{g m}^{-2} \text{ week}^{-1}$ and $0.98 \pm 0.68 \mu\text{g m}^{-2} \text{ week}^{-1}$ for TF at LGS, TSP and LCG, respectively. Compared with the values in WP, the elevated Hg fluxes from TF implied that the forest canopy is important for increasing Hg input to the forest floor, most likely due to atmospheric scavenging. Hg fluxes from TF were slightly higher in spring than other seasons at these catchments, which may be due to wash-off dry deposition of Hg that accumulated on the forest canopy in the dry winter season. The ratios of weekly Hg input fluxes via TF to those from WP were 3.0 for LGS and 3.2 for TSP, which were greater than the value of

Fig. 1. Weekly Hg deposition fluxes from wet-only precipitation and throughfall.

Fig. 2. The fluxes of litterfall, Hg concentrations and Hg fluxes from litterfall at TSP (A) and LGS (B), and percentage of dominant vegetation with different defoliation ranges (C) (C data from IMPACTS, 2004).

1.5 suggested by Munthe et al. (1995) and also higher than the overall mean ratio of 1.8 for temperate and boreal forest reviewed by Grigal (2002). This is in accordance with deposition of main compounds (e.g. sulphur and calcium) in these catchments, where also the fraction of dry deposition relative to wet is higher than typical values in Europe and North America (Larssen et al., 2006). This indicated that the dry deposition of atmospheric Hg plays more significant role for the mercury cycling in these subtropical forested catchments.

3.2. Litterfall

Hg average concentrations in LF were $135.1 \pm 31.7 \mu\text{g kg}^{-1}$ for LGS and $104.8 \pm 18.6 \mu\text{g kg}^{-1}$ for TSP, which were higher than those in conifer and hardwood at temperate and boreal forest (Lindberg,

1996; St. Louis et al., 2001; Larssen et al., 2008) but much lower than the levels (10^5 – $10^6 \mu\text{g kg}^{-1}$) in seriously pollutant areas (Siegel et al., 1984; Fischer et al., 1995). The monthly Hg input fluxes from LF at TSP ($18.4 \pm 18.1 \mu\text{g m}^{-2} \text{mon}^{-1}$) were much higher than that at LGS ($6.5 \pm 4.8 \mu\text{g m}^{-2} \text{mon}^{-1}$), even if lower Hg contents were found in LF at TSP. From Fig. 2A and B, the seasonal variation of Hg fluxes via LF was contrary, and high Hg fluxes occurred in spring and summer at TSP and yet in autumn and winter at LGS. These were attributed to the large difference of the fluxes from litterfall input between two sites, and seventy two percent of litterfall deposited to the floor during March to July at TSP. Furthermore, from Fig. 2C, the three years (2001–2003) mean defoliation percentage was 45% at TSP and 3% at LGS, and a recent study showed that the pressure of air pollution and defoliating insects play major roles for forest damage at TSP (Wang et al., 2007b). As

Fig. 3. Mercury concentrations in stream water from forested catchments.

Fig. 4. Hg distribution in soil profile at three catchments.

thus elevated Hg concentrations in litterfall at LGS may be the consequence of long growing seasons for foliage to uptake Hg from atmosphere. Several studies have indicated that Hg concentrations in foliage increase with the growing season (Fleck et al., 1999; Rea et al., 2001).

3.3. Stream water

At the three forested catchments, Hg concentrations in stream water ranged from 1.9 ng L^{-1} to 36.3 ng L^{-1} and the mean concentration was $4.3 \pm 2.5 \text{ ng L}^{-1}$ for LGS, $6.2 \pm 3.5 \text{ ng L}^{-1}$ for TSP and $8.9 \pm 6.7 \text{ ng L}^{-1}$ for LCG (Fig. 3), which were comparable to those from temperate and boreal forested catchments. Significant difference of Hg concentrations in stream water was found among these sites (Kruskal–Wallis test, $P = 0.0003$), which may be attributed to the characteristics of catchments and the disturbance of anthropogenic activities such as deforestation and disarrange soil. LCG locates in a botanical garden of the Guiyang suburban and cutting and trample disturbances occurred frequently, and yet LGS stands at a remote and natural forest area. Fostier et al. (2000) discovered that Hg concentrations in stream water from natural

forest areas (2.5 ng L^{-1}) were much lower than those (8.9 ng L^{-1}) from disturbed forest areas.

The annual output fluxes of Hg from stream water were $3.0 \mu\text{g m}^{-2} \text{ a}^{-1}$ for LGS, $3.5 \mu\text{g m}^{-2} \text{ a}^{-1}$ for TSP and $4.5 \mu\text{g m}^{-2} \text{ a}^{-1}$ for LCG, which were calculated using the weekly Hg concentration and the estimated discharge of stream water. In this study, the water fluxes of stream water were calculated using 70% of the actual throughfall in TSP and 50% in LCG, which were based on the results of four years (2001–2004) monitoring of IMPACTS (Larsen et al., 2006) and referred to the report of Fujieda et al. (1997) and Wei et al. (2005). In addition, no statistically significant correlations were discovered between Hg in stream water and Hg input from WP and TF during study period, implying that Hg output from stream water is regulated by processes other than current deposition input in these subtropical forested catchments.

3.4. Catchments pools

At these forested catchments, Hg contents in soils at LGS were much higher than other two sites (Fig. 4). Hg contents in soils decreased with the increase of soil depth at three catchments, and the highest surface contents were $259 \mu\text{g kg}^{-1}$, $239 \mu\text{g kg}^{-1}$ and $167 \mu\text{g kg}^{-1}$ for LGS, TSP and LCG, respectively. Furthermore, soil water was aperiodically sampled using lysimeter at TSP during study period, and Hg concentrations in soil water were much higher at 2 cm depth ($59.6 \pm 40.3 \text{ ng L}^{-1}$) than those at 20 cm depth ($10.1 \pm 7.9 \text{ ng L}^{-1}$), indicating that Hg from atmospheric deposition was main retention in upper layer soil in forested catchments. The storage of Hg in soil (0–90 cm depth), estimated through the soil bulk density of corresponding horizon (IMPACTS, 2004) and Hg contents in each soil profile, was 1523 g ha^{-1} for LGS, 1647 g ha^{-1} for TSP and 938 g ha^{-1} for LCG.

3.5. Catchments budgets

The annual budgets of Hg at these catchments are presented in Fig. 5. Total input of Hg, the sum flux of TF and LF, was $119.5 \mu\text{g m}^{-2} \text{ a}^{-1}$ for LGS and $291.2 \mu\text{g m}^{-2} \text{ a}^{-1}$ for TSP, which was much higher than those from temperate and boreal forest catchments (Schwesig and Matzner, 2000; Lee et al., 2000; Munthe et al., 1998; St. Louis

Fig. 5. Hg budgets for three forested catchments (units: $\mu\text{g m}^{-2} \text{ a}^{-1}$, SV represents soil volatilization and TI shows total input).

Table 3

The comparison of Hg input/output in this study with other reported.

Site	Location	Forest type	Period	Input		Output SW	Soil storage (g ha ⁻¹)	LF/input (%)	Input/ storage (%)	SW/input (%)	Ref.
				TF ^a	LF ^a						
LGS	108° 11'E	<i>Pinus armandii</i>	2005/3–2006/2	41.2	78.3	3	1092 ^b	65.5	0.11 ^b	2.5	This study
Southwest China	26° 22'N						1523 ^c		0.08 ^c		
TSP	104° 41'E	Masson pine	2005/3–2006/3	71.3	219.9	3.5	1205 ^b	75.5	0.24 ^b	1.2	
Southwest China	29° 38'N						1647 ^c		0.18 ^c		
LCG	106° 43'E	Massone pine	2005/1–2006/1	49.0		4.5	695 ^b	–	–	–	
Southwest China	26° 38'N						938 ^c				
Lehstenbach	11° 52'E	Norway spruce	1998/4–1999/4	38.4	15.7	7	891	29	0.06	13	Schwesig and Matzner, 2000
Germany	50° 08'N						(0–60 cm)				
Steinkreuz	10° 27'E	<i>Fagus sylvatica</i> L.	1998/4–1999/4	27.8	34	2.8	193	55	0.32	4.5	
Germany	49° 52'N						(0–60 cm)				
Svartberget	19° 46'E	Norway spruce	1993–1998	15	18	1.6	–	54.5	–	4.8	Lee et al., 2000
Sweden	64° 14'N										
Gårdsjön	12° 03'E	Norway spruce	1995–1997	23	23	1.8	280	50	0.16	3.9	Munthe et al., 1998
Sweden	58° 04'N						(0–60 cm)				
Marcell	93° 28'W	Black spruce	1995/4–11	13	12.3	2.81	48	48.6	0.53	11	Grigal et al., 2000
Minnesota	47° 32'N	Aspen					(0–50 cm)				
ELA, Ontario	93° 43'W	Tamarack and black spruce	1998–1999	8	12			60			St. Louis et al., 2001
	49° 40'N										
Lake Huron	84° 48'W	Maple and beech	1996/6–1997/6	10.5	11.4			52.1			Rea and Lindberg, 2001
	45° 34'N										
Hadlock Brook	44° 20'W	Maple and	1999–2004	10.2	10.1	2.46		49.8		12.1	Johnson and Haines, 2007; Sheehan and Fernandez, 2006; Nelson and Johnson, 2007 Larssen et al., 2008
Cadillac Brook	68° 13'N	paper birch		9.4	10	0.76		51.5		3.9	
Langtjern	9° 39'E	Scots pine	2004/2005	6.7	2.7	2.5	174	28.7	0.05	26.6	Grigal, 2002
Sutheast Norway	60° 22'N						(0–B)				
Review	Temperate and boreal zones		1990–2000	17	21	1.7		55.5		4.5	

^a The unit of input and output fluxes was $\mu\text{g m}^{-2} \text{a}^{-1}$.^{b,c} Represent the soil of 0–60 cm and 0–90 cm, respectively.

et al., 2001; Grigal, 2002; Johnson and Haines, 2007). This implied that enhanced atmospheric Hg deposition exists in forest catchments at southwest China, and it may be attributed to the elevated Hg emission from coal combustion and mining. In this study, Hg volatilization fluxes from forest floor were estimated through the Hg release velocity measured by Wang et al. (2006b) using dynamic flux chamber from forest soils surface at remote and suburban sites in Chongqing of southwest China. And we also referred the emission fluxes of Hg from soil in Guiyang (Feng et al., 2003) and from forest floor of the Adirondacks in New York (Choi and Holsen, 2009). The estimated fluxes of Hg volatilization were $67.4 \mu\text{g m}^{-2} \text{a}^{-1}$ for LGS and $73.6 \mu\text{g m}^{-2} \text{a}^{-1}$ for TSP and LCG. Therefore the annual net retention Hg from the atmosphere was $49.1 \mu\text{g m}^{-2} \text{a}^{-1}$ for LGS and $214.1 \mu\text{g m}^{-2} \text{a}^{-1}$ for TSP, which only accounted for about 0.03% and 0.13% of the soil (0–90 cm) storage Hg in LGS and TSP, respectively.

3.6. The role of forested catchments in mercury cycling in China

A comparison of Hg input/output in subtropical forested catchments with those in temperate and boreal forested catchments is shown in Table 3. The input fluxes of Hg via LF accounted for 65.5% and 75.5% of total input at LGS and TSP, respectively, which were significantly higher than those from temperate and boreal forested catchments, suggesting that Hg input via litterfall may play a more important role to the mercury cycling in subtropical forested catchments. The output fluxes of Hg from stream water in this study were comparable with those in temperate and boreal forested catchments, but inputs considerably higher and hence the ratios of Hg output to total input in these catchments were clearly lower, due to strong retention of atmospheric mercury deposition in these catchments. The current annual input Hg was 0.8 and 1.8 per mille to the Hg stored in the upper 90 cm of the soil in LGS and TSP. Hence, currently most of the atmospherically deposited Hg is

retained in the soil rather than exported to surface water. The long term implication of this, however, is unknown, and in the long term leaching from stored Hg in forested catchments may occur to a larger extent than today even if the reductions of Hg emissions from anthropogenic sources to the atmosphere.

4. Conclusions

In this study weekly wet-only precipitation (WP), throughfall (TF), stream water (SW), monthly litterfall (LF) and soil profiles were collected to investigate the Hg fluxes and pools at three subtropical forested catchments (Leigongshan (LGS), Tieshanping (TSP) and Luchongguan (LCG)) in southwest China. Hg annual volume-weighted mean concentrations in TF were 36.7 ng L^{-1} , 69.7 ng L^{-1} and 43.6 ng L^{-1} for LGS, TSP and LCG, respectively, which were two times more than those in WP during this study and approximately three times higher than those in TF reported from temperate and boreal forested catchments. The annual total input fluxes of Hg were $119.5 \mu\text{g m}^{-2} \text{a}^{-1}$ for LGS and $291.2 \mu\text{g m}^{-2} \text{a}^{-1}$ for TSP, which were several-fold of those in temperate and boreal forested catchments, suggesting significantly enhanced atmospheric Hg deposition at subtropical forested catchments in southwest China. Output fluxes of Hg from stream water were $3.0 \mu\text{g m}^{-2} \text{a}^{-1}$, $3.5 \mu\text{g m}^{-2} \text{a}^{-1}$ and $4.5 \mu\text{g m}^{-2} \text{a}^{-1}$ for LGS, TSP and LCG, respectively, which were comparable to those reported from Europe and North America. The current annual input Hg only accounted for 0.8 and 1.8 per mille to the Hg stored in the upper 90 cm of soil in LGS and TSP. Therefore, for a long term, the large Hg pools in forest soil may be significant sources for associated lakes even if the reduction of Hg emission to the atmosphere from the anthropogenic sources in southwest China.

The comparison of Hg input/output in this study with those reported from temperate and boreal forested catchments showed that the litterfall input of Hg from the atmosphere was significantly

higher at TSP and LGS, suggesting that Hg input via litterfall may play a more important role to the mercury cycling at subtropical forested catchments in China. The ratios of Hg output from stream water to total input at these catchments were obviously lower than those from temperate and boreal areas although there is a significantly high Hg input in southwest China. These suggest that subtropical forested catchments are important for retention of atmospheric mercury deposition in southwest China.

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