Supporting Information for

Subtropical forests act as mercury sinks but net sources of gaseous elemental mercury in South China

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Table S1~S2

Text S1. Sites description

The Qianyanzhou (QYZ) and Huitong (HT) experimental stations are managed by the Chinese Academy of Sciences (CAS) and Central South University of Forestry and Technology (CSUFT), respectively. The QYZ station (115º04'E, 26º45'N) is located in Taihe county, Jiangxi province (Figure 1, Table S1), surrounded by farmland, with no obviously anthropogenic mercury sources such as coal-fired power plants and metal smelters in 25 km around. The HT station (109º45'E, 26º50'N) is located in Huitong county, Hunan province, about 100 km away from the Wanshan Mercury Mine (WS), which used to be the largest mercury mine in China. The two study sites have the similar climate condition. The dominant soil and vegetation types (Table S1) are widely distributed in subtropical monsoon climate zone in south China. The subtropical evergreen coniferous forests have fairly thick canopy, even in winter.

Text S2. The observations of air-canopy and air-foliage Hg^0 exchange

The air-canopy Hg^0 exchange flux was measured by the micrometeorological method (MM) for a full year in 2014 (same period for other measurements) at both sites QYZ and HT, using flux tower and Tekran 2537 (Tekran Instruments Inc. detection limit < 0.1 ng m^{-3}).[1] Air intakes were placed at two different heights (25 and 35 m of the 41 m-high flux tower at QYZ site; 22.5 and 30.5 m on the 33m-high flux tower at HT site). The difference of Hg^0 concentrations between the two air intakes divided by the height difference was assumed to be the vertical gradient of atmospheric Hg^0 concentration. The turbulent transfer coefficient was calculated based on the measurement of the wind speed and temperature profile. Then, the Hg^0 fluxes over forest canopy were calculated combined the Hg^0 concentration gradient and turbulent transfer coefficient, and had annual average values in 2014 of 58.5 and 2.65 μg m^{-2} yr^{-1} at site QYZ and HT, respectively.[1] The monthly variations of Hg^0 concentrations and fluxes was showed as Figure S1.

The foliage-air Hg^0 exchange flux was observed using dynamic flux chamber

(DFC) over four seasons at site QYZ in 2014.[2] The DFC was made by 3 mm thick polycarbonate plates with external dimensions of 200mm× 200mm× 400mm (16 L). One side of the chamber (200mm× 200 mm) was open to enclose the branch, and this opening was sealed using flexible material of poly (vinyl formal) film (Figure 1). Air inlets (12 holes in total and 4 mm in diameter) were evenly distributed at the end of the chamber. To increase the turnover rate and turbulence in the chamber, a plastic DC brushless computer fan was mounted in the chamber using nylon screws, supplementing the 1 L min^{-1} pump flow of the Hg analyzer and an external pump with 10 L min^{-1} flow. The DFC was installed at a height of about 12 m supported by a tower located in the central of the forest experimental station. The branch measured was taken from the Masson Pine around the tower. The field observations were carried out during four seasons, each lasting 2–4 weeks and using 5 branches from different tree.

The Hg^0 concentrations in the chamber and ambient air were measured alternatively using an automatic Hg analyzer (Tekran 2537, Tekran Instruments Inc.). A solenoid valve at the front of Tekran 2537X was switched every 10 min synchronously to allow air from inside or outside the chamber successively to enter the analyzer. Foliage-air exchange flux from the foliage enclosed in the DFC was calculated using the Hg^0 concentrations in the DFC and in the ambient air, the flushing flow rate through the chamber, and the total enclosed leaf surface area. Combined with the leaf area and leaf area index (LAI), the Hg^0 emission flux from the forest was obtained. The current-year needles and previous-year needles were observed separately except during winter, when most of the previous-year needles have fallen. The results showed a bidirectional Hg^0 exchange between foliage and atmosphere, manifesting as emission and adsorption/deposition peaked midday and in the morning. The annual average net emission of Hg^0 in 2014 was measured as 10.9 μg m^2 yr^{-1}, including 1.7 μg m^2 yr^{-1} from current-year needles and 9.2 μg m^2 yr^{-1} from previous-year needles (Figure S2).[2]

Text S3. The observations of Hg fluxes in precipitation and throughfall

Samples of precipitation and throughfall were collected weekly from December 2013 to November 2014 at sites QYZ and HT.[3] In each 10 m × 10 m forest plots (three plots at each site), where the soil water and literfall were collected, the self-made collectors were equipped in triplicate for throughfall collection. Synchronously, the precipitation was sampled in quintuplicate also using self-made collectors at the adjacent open areas. The collectors consisted of an 8.6-cm-diameter funnel with nylon gauze to exclude canopy litter and an opaque 3L bottle to store water. The sample bottles were acidified with 4 mL of 6 M ultrapure HCl prior to collection, to inhibit transformations and adsorption of Hg. Water samples were added 6 M HCl to pH < 2 and stored at 4°C immediately after collection. Every four weeks, the samples were pooled into monthly bulk samples prior to analysis, and the total volumes of precipitation and throughfall were recorded to calculate the water fluxes. All the water samples were measured for the total Hg concentrations by cold atomic fluorescence method (Tekran 2600. Tekran Inc., Canada), following the modified US-EPA Method 1631 guideline (revision E, August 2002). The acid-preserved water samples were transported to the laboratory every month, then were oxidized with BrCl addition to the original samples bottles (re-solubilize Hg condensed on the bottle walls during storage) at least 24 hours prior to analysis. A 100 mL-subsample were filtered (0.45 μm, glass fiber) after BrCl addition to prevent line clogging of Tekran 2600. The oxidization, filtration, and the subsequent reduction and detection were performed in a clean room to prevent contamination.[4]

The annual concentrations (water flux-weighted of monthly concentrations) in precipitation and throughfall had values of 12.5 and 22.2 $ng \cdot L^{-1}$ at QYZ site, and 8.62 and 22.0 $ng \cdot L^{-1}$ at HT site. The annual Hg fluxes (in $\mu g \cdot m^{-2} \cdot yr^{-1}$) were calculated by multiplying the annual concentrations with the water fluxes. Consequently, the wet and throughfall deposition were 18.4 and 30.6 $\mu g \cdot m^{-2} \cdot yr^{-1}$ at site QYZ. At HT, the annual fluxes were 15.6 and 37.3 $\mu g \cdot m^{-2} \cdot yr^{-1}$ in precipitation and throughfall, respectively.[3] The seasonal Hg fluxes via precipitation and throughfall were showed in Table S2.

Text S4. Foliar biomass and Hg pool

The biomass of current-year and previous-year needles of Masson pine in summer was measured at site QYZ.[5] In order to get the biomass in other seasons, we collected two hundred pairs of needles (two needles is one bundle for Masson pine) randomly in each branch from the six Masson pines around the observation site every month and measured dry weight. The biomass in other seasons (M_i; kg·m^{-2}) was calculated according to the biomass in summer and the dry weight ratio between summer and other seasons by the following equation:

$$M_i = M_s \times \frac{m_i}{m_s} \quad \text{(S1)}$$

Where M_s is the biomass of current-year needles in summer (kg m^{-2}); m_i and m_s are the dry weights of needle per pair in spring/autumn and summer, respectively. Here the current-year needles were assumed not to fall during the growing season. The foliar biomass at HT was about 1080 g m^{-2},[6-8] and the foliar primary production was about 162 g m^{-2} yr^{-1},[6] which is approach to the total mass of litterfall measured (205 g m^{-2} yr^{-1}).

The total foliar Hg pool is consisted of the Hg in both current-year and previous-year needles, which is equal to the Hg content of needles multiplied by the foliar biomass. For the current-year needles at QYZ, the monthly foliar Hg pool was calculated according to the monthly Hg contents and biomass of needles showed in Figure 2b. For the previous-year needles at QYZ, the Hg content was relatively stable in different months according to Figure 2b, with a mean value of 51.3 ng g^{-1}. For Masson pine, the current-year needles are in dormant after December, and become previous-year needles in the next year, then begin to fall till bald in December. Thus, the biomass of previous-year needles in Month i (B_{pi}) was calculated by the biomass of current-year needles in December ($B_{c\text{-}12}$) and the cumulative biomass of litterfall ($\sum B_{Li}$):

$$B_{pi} = B_{c\text{-}12} - 0.65 \times \sum B_{Li} \quad \text{(S2)}$$

where the 0.65 is the ratio of the current-year needle biomass in December (402 g m^{-2}) to the annual litterfall biomass (619 g m^{-2}), which is less than the unit due to the fall of

current-year needles, branch and bark. The total foliar Hg pool was relatively stable in different months, with a mean value of 18.3 μg m^{-2}.

At HT site, the foliage for Chinese fir may live for many years, with the foliar biomass of 162 g m^{-2} (primary production) for current-year needles, and 918 g m^{-2} for previous-year (total biomass minus biomass of current-year needles). The mean Hg content in current-year needles was 87.0 ng g^{-1} according to Figure 2b. For previous-year needles, the Hg content was substituted by that of litterfall, with an average value of 178 ng g^{-1}, due to no direct-measured data. Thus, the foliar Hg pool was approximately 177 μg m^{-2}. The foliar Hg pool was assumed to be stable for the mature forest with stable biomass. The difference between the foliar primary production (162 g m^{-2} yr^{-1}) and the annual mass of litterfall (205 g m^{-2} yr^{-1}) might be caused by the fall of current-year needles, branch and bark.

Text S5. QA/QC and uncertainty analysis

The Quality assurance/Quality control (QA/QC) of the observations of Hg^0 fluxes above forest canopy included the maintenance and calibration of the instrument (e.g. regularly renewal of filter membrane on the air intake and the soda-lime tank; automatic calibration and manual calibration), the blank experiments (e.g. measuring the detection limit of the concentration gradient for the monitoring systems before the installation, detection limits were 0.021 ng·m^{-3} and 0.034 ng·m^{-3} at QYZ and HT sites, respectively), and the cleaning of the systems (soaked in 10% HCl for 24 h and washed by ultrapure water to clean, then sealed by polyvinyl chloride film). The observation time with valid data (elimination of the values outside the range of the monthly mean ± 3 standard deviations, and the problematic data during the high atmospheric stability, instrument failure and instability operation) have 86% and 91% coverage in the year.

Similar QA/QC was conducted during the foliar Hg^0 flux measurement. The mean difference in Hg^0 concentrations between inside and outside of the chamber was not significant (0.08 ± 0.12 ng·m^{-3}) (one-way ANOVA; $n > 40$) during the blank experiment. And the detection limit of flux was 0.13 ng·h^{-1}, defined as the sum of mean

value and standard deviation. The contribution of the fluxes data over 0.13 $ng \cdot h^{-1}$ (detection limit) was 91.8%. In addition, the uncertainty might come from the possible influences from DFC, for instance the positive effects from greenhouse effect and negative effects from UV blocking) on foliar Hg^0 emission.

For quality control on the analysis of Hg in water/plant/soil samples, a Hg^{2+} standard solution was prepared from a reference standard (1000 $\mu g \cdot mL^{-1}$ in 3% HNO_3, Ricca Chemical Company), plant standard reference materials (GBW07410) and soil standard reference materials (GBW07405) were purchased from the National Research Centre for Reference Materials of China. When measured values deviated by more than 5% from the standard values, the analyzers were recalibrated. The recovery percentages from spiked samples ranged from 90% to 110% for Hg in water samples and ranged from 95% to 105% for Hg in solid samples. The collectors for water samples, including self-made collectors for precipitation and throughfall and lysimeters for soil water, were soaked in 10% HCl for 24 h and washed by ultrapure water to clean before setup. Considered the possibility loss of Hg due to long times storage, we measured the bulk water samples every month after combined, so the bulk samples stored no longer than one week.

The uncertainties of annual forest-atmosphere Hg^0 flux (i.e. air-canopy flux) were 3% and 5% at sites QYZ and HT, respectively, calculated by error propagation of hourly averages for each day of every month and propagated to annual flux. The uncertainty associated with the annual air-foliage Hg^0 flux was 10% at QYZ site, using the same method applied in uncertainties estimate of air-canopy exchange flux. The uncertainty of Hg fluxes in precipitation, throughfall, and leachate was derived from the errors of Hg analysis and the spatial variation. The error for THg concentrations is 8% (according to the precision of EPA method 1631 for fresh water). The spatial variation was defined as ± relative standard deviation (RSD, %) of monthly samples in three plots at each site. The annual concentrations are the volume-weighted of monthly concentrations, and multiplying with water flux to obtain annual flux. Based on the uncertainties propagation, the uncertainties of Hg fluxes in precipitation, throughfall,

and leachate were 9%, 11%, and 11%, respectively at site QYZ, and 12%, 15% and 8%, respectively at site HT. Similarly, the uncertainty of monthly Hg concentration in litterfall was defined as ±1 RSD of nine samples in each month (three collectors in each plots, and 3 plots at both site). The annual Hg flux from litterfall was calculated by summing monthly fluxes, which were obtained by multiplying the Hg content by the mass flux of each litter sample. According to the uncertainty propagation, the uncertainties of annual Hg flux in litterfall were 9% and 6% at site QYZ and HT, respectively. Overall, the uncertainties of dry Hg^{II} deposition and Hg sink for entire forests were 20% and 21%, respectively, at QYZ site, and 20% and 19%, respectively, at HT site, according to errors propagation.

Text S6. Hg budgets for entire forest and soil

The total Hg sink for entire forest (S_{forest}) can be calculated according to the following equation (i.e. Eq 5):

$$S_{forest} = F_{WD} + F_{DD} - F_{CE} - F_{LE} \quad \text{(S3)}$$

where F_{DD} was estimated based on the equation (S4), i.e. Eq 4

$$F_{DD} = F_{TF} - F_{WD} + F_{LF} + F_{FE} \quad \text{(S4)}$$

Then the S_{forest} can estimate by:

$$S_{forest} = F_{TF} + F_{LF} - (F_{CE} - F_{FE}) - F_{LE} \quad \text{(S5)}$$

The total Hg budgets for soil (S_{soil}) was showed as Figure S3b, having Hg input via throughfall (F_{TF}) and litterfall (F_{LF}), and output via soil emission (F_{SE}) and leaching (F_{LE}), and can calculate according to:

$$S_{soil} = F_{TF} + F_{LF} - F_{SE} - F_{LE} \quad \text{(S6)}$$

The Hg^0 emission form entire forest (i.e. Hg^0 flux of air-canopy, F_{CE}) is the sum of Hg^0 emission for soil (F_{SE}) and vegetation (F_{FE}), showing as:

$$F_{CE} = F_{SE} + F_{FE} \quad \text{(S7)}$$

Consequently, the S_{forest} is equal to S_{soil} based on Eq S5, S6, and S7. In addition to mathematical derivation, it is because the Hg keep balance for vegetation (Figure 3a) in theoretical understanding.

Tables

Table S1. Description of QYZ and HT experimental station.

Station sites	**QYZ**	**HT**
Altitude (m)	30~60	280~390
Climate type	Humid subtropical monsoon climate	
MAT (°C)	18.6	15.8
MAP (mm)	1361	1200
Dominated tree species (relative abundance)	*Pinus massoniana* (86.5%)	*Cunninghamia lanceolata* (92.4%)
Other predominant vegetative species	*Pinus elliottii; Quercus fabei; Vitex negundo; Rhododendron plonch; Ischaemum indicum*	*Marsa japonica ; Ilex pirpurea; Cyclosorus parasticus; Woodwardia prolifera*
Forest age (yr)	31	27
Canopy height (m)	16	14
Leaf area index (LAI) in summer	4.31	7.00
Canopy density	0.7	0.8
Radiation transfer under canopy	3.0%	2.7%
Dominant soil type (Chinese soil name)	Udic Ferrisols (Red Earth)	Haplic Acrisol (Yellow Earth)
Annual air Hg^0 conc. (ng m^{-3}) [a]	3.64 ± 1.82	5.93 ± 3.16
Soil pH[b]	4.52	3.85
Surface soil organic matter (g kg^{-1})	10~15	28.3
DOC of soil water in 30 cm (mg L^{-1})	4.28 ± 0.73	5.74 ± 1.21
Hg content in soil organic layer (ng g^{-1}) [b]	76.2 ± 6.0	153 ± 28
Hg content in 0~5 cm soil (ng g^{-1}) [b]	42.6 ± 2.3	167 ± 32
Oi Hg pool (μg m^{-2})	39.4	27.5
Oa&Oe Hg pool (μg m^{-2})	125	146
A Hg pool (μg m^{-2})	3731	1187
B Hg pool (μg m^{-2})	15104	5401

[a] Mean value of the measurements at the height of 25 m and 35 m at QYZ site, 22.5 and 30.5 m at HT site;

[b] Analyzed based on 18 samples using a direct Hg analyzer (DMA80, Milestone Inc., Italy).

Table S2. The atmospheric Hg^0 concentrations (Hg^0 conc., ng m^{-3}**)**, Hg concentrations in litterfall (LF Hg conc., ng g^{-1}), and Hg fluxes (μg m^{-2} yr^{-1}) via precipitation (F_{WD}), throughfall (F_{TF}), litterfall (F_{LF}), soil emission (F_{SE}), and leaching (F_{LE}), as well as Hg sink (μg m^{-2} yr^{-1}) and pool (μg m^{-2}) for soil at different sites.

Country[a]	**Site**	**Period**	**Lat Lon**	**Vegetation**	**Hg^0 conc.**	**LF Hg conc.**	**F_{WD}**	**F_{TF}**	**F_{LF}**	**F_{SE}**	**F_{LE}**	**Sink**	**Pool**[b]	**Reference**
China [S]	QYZ annual	2014/1 2014/12	26.75 115.07	Masson pine	3.64	42.9	18.4	30.6	26.2	47.6	3.89	5.31	6461[15]	This study
	QYZ spring	2014/3 2014/5			3.47		21.6	24.0	17.1	43.2	2.24	-4.30		
	QYZ summer	2014/6 2014/8			3.30		30.0	51.6	30.0	48.6	4.04	28.9		
	QYZ fall	2014/9 2014/11			3.75		9.40	17.3	33.5	68.9	7.56	-25.6		
	QYZ winter	2014/12 2014/2			4.05		12.6	29.7	24.1	29.2	1.68	22.9		
	HT annual	2014/1 2014/12	26.83 109.75	Chinese fir	5.93	177.9	15.6	37.3	36.4	2.65	3.80	67.3	19000[15]	This study
	HT spring	2014/3 2014/5			5.31		16.3	33.7	34.4	7.30	2.24	58.6		
	HT summer	2014/6 2014/8			4.99		25.5	53.6	43.0	38.5	4.04	54.0		
	HT fall	2014/9 2014/11			6.30		7.80	38.0	36.1	-3.50	7.56	70.1		
	HT winter	2014/12 2014/2			5.87		12.1	22.3	32.0	-31.7	1.68	84.3		
	Simian	2012/3 2013/2	28.62 106.40	Evergreen broad leaf	3.80	106.7	16.0	32.2	42.9	18.6	7.23	49.2	25341[98]	9
	Jinyun	2012/3 2013/2	29.78 106.36	Evergreen broad leaf	4.20	104.5	15.9	21.8	43.5	12.2	6.53	46.6	-	10
	Gongga	2005/5 2005/4	29.65 102.12	Fir/Cuculidae	3.98	35.7	26.1	57.0	35.5	14.0	8.60	69.9	-	11
	Tieshanping	2009/10 2010/10	29.63 106.68	Masson pine	5.30	115	-	67.5	22.3	2.70	12.6	74.6	28159[30]	12, 13
	LeiGongShan	2005/3 2006/2	26.37 108.18	Pinus armandii coniferous-broad mixed	4.60	135	16.8	41.2	78.3	67.4	3.00	49.1	58500[30]	14
	TieShanPing	2005/3 2006/3	29.63 104.68	Masson Pine coniferous-broad mixed	6.74	105	29.0	71.3	220	73.6	3.50	214	80300[30]	

	LuChongGuan	2005/1 2006/1	26.63 106.72	Massone pine broad -coniferous mixed	-	-	-	49.0	-	73.6	-	-	38600[30]	
	Leigong	2008/5 2009/5	26.39 108.2	Evergreen broad leaf	2.80	91.0	6.10	10.5	39.5	-	4.50	-	-	15
	Ailao	2011/6 2014/5	24.53 101.02	Evergreen broad-leaved primary forest	2.09	54.0	4.90	22.9	75.0	-	-	-	191300[80]	16
	Damei	2012/8 2013/7	29.63 121.57	-	3.31	46.6	7.00	-	26.0	-	-	-	-	17
China[T]	Changbai	2009/1 2016/1	42.4 128.14	Korean pine/China ash/ Acer mono mixed forest	1.60	43.0			20.9					18
Korea[T]	Yangsuri	2008/8 2010/2	37.53 127.3		2.15	50.2	4.30	6.40	4.60	6.80	0.60	3.60		19
Sweden[T]	Gårdsjön	1993 1998	58.07 12.05	Norway spruce			10.0	23.0	23.0		1.80			20
	Svartberget	1995 1997	64.23 19.77	Norway spruce			7.00	15.0	18.0		1.60		28000[60]	
Germany[T]	Lehstenbach	1998/4 1999/4	50.13 11.87	Norway spruce			36.0	38.4	15.7		7.00		89100[60]	21
	Steinkreuz	1998/4 1999/4	49.87 10.45	Fahus sylvatica L			37.5	27.8	34.0		2.80		19300[60]	
Norway[T]	Langtjern	2004 2005	60.37 9.65	Scots pine				6.70	2.70		2.50		17400[60]	22
America[T]	Huntington Wildlife Forest	2009 2010	43.97 74.22	Hardwood mature sugar maple	1.4	6.7	6.9	8.9	7				40600[100]	23
				Conifer white pine			18.2	8.7	-1				33800[100]	
Review[T,B]	Review	1990 2000					9.60	17.0	21.0	11.0	1.70	25.3		24
North America[T]	Review				1.32-1.66	32-57	2.5-21.5	1.6-12	7.2-15					17
Europe[T]	Review				1.4-1.93	28-68	7-36	15-39	4-32.5					

[a] The superscript presents the climate type, S, T, and B means subtropical, temperate, and boreal, respectively;

[b] The superscript presents the total soil depth, cm.

Figures

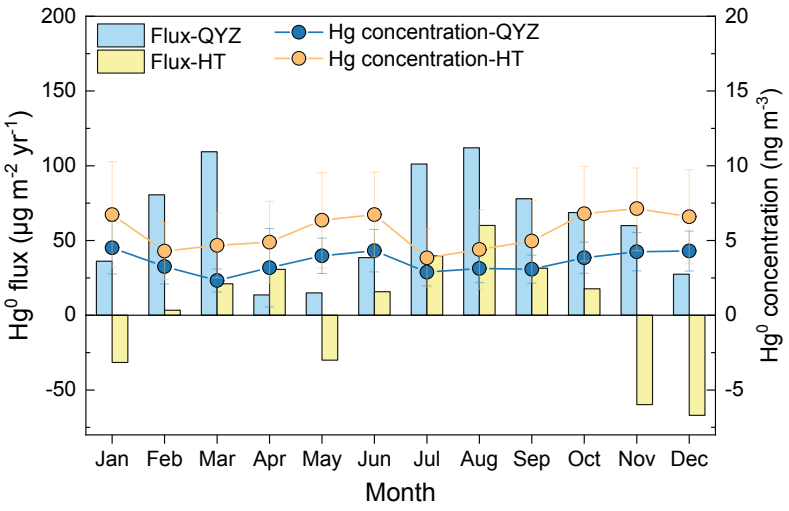

Figure S1. Monthly variations in atmospheric Hg^0 concentrations and Hg^0 exchange fluxes (positive means net emission, and negative means sorption) over the forest canopy at sites QYZ and HT in 2014.

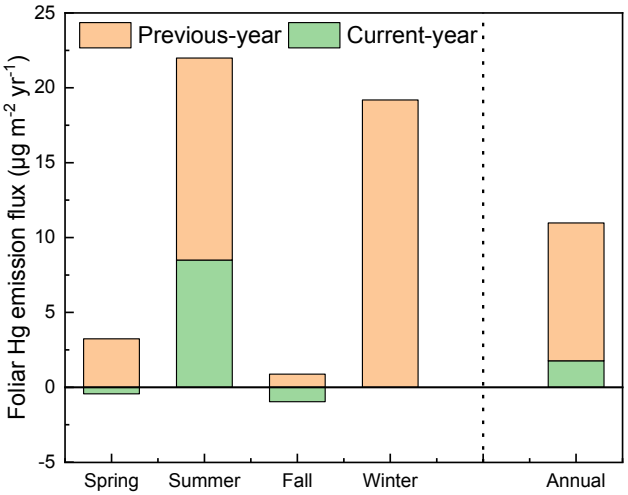

Figure S2. Seasonal and annual foliar Hg^0 emission fluxes (positive means net emission, and negative means sorption) at site QYZ in 2014.

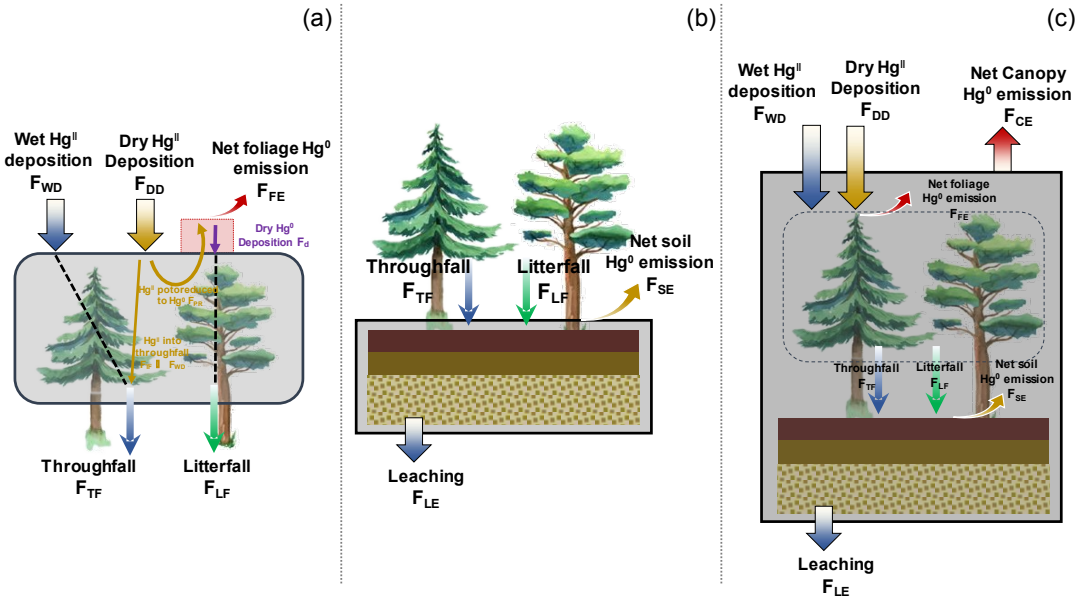

Figure S3. Concept map of Hg budgets for forest canopy (a), soil (b), and entire forest (c)

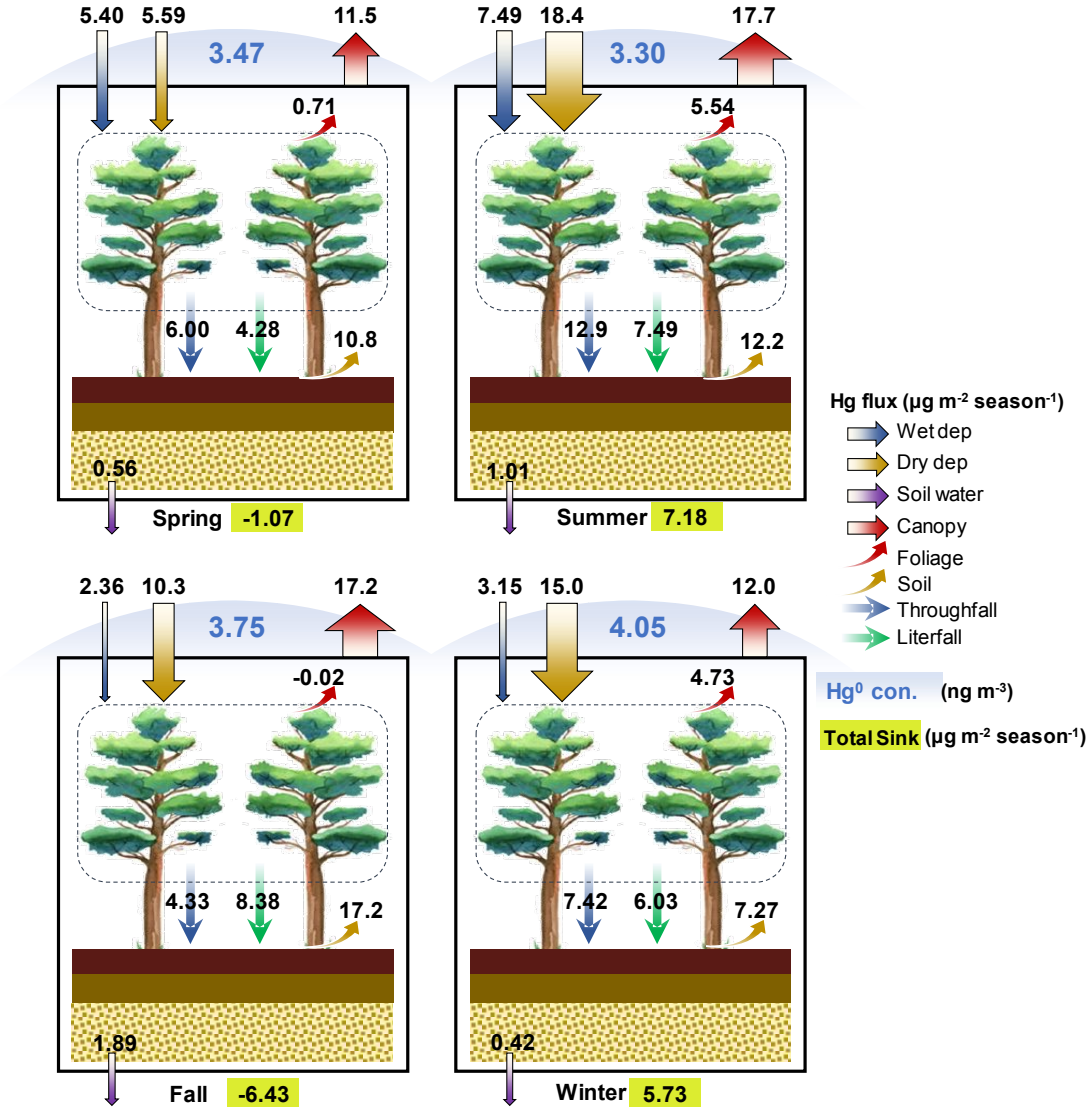

Figure S4. Seasonal Hg mass balance at site QYZ in 2014. Fluxes (μg m^{-2} $season^{-1}$) are presented by arrows.

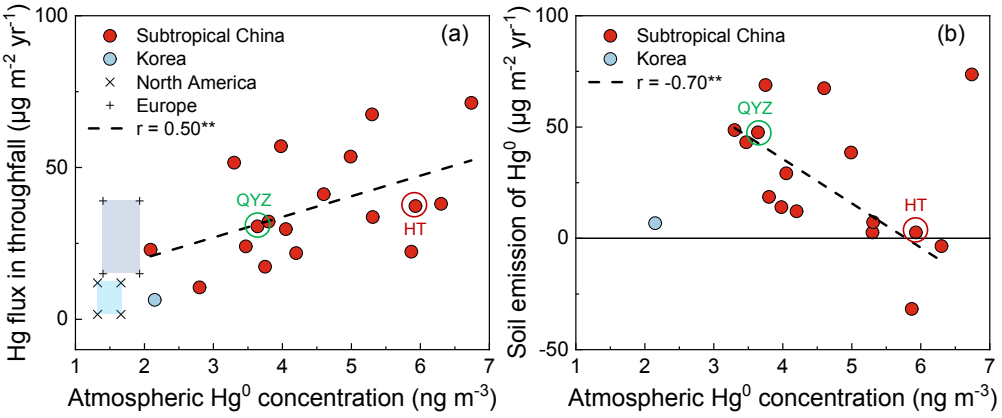

Figure S5. Correlations between atmospheric Hg^0 concentrations and Hg fluxes in throughfall (a), and between atmospheric Hg^0 concentrations and soil emission fluxes (b). Detail data showed in Table S2.

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