



Mercury distribution in the foliage and soil profiles of the Tibetan forest: Processes and implications for regional cycling



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ABSTRACT

Remote forests are considered a pool of Mercury (Hg) in the global Hg cycle. However, notably few studies have investigated the fate of Hg in the Tibetan forest. In this study, fifty-two foliage samples and seven litter/soil profiles were collected throughout the Tibetan forest. The concentrations of total Hg (THg) in foliage were positively correlated with longitude and negatively correlated with altitude, indicating that the emission of Hg is expected to decrease with increasing distance from emission sources to the Tibetan forest. The deposition flux of THg in the Tibetan forest (with an air-to-forest ground flux of 9.2 $\mu\text{g}/\text{m}^2/\text{year}$) is ~ 2 times the flux in clearings, which is suggestive of enhanced Hg deposition by the forest. The depositional Hg is eventually stored in the forest soil, and the soil acts as a net 'sink' for Hg.

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1. Introduction

Mercury (Hg) and its compounds (especially methylmercury) are toxic to human beings (Tchounwou et al., 2003). Because of its volatility and persistence, mercury vapor can remain in the atmosphere for months or even years (Schroeder and Munthe, 1998), and thus, Hg can reach remote areas, where it has never been produced or used (Seigneur et al., 2001). Therefore, Hg is considered to be a global pollutant. This view is supported by global biogeochemical Hg models (Corbitt et al., 2011) and sedimentary archives of historic Hg fluxes (Biester et al., 2007; Engstrom and Swain, 1997).

Forests are one of the important ecosystems in remote areas and are considered a large pool of atmospheric Hg deposition (Graydon et al., 2006, 2012; Obrist, 2007; Selin, 2009; Stankwitz et al., 2012). The atmospheric Hg can be uptaken by the stomata (gaseous elemental Hg) and/or absorbed by leaf surfaces (particulate Hg, reactive gaseous Hg, and Hg in wet deposition) (Bargagli et al., 1986; Graydon et al., 2009; Lindberg et al., 1992; Millhollen et al., 2006). Because of the relatively longer half-lives time of Hg in foliage (110–180 days, Graydon et al., 2012), leaves are considered a bio-monitor for studies of the spatial distribution of Hg (Amado Filho et al., 2002; Figueiredo et al., 2007; Siwik et al., 2009). The

processes of Hg deposition from air to forest soil include through-fall, litterfall and dry deposition of gaseous Hg directly to the ground surface (Pokharel and Obrist, 2011; Siwik et al., 2009; St. Louis et al., 2001). Although deposited Hg is mostly absorbed by organic matter (with C–, N–, or S– functional group) in soil after mineralization (Grigal, 2003; Obrist et al., 2009; Pokharel and Obrist, 2011), Hg can re-emit to the atmosphere (Pokharel and Obrist, 2011; Selin, 2009) and be leached with dissolved organic matter (DOM) into runoff or soil solution (Akerblom et al., 2008; Kocman et al., 2011). Forest canopies and soil regulate the magnitude and timing of the delivery of Hg to downstream aquatic ecosystems. Thus, forests influence regional Hg cycling.

The Tibetan Plateau is considered a remote area with minimal anthropogenic activities and is surrounded by high-pollution regions (South/East Asia). Loewen et al. (2007) reported the atmospheric transport of mercury to the Tibetan Plateau. They found Hg deposition in association with particulate matter as a significant depositional process (Loewen et al., 2007). Recently, Yang et al. (2010) reported the Hg accumulation rates in lake sediment of the Tibetan Plateau. In their study, the increase in Hg deposition flux over the last few decades is synchronous with the recent economic development in Asia (especially China and India, Yang et al., 2010). In addition, Zhang et al. (2002) investigated the concentrations of 13 elements in the soils of the Tibetan Plateau and noted that the element's distribution patterns were influenced by vegetation type. The mercury concentration pattern was found to

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be the greatest in forest soils, followed by shrub soils and meadow soils, with grassland soils having the lowest Hg concentration (Zhang et al., 2002).

The forest region of the Tibetan Plateau is the highest-altitude forest in the world, extends from the central to the east Himalayas and forms the border between China and countries in south Asia. Fu et al. (2008a,b,c, 2009, 2010) conducted a series of studies on the patterns of Hg in air, foliage and soil throughout the eastern fringe of the Tibetan Plateau. They concluded that due to the local emission, the total gaseous Hg and Hg deposition flux displayed relatively higher values in low-altitude regions (Fu et al., 2009, 2010). It was also reported in Fu et al.'s study that the forest in the eastern Tibetan Plateau was a large pool of atmospheric Hg, and nearly 76% of the atmospheric Hg input was stored in forest soil (Fu et al., 2010). However, the area of the Tibetan forest is more than 3×10^4 km², and it is essential to investigate the fate and cycling of Hg in the Tibetan forest system on a larger scale.

In this study, we investigated the Hg concentrations in foliage and soil profiles of the Tibetan forest, which is located in the eastern/southeastern Tibetan Plateau (Fig. S1). The sampling sites span 2000 km in longitude and 500 km in latitude. The primary objectives of this study were (1) to investigate the longitudinal and altitudinal distribution of foliar Hg to study the sources and transport of Hg in the Tibetan forest, (2) to estimate the air-to-forest ground Hg deposition loads and the Hg losses from surface ground and (3) to study the vertical distribution of Hg and the total amount of Hg stored in forest soil pits to investigate the Hg cycling in the Tibetan forest.

2. Materials and methods

2.1. Study area and sample collection

The sampling sites were located between 88 and 105°E, and altitudes ranged from 351 to 4400 m (Table S1 and Table S2 in the Electronic Supplementary Material). The foliage was collected from west to east in the following four transects: Yadong (YD), Linzhi (LZ), Nujiang River (NR), and Sichuan (SC) (Fig. S1). Detailed information on the four transects is provided in Text S1. The foliage was collected in August 2008, and fully developed undamaged needles (approximately 1 year old) were cut from trees at heights of ~2 m above the ground using scissors, before being subsequently stored in plastic bags at -18 °C until extraction (Mélières et al., 2003).

Soil samples were collected from a total 7 soil pits dug in the forested range (Sygera Mountain, 3300–4400 m) of the LZ transect. The ecosystem, bedrock/soil characteristic, and hydrology of this region are described in Text S2. Four of the soil pits were dug on the northern slope between 4170 and 4400 m in both forested areas (two of pits) and clearings (two of pits). The remaining three soil pits were dug on the southern slope with two under forest canopy (4170 and 4400 m) and one in a clearing (at 3300 m) in a valley. The soil layers were classified using the United States Department of Agriculture (USDA) Soil Taxonomy guidelines, and the information of layers is listed in Text S3 and Table S2. The pits were sampled using stainless-steel hand trowels and clean latex gloves. To avoid contaminating samples, the trowels were washed using deionized water before sampling. Approximately 100 g of soil was collected for each layer, packed in plastic bags, mailed to laboratory immediately and frozen until analysis.

Before analysis, the soil samples were freeze dried, manually ground, and sieved through a 0.2-mm mesh to remove large debris, roots and other rubble. The foliage was also freeze dried and ground.

2.2. Analytical methods

A soil sample of approximately 0.1 g was extracted using 5 mL of mixed solution (2 mol/L HNO₃ and 4 mol/L HCl) at 100 °C for 1 h in rigorously acid-leached silicic borate beakers. Litter or foliage sample (0.05 g) was digested by 10 mL pressurized acid digestion using a mixture of 2 mL HNO₃/5 mL H₂O₂ at 120 °C for 3 h. After digestion, 2 mL of K₂Cr₂O₇ solution (1 mmol/L) was added to all solutions, which were then diluted to 10 mL with deionized water. Total Hg (THg) concentrations were measured by cold vapor atomic fluorescence spectrophotometry (CVAFS) following reduction with SnCl₂.

The carbon contents of the soil (total carbon, TC; and total organic carbon, TOC) were measured using a TOC analyzer (Shimadzu 5000-A, Japan). Total nitrogen (TN) concentrations of the soil samples were measured with a Kjeldahl nitrogen analyzer (KDY-9820, Beijing Ruihang Xingye Science & Technology Co., LTD). The dry bulk density (BD) was determined by drying and weighing in cylinders of known volume.

2.3. Quality assurance/Quality control

The accuracy of the Hg method was assessed by analyzing standard reference materials. The standard reference materials (soil GB GSS-2 and foliage GB GSW-11) were supplied by National Institute of Metrology P.R. China (NIM), and their certified values were 15 ± 3 and 150 ± 20 ng/g, respectively. The values obtained from standard reference material were within $104.8 \pm 22.7\%$ and $94.6 \pm 9.7\%$ of the certified values. To evaluate potential laboratory contamination, procedural blanks were processed every 15 samples. Method detection limits (MDLs) were set at 3 times the standard deviation of the mean blank concentrations. The MDL based on a 0.1 g soil sample was 1.8 ng/g (0.02 pg/μl). All results were blank corrected, and three replicates were taken for each sample.

2.4. Estimated methods of soil age

Soil age is considered a factor influencing the distribution of Hg in soil profile. ²¹⁰Pb was used as an indicator of soil age and Hg deposition (Klaminder et al., 2008; Stankwitz et al., 2012). In this study, the dating methods included the age estimation of the Oi layer using litter decomposition rates and of other layers using sedimentation rates derived using radioactive ²¹⁰Pb (Moeckel et al., 2009). The detailed methods are described in Text S4. In addition, the Pb leaching may introduce uncertainties for dating the soil age, as discussed in Text S4.

2.5. Estimation methods of input and output fluxes

For studying the processes of Hg cycling, the fluxes of Hg input (air-to-forest ground input, litterfall deposition) and the fluxes of Hg losses from organic layers (leaching to deeper layers and runoff) were estimated in this study.

2.5.1. Input of Hg

The litterfall deposition fluxes in the forest (F_{litter} , μg/m²/year) are an important process for 'dumping' atmospheric Hg to forest ground. F_{litter} can be estimated by multiplying the annually deposited litter masses ($m_{litter/year}$, g/m²/year) and the Hg concentrations in the litter (C_{litter} , μg/g). The equation is as follows:

$$F_{litter} = C_{litter} m_{litter/year} \quad (1)$$

Assuming THg concentrations in leaves accumulate linearly over the growing season (Bushey et al., 2008; Laacouri et al., 2013; Millhollen et al., 2006; Siwik et al., 2009), the litterfall THg concentrations were approximately estimated by multiplying the measured foliar THg concentration in LZ forest by a factor of 1.5 (Fu et al., 2010).

The net air-to-forest ground fluxes (F_{Oi} , ng/m²/year), including litterfall deposition, were estimated from the age of the Oi layer (t_{Oi}), the THg concentration in the Oi layer (C_{Oi}) and the forest ground area specific mass of the Oi layer (m_{Oi}):

$$F_{Oi} = \frac{C_{Oi} m_{Oi}}{t_{Oi}} \quad (2)$$

2.5.2. Losses of Hg

Interflow will carry the Hg in the ground surface to the deeper soil layers and finally to the low-land water bodies. The leaching of Hg to deeper layers is influenced by dissolved organic carbon (DOC) (Akerblom et al., 2008; Grigal, 2003; Schwesig and Matzner, 2000; Tipping et al., 2011). The leaching from the ground surface to the deeper layers (Hg_{leach} , μg/m²/year) can be calculated by an equation introduced by Tipping et al. (Tipping et al., 2011):

$$Hg_{leach} = c_{Oi} DOM_{leach} / OM_{Oi} \quad (3)$$

where c_{Oi} is the average concentrations of Hg measured in the organic layers (μg/g), and DOM_{leach} and OM_{Oi} are the dissolved organic matters in the stream leaching into the deeper layer (g/m²) and the total organic matter in the litter or mineral soil (g/m²), respectively. The DOM in the organic layer is calculated using the C/N ratios (Aitkenhead and McDowell, 2000):

$$DOM = 1.5908 \times (C/N) + 13.342 \quad (4)$$

and the DOM_{leach} is estimated using the following equation (Tipping et al., 2011):

$$DOM_{leach} = 21.6\% P \times DOM \quad (5)$$

where P is the annual precipitation. The coefficient of 21.6% is the correlation between the rainfall and the leaching water (Text S2, Wang, 2002).

Runoff is also an important pathway of Hg loss. The loss flux of Hg in runoff (Hg_{runoff}) was calculated as (Scholtz et al., 2003):

$$Hg_{runoff} = F_W \times c_W \quad (6)$$

where F_W is the water flow of surface runoff, which was assumed to be $0.8\text{--}2.5$ m³/m²/year (10%–25% of precipitation); c_W is the concentration of THg in river water. The c_W was assumed to be 1.5 μg/m³, which was the concentration of THg measured in the river water of LZ region reported by Zheng et al. (2010).

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