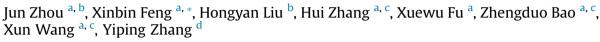
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Examination of total mercury inputs by precipitation and litterfall in a remote upland forest of Southwestern China



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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.

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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 μ g m⁻² yr⁻¹. Litterfall Hg depositions were the major pathway for Hg loading to the forest catchment, which were 71.2 μ g 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.

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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





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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 $(24^{\circ}32'N, 101^{\circ}01'E)$ – 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% HNO₃), rinsing with ultrapure deionized water (18 MQ cm), and then triple rinsed with ultrapure deionized water (18 MQ cm) and finally, doubled-bagged, stored in a plastic boxes

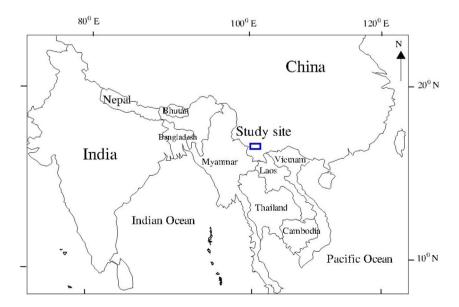


Fig. 1. The location of the study site.

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