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Mercury in terrestrial forested systems with highly elevated mercury deposition in southwestern China: The risk to insects and potential release from wildfires

Jun Zhou [a, b], Zhangwei Wang [a, *], Ting Sun [a, b], Huan Zhang [a, b], Xiaoshan Zhang [a]

[a] Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, Beijing 100085, China
[b] University of Chinese Academy of Sciences, Beijing 100049, China

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ABSTRACT

Forests are considered a pool of mercury in the global mercury cycle. However, few studies have investigated the distribution of mercury in the forested systems in China. Tieshanping forest catchment in southwest China was impacted by mercury emissions from industrial activities and coal combustions. Our work studied mercury content in atmosphere, soil, vegetation and insect with a view to estimating the potential for mercury release during forest fires. Results of the present study showed that total gaseous mercury (TGM) was highly elevated and the annual mean concentration was 3.51 ± 1.39 ng m^{-2}. Of the vegetation tissues, the mercury concentration follows the order of leaf/needle > root > bark > branch > bole wood for each species. Total ecosystem mercury pool was 103.5 mg m^{-2} and about 99.4% of the mercury resides in soil layers (0–40 cm). The remaining 0.6% (0.50 mg m^{-2}) of mercury was stored in biomass. The large mercury stocks in the forest ecosystem pose a serious threat for large pluses to the atmospheric mercury during potential wildfires and additional ecological stress to forest insect: dung beetles, cicada and longicorn, with mercury concentration of 1983 ± 446, 49 ± 38 and 7 ± 5 ng g^{-1}, respectively. Hence, the results obtained in the present study has implications for global estimates of mercury storage in forests, risks to forest insect and potential release to the atmosphere during wildfires.

1. Introduction

Mercury is a significant global contaminant due to its biogeochemical properties and its toxicity that can affect the health of people and ecosystems (US EPA, 1997). Anthropogenic activities related to industrialism disperse mercury, mainly inorganic mercury, which can be transformed by bacteria to methylmercury and biomagnified through aquatic and terrestrial food webs resulting in human health concerns through fish and rice consumption (Clarkson, 1998; Zhou et al., 2015a). Unlike other heavy metals, the biogeochemical cycling of mercury between atmosphere and forest terrestrial ecosystem is particularly important for subtropical ecosystems because huge amount of biomass was contained in subtropical forests and peat lands and forest was supposed to a large sinks of atmospheric mercury (Zhou et al., 2013, 2015b; Stankwitz et al., 2012). Coupled with global warming accelerated in northern latitudes (Raisanen, 1997) and high drought frequency in southwest China (Zhang and Zhou, 2015), these were of great scientific and public interest because of heightening wildfire activity and increasing mercury and carbon release to the atmosphere (Melendez-Perez et al., 2014; Friedli et al., 2007).

Vegetation is known to exert significant influence on the atmospheric mercury deposition and soil mercury evasion in the terrestrial ecosystem (Wang et al., 2009; Choi and Holsen, 2009; Mazur et al., 2014). Total gaseous mercury (TGM) in the atmosphere makes it likely that the dry deposition flux via stomatal uptake constitutes a large component of total dry deposition (Zhou et al., 2013). Upon deposition to the ground in throughfall or contained in biomass material (senesced leaves, needles, bark, twigs and dead wood), mercury is sequestered by reduced sulfur groups in the organic soil (Grigal, 2003; Obrist et al., 2009), re-emitted back to the atmosphere (Choi and Holsen, 2009; Mazur et al., 2014) and leached into runoff or soil solution (Wang et al., 2009; Grigal et al., 2000). Atmospheric mercury deposition and soil

* Corresponding author.
E-mail address: wangzhw@rcees.ac.cn (Z. Wang).

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mercury pool in terrestrial ecosystems have been extensively studied over the past decade (Grigal et al., 2000; Grigal, 2003; Obrist et al., 2009), yet little work has been done to assess mercury concentrations in organisms occupying forest ecosystems. Insects are the important constituent of the ecosystem and provide a significant source of food for wildlife such as spider, birds and some other small mammals. It is reported that some insects accumulate large amount of mercury in their bodies and result in physiological toxicity (Wyman et al., 2011; Townsend et al., 2014). Additionally, mercury would be transported to organisms on the higher position from insects along food chains (Zhang et al., 2010).

As an economic giant of the world, China has experienced dramatic economic growth over the past three decades, which was proved to be the world's largest energy consumer and anthropogenic mercury emission. According to recent studies at subtropical forest in China, the biome has high concentrations of mercury in litter (Fu et al., 2010; Zhou et al., 2013). Some animals in forest ecosystems may be uniquely adapted to this environment (Holmes and Sawyer, 1975; Townsend et al., 2014) and may be highly vulnerable to increased mercury loads. However, few assessments exist to determine if biota in forest systems is bioaccumulated and impacted by atmospherically deposited mercury. Furthermore, in recent years, sections of forests in China suffer from massive wildfires, consuming on average up to millions of hectares each year and the annual average mercury emission from forest fires was 0.78 t during 2000–2010 (Chen et al., 2013).

In the current study, we investigated the mercury concentrations in air, soil, plant and insect of the TFP forest in southwestern China. However, this is the first study, to the best of our knowledge, to explore mercury bioaccumulation in insects and to assess the mercury distribution and pool in biomass as well as potential risk of mercury emission caused by fires in forest of China. The primary objective of this paper is (1) to quantify the mercury distribution in air, soil, vegetation, insect and mercury stocks in the forest ecosystem, and (2) to estimate the potential risk to insects and mercury release during future wildfires.

2. Materials and methods

2.1. Study area

A field experiment was carried out in a Masson pine (*Pinus massoniana* Lamb.) stand planted in 1962 following the clear cutting of a natural Masson pine forest at Tieshanping Forest Park (106°41.24′E, 29°37.42′), which is located about 20 km northeast of the center of Chongqing City, at an elevation from 200 to 550 m (Fig. 1). TFP is one of the Sino-Norwegian multidisciplinary Integrated Monitoring Program on Acidification of Chinese Terrestrial Systems (IMPACTS) project monitoring sites. The climate is mainly controlled by the southwest monsoon with a mean annual precipitation of 1028 mm and 75% of the rainfall occurs during wet seasons (May–October). Annual mean air temperature in the study area is 18.2 °C. Due to warm and wet summer, soil organic matter turnover rates are high (Raich and Schlesinger, 1992; Zhu et al., 2013). The stand is homogeneous, dominated by Masson pine and some associated species such as *Cunninghamia lanceolata* (Lamb.) Hook. and *Schima superba* Gardn. et Champ. Clay mineralogy is dominated by kaolinite and the soil is typically mountain yellow earth (corresponding to a Haplic Acrisol in FAO; Surhone et al., 2010) and its texture is haplic acrisol/alisol.

Chongqing is an important industrial region in southwest China, and consumes large amount of coal. Consequently, annual mercury emissions just from coal-fired was 4.97 t (Wang et al., 2006) and mercury pollution was regarded as major environmental burdens in Chongqing (Yang et al., 2009). Via atmospheric long-distance transport, large amounts of mercury deposited to the surrounding region of the city. Based on our previous studies here, the annual total input fluxes of mercury were 291.2 μg m^{-2} yr^{-1} (Wang et al., 2009), which was several or even dozens of times higher than those in North America and Europe (Zhou et al., 2016). About over two-thirds of the deposited mercury was resident in the forest, which resulted in elevated mercury concentration in organic horizons (191 ± 65 ng g^{-1}) and mercury pool (8.2 ± 2.6 mg m^{-2}), (Zhou et al., 2015b), and thus it can be deduced that the subtropical forest was a great sink of atmospheric mercury.

2.2. TGM in atmosphere

The collection and measurement of TGM samples as well as the quality control procedure have been described in details in our previous study (Zhou et al., 2015b). Briefly, TGM was continuous monitoring in four seasons from March 2014 to January 2015 over the canopy in the forest with manual pure gold quartz trap. The gold traps were sampled at 8:00 am and 6:00 pm every day, representing daytime and night, respectively. All the gold traps were brought back to TFP Forestry Station for mercury quantification by CVAFS detector (Brooks Rand III, US EPA, 1999) using dual gold trap amalgamation procedure after every sampling.

2.3. Mercury in soil, vegetation and insect

In this study, three sample plots of 10 × 10 m^2 were set up in January 2015. A buffer strip of about 100 m surrounded each of the plots. The plots were all quite flat, with slope less than 10°. Ground organic material and mineral soil samples were collected from 20 cm × 20 cm squares and transported in polyethylene bags with airtight seals. The organic material included litter (leaves and needles, small twigs), organic soil layers (O horizons) which were collected as horizontal slices and assigned Oi (undecomposed surface litter), Oe (partially decomposed litter), Oa (decomposed organic humus, and coarse woody debris) designations based on the approximate degree of decomposition. Samples in the depth mineral soil profiles were collected from top layer to the depth of 40 cm.

Samples of herb standing vegetation (*Dicranopteris pedata*, *Woodwardia japonica* and moss) were collected from the subplots and separated into root, stem and leaf. Samples of shrub were taken from *Randia cochinchinensis* and *Loropetalum chinense including root, stem, branch and leave in each subplot. Samples of overstory collected from* Masson pine were needles, bark, cores and roots. Each sample was collected in triplicate in each plot. To calculate the mercury stocks in vegetation, concentrations were assumed to be the same for understory and overstory plant components.

Three insect species which were cicada (*Platypleura kaempferi*, *Chremistica ochracea* and *Oncotympana maculaticollis*), longicorn (*Anoplophora chinensis*) and dung beetle (*Geotrupidae*) were collected by hand in June and August, 2014 and sealed in polythene bags. Samples of insects were froze immediately in a refrigerator at −4 °C until used.

Soil samples were air-dried in a clean environment and vegetation samples were washed thoroughly with tap water and then with deionized water, dried at 60 °C for 48 h to constant weight. The insect samples were washed thoroughly with deionized water and then freezing-dried for 72 h in the laboratory. Subsequently, plant and insect samples were completely ground to a fine powder in a pre-cleaned food blender, passed through 100 mesh and stored in polyethylene bags to avoid cross-contamination. Practices to avoid mercury contamination included wearing clean polyethylene gloves, using pre-cleaned plastic tools and double-bagging all samples.