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The fate of mercury and its relationship with carbon, nitrogen and bacterial communities during litter decomposing in two subtropical forests

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ABSTRACT

Forests play a significant role in the biogeochemical cycling of mercury (Hg). Litterfall represent a dominant pathway for Hg to reach the ground surface under a forest canopy. In order to understand Hg accumulation in the litterfall, the dynamics of Hg, carbon (C), nitrogen (N), microbial C and N, as well as bacterial community in the decaying litterfall of two typical subtropical forest stands in southwest China were investigated for one year. THg levels in the litterfall after one-year increased to 124.64% and 135.90%, and MeHg levels in the litterfall increased up to 295.65% and 209.38% of the initial values in the mixed broadleaf-conifer forest and evergreen broadleaf forest respectively. Differently, the concentrations of THg and MeHg in the organic layer of the underlying soil were quite stable. The concentrations of THg in the decaying litterfall corresponded negatively with C/N ratios. Bacterial community analysis found that the bacteria previously being confirmed as Hg methylators did not occur in the genera of the decomposing litterfall in the two forest stands, which might imply that the increase of MeHg during decomposing did not mainly due to the contribution of confirmed Hg methylators, and other sources of MeHg might exert certain roles.

1. Introduction

Mercury (Hg) is regulated as a hazardous pollutant, which is released from both natural and anthropogenic sources. Large amounts of Hg have been emitted into the environment, which makes it a global health concern due to its toxicity (Abelson, 1970; Boening, 2000; Driscoll et al., 2013). Forest ecosystem is the largest terrestrial ecosystem, the area of which is approximately one third of the Earth, and the biomass of which accounts for about 85% of the terrestrial ecosystems (Abella, 2015). Forest ecosystem is generally considered to be an active library of Hg, and environmental behavior of Hg in the forest ecosystem is believed to be an important part of global Hg cycle (Ericksen et al., 2003; Kim et al., 1995). Forest canopies can uptake atmospheric Hg more rapidly than other landscapes due to their large leaf areas and rough surfaces (Risch et al., 2012). Mercury input to the forest floor includes dry deposition on the leaf surface, litterfall, throughfall, dry deposition directly on the forest floor and wet deposition during leaf-off periods (Choi, 2007; Demers et al., 2007). Mercury dry deposition through litterfall means that the leaves become contaminated via atmospherically deposited Hg and accumulated Hg externally and internally as a function of leaf age (Ericksen et al., 2003; Rea et al., 2000; Tyler, 2005; Tyler and Olsson, 2006; Zhang et al., 2005). Previous studies have indicated that Hg deposition through litterfall is significantly greater than wet deposition (Demers et al., 2013; Grigal, 2002; Jiskra et al., 2015) and represents a dominant pathway for Hg to reach the ground surface under a forest canopy (Lindberg et al., 1994; Munthe et al., 1995; Rea et al., 1996, 2001). Namely Hg deposition through litterfall have much greater effect than wet deposition on the size of Hg storage in the forest soil. Therefore, Hg dry deposition from the air through litterfall had attracted more

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and more attention from researchers worldwide (Fu et al., 2015; Risch et al., 2012; Zhang et al., 2012). The annual quantity of global Hg deposition through litterfall is estimated to be 1180 ± 710 mg yr^{-1} through statistical analysis based on published data, approximately 70% of which occurs in the tropical and subtropical regions (Wang et al., 2016). This systematic evaluation on the role of Hg deposition through litterfall pointed out the scarce studies on the fate of litterfall Hg in the tropical/subtropical forests, and significantly highlighted the role of tropical/subtropical forests in global Hg cycling (Wang et al., 2016). The distribution and dynamics of soil Hg of Asian subtropical forest have been documented for seven years (Lu et al., 2016), and Hg in leaf litter in typical suburban and urban broadleaf forests were also reported (Niu et al., 2011). However, there is still limited understanding about the dynamics and fate of accumulated Hg through litterfall in the subtropical forests.

Moreover, researchers found that MeHg concentrations in the litterfall increased in the decaying process, depending on the plant type (Hall and St Louis, 2004). Our unpublished results also found this interesting phenomenon and proposed a hypothesis that MeHg levels in the decomposing litterfall increased in the subtropical forests. If this hypothesis establishes, then it implies that the risk of Hg migration from the forest system to the aquatic system may also increase. Previous research speculated that THg levels corresponded with microorganisms that involved in nitrogen fixation, especially fungi (Demers et al., 2007; Hall and St Louis, 2004), and found that the quantities of bacteria during litter decomposing were much larger than fungi (Wang et al., 2006). Moreover, MeHg is produced in the environment via biotic and abiotic processes (Li and Cai, 2013), among which a series of anaerobic bacteria and archaea is the predominant way (Raposo et al., 2008). That is to say, the concentrations of THg and MeHg change with the degradation of litterfall, in which a series of microorganisms are mainly involved. Therefore, it is important to research on the biotic formation of MeHg and understanding the role of the bacterial community structure in the litterfall decomposing process (Strickman and Mitchell, 2016; Macalady et al., 2000; Bae et al., 2014). In addition, nitrogen fixation seems to play significant roles in Hg emission and accumulation, which means that it is essential to research on the dynamics of Hg and its relationship with the concentrations of nitrogen during litter decomposing. Therefore, we need to explore the relationship of Hg concentrations with microorganisms that maybe involved in this process. However, little is known about whether bacteria play certain roles in the emission and accumulation of Hg during the degradation of litterfall and what kind of bacteria dominate in this process, even the bacterial community structures was unknown so far. To date, less research on the dynamics of bacteria during litterfall decaying had been conducted. Thus, the objectives of this study were: 1) to characterize Hg dynamics and fate in the litterfall and forest soil in two typical subtropical forest in southwest China; 2) to analyze the relationship between the dynamics of litterfall THg and carbon nitrogen ratios during litterfall decaying; 3) to determine the relationship between the dynamics of litterfall MeHg and bacterial community structure during the whole degradation process.

2. Methods

2.1. Study area description

Chongqing is situated at the transitional area between the Qinghai-Tibet Plateau and the plain on the middle and lower reaches of the Yangtze River in the subtropical climate zone. This study was conducted at the Simian Mountain, a national nature reserve (106° 22′–106° 25′ E, 28° 35′–28° 39′ N) located about 200 km from urban Chongqing. Mt. Simian is a typical subtropical forest of southwest China. It possesses the largest and most well preserved primitive subtropical evergreen broadleaf forest at 28° north latitude in the northern hemisphere, and the forest coverage has reached 95.8%. It is known as the “natural species gene pool” by the specialists of ecological protection of the United Nations (Zhang et al., 2011). The dominant land over of Mt. Simian is natural or secondary vegetation with more than 1422 species of vascular plants, mainly including mixed broadleaf-conifer forest (90%) and evergreen broadleaf forest (8%). The natural conditions are suitable for vegetation growing and landscape resources are abundant in this area. The topography is characterized by a low mountainous landscape. The climate is predominantly subtropical humid monsoon with a relative humidity over 90%, annual average temperature and rainfall of 13.7 °C and 1127 mm, respectively. At present, there are no significant point source discharges of Hg in the forest floor of Mt. Simian. In our research, the mixed broadleaf-conifer forest (M forest for short) and evergreen broadleaf forest (B forest for short) were selected as the two forest stands. The two forest stands are two adjacent sites, about 500 m apart from each other. Within the organic horizon, the *Oi* horizon means soil at the range of 0–10 cm, and *Oe* horizon is the soil at 10–20 cm in thickness in each stand. The background information about the two forests are shown in Table S1.

2.2. Decomposition experiment design and sample collection

Experimental research on the litter decomposition at the two forest stands was investigated by litterbag technique. A total of 10 nylon-mesh-lined baskets were acid cleaned and randomly placed throughout each forest stand to collect fresh leaf litter. Leaves were collected during rain-free periods to make sure that Hg was not leached from the fallen litter before the deployment of litterbags (Demers et al., 2007). Leaves from each stand were weighed into 108 acid-cleaned 1-mm mesh bags (10*15 cm, 10.0 g/bag), labeled, and heat-sealed, with 36 bags per plot. Nine litterbags (3 bags per plot) were immediately removed from each forest stand for analysis of initial mass, carbon, nitrogen, THg and MeHg contents on a dry mass basis and DNA extraction. The remained 99 litterbags were returned to the field and split between three 1 × 1 m plots (33 bags per plot) in each forest stand. Each litterbag was pinned to the ground with small plastic stakes at the four corners. Litterbags were not covered with additional fresh litter upon deployment. The litter was collected monthly for one year from March 2014 to February 2015. Namely 9 decomposing litterbags were taken from the three plots of each forest stand every month. For the three bags moved to the lab from one plot each month, one bag was used to measure various environmental factors, the second was used to analyze the concentrations of THg, MeHg, microbial C and N, and the last one was used to extract DNA. For each sampling, the litter samples were shipped to the laboratory and air dried within 2 h, and then weighed to determine dry mass loss. The corresponding soils under the bags were also collected at depth of 0–10 cm (O_i) and 10–20 cm (O_e) each month together with bags.

According to the exponential regress model, the relative rates of decomposition (k) were calculated after Olson model (Olson, 1963):

$$X_t = X_o\, e^{-kt} \quad (1)$$

where: X_o is a weight of initial material, Xt is a weight of decaying material after time t, K is a rate of decomposition, e is a base of natural logarithm.

The mass of THg and MeHg in each litterbag was calculated by multiplying the THg or MeHg concentrations by the final freeze-dried weight of litter tissues in the litterbags (Hall and St Louis,