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Research article

High mercury accumulation in two subtropical evergreen forests in South China and potential determinants

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ABSTRACT

Forests play an important role in global mercury (Hg) cycling. To explain the high Hg accumulation in subtropical forest ecosystems, we studied temporal dynamics of Hg, carbon (C), nitrogen (N), and sulfur (S) in forest soil profiles, as well as litterfall flux and precipitation, in an old-growth moist evergreen broadleaf (EB) forest and a mossy coppice (MC) forest from South China over seven years. The mean soil Hg concentration was 257 ± 14 ng g^{-1} in the O-horizon and 248 ± 15 ng g^{-1} in the A-horizon for the EB forest, and 94 ± 27 ng g^{-1} in the O-horizon and 70 ± 11 ng g^{-1} in the A-horizon for the MC forest. Annual variations in Hg concentration were suggested to be associated with variations in precipitation and litterfall biomass. Significant vertical Hg transport was only observed in the MC forest, which was attributed to its lower organic matter content. Correlation and stoichiometry analyses further suggested that the dynamics in Hg concentration in the forest floor was also closely linked to the variation in S concentration. Additionally, the difference in the soil Hg pool between these two forests was attributed to different litterfall biomass fluxes.

1. Introduction

Mercury (Hg) has been treated as a primary pollutant by the United States Environmental Protection Agency and the World Health Organization. Forest ecosystems play an important role in global Hg cycling (Lindberg et al., 2007). Forest Hg mass assessments have shown that the forest floor is a stable Hg sink (Fu et al., 2010; St Louis et al., 2001; Wang et al., 2009). Hg accumulated in the forest soil can be methylated through microbial and abiotic processes (Allan et al., 2001; Carpi et al., 1997). The product methyl Hg (MeHg), which has a strong bioaccumulation capability, is transported to aquatic ecosystems via surface runoff leading to elevated MeHg levels in aquatic food webs (Allan et al., 2001; Eklof et al., 2013, 2014; Hultberg et al., 1995). Recently, a study of the spatial distribution of soil Hg suggests that the forest soil in South China is more prone to accumulating Hg than that in North China (Luo et al., 2014). Forest type has been identified as an important factor that influences soil Hg sink function (Luo et al., 2014; Obrist et al., 2011). Forests in South China are mainly subtropical evergreen broadleaf forests. The distribution and dynamics of soil Hg of these forest ecosystems have not been well documented. In addition, these forests are facing threats from human activities, and their areas are decreasing. Therefore, understanding the process of Hg accumulation in the forest floors of these forests will greatly help the assessment of ecological risk of deforestation in South China.

Hg geochemical processes in forest soil include litterfall input, wet deposition, soil evasion, surface runoff, and groundwater output. The surface soil is rich in organic matter, which has a strong affinity for Hg. Therefore, groundwater output, surface runoff, and soil evasion would not substantially affect soil Hg accumulation (Grigal, 2002, 2003), and the soil Hg pool is mainly shaped by atmospheric Hg inputs. Earlier studies have shown that litterfall Hg inputs were 2–7 times higher than those of wet deposition, suggesting that Hg uptake by foliage is the key process that shapes soil Hg accumulation (Grigal, 2003). As Hg concentration in foliage correlates with atmospheric Hg concentration (Frescholtz et al.,

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2003; Gustin et al., 2008), the elevated atmospheric Hg concentration in South China (Fu et al., 2015) may lead to higher Hg deposition from litterfall in forests from this region.

The stoichiometry of Hg, carbon (C), nitrogen (N), and sulfur (S) in forest soil could help to understand Hg accumulation in forests. Legacy sequestration of atmospheric Hg in the forest has been shown to strongly link with C pools (Obrist et al., 2011, 2012). Subtropical forests in South China are among the largest terrestrial carbon sinks (Tan et al., 2012); however, it is unknown whether high carbon sequestration is associated with high Hg accumulation in the subtropics. Moreover, soil Hg also tends to have an affinity for reduced S groups (S^{-2}, SH^{-1}) and organic S groups (Zhang and Lindberg, 1999). In the presence of reduced S groups, the soil Hg storage capacity can reach up to 2.4×10^{-2} g g^{-1} (Smith-Downey et al., 2010). Understanding relationships and interactions among Hg, C, N, and S is important for assessing their potential contribution to the regional and global C and Hg cycles.

In the present study, we monitored soil Hg storage, soil C, N, and S dynamics, and litterfall production for seven years in two subtropical forests in South China. Our objectives were: (1) to characterize the soil Hg dynamics in Asian subtropical forests; (2) to quantify the interactions between Hg and other nutrient elements, such as C, N, and S for subtropical forests; and (3) to understand potential ecological mechanisms associated with the high Hg accumulation in remote forest ecosystems of South China.

2. Material and methods

2.1. Site descriptions

Our study sites are located in the Ailaoshan Station for Subtropical Forest Ecosystem Research Studies (ASSFERS, 24°32′N, 101°01′E, Fig. 1) at 2450–2650 m above sea level. ASSFERS is in a pristine remote area far away from air pollution sources, and the mean atmospheric Hg concentration of the forests in ASSFERS is ~2 ng m^3 (Fu et al., 2015). Two types of forests along the elevation gradient were studied. At around 2450 m of elevation, the ecosystem is an old-growth, montane, and moist evergreen broadleaf (EB) forest. The forest has a distinct vertical structure, with a 20–30 m canopy and a 95% average canopy coverage. Dominant species include *Lithocarpus xylocarpus*, *L. chintungensis*, *Schima noronhae*, and *Manglietia insignis*. At high elevations (above 2600 m), close to the top of the mountain, the ecosystem is a mossy coppice (MC) forest, with a 5–7 m high tree layer and an 85% average canopy coverage. The dominant species of the MC are not significantly different from those of the EB. The climate of this region is influenced by the southwest monsoon throughout the year, and it has distinct dry (from November to May) and rainy (June to September) seasons. The annual temperature at the EB forest site is 11.0 °C and is 10.0 °C at the MC forest site. The annual precipitation for both the EB and MC forest sites are ~1900 mm.

2.2. Sample collection and measurements

Soil profile samples were collected in the EB and MC forests. A typical forest site was chosen for each forest type for sampling. Twelve plots (10 × 10 m and numbered 1, 2, 3 … 12) were randomly selected in a 30 × 40 m transect for both the EB and MC forest sites. Two groups of six random plots (selected from the 12 plots) were sampled at the end of dry and rainy seasons for each forest site. Hence, 6 plot values × 7 years were obtained for each site. The mean Hg concentration from the 6 random plots did not show a significant difference from the mean concentration of the total 12 plots ($P = 0.957$) based on a test measurement in 2007. In each plot, the "S" type soil sampling method was used according to the operations of the Chinese Ecosystem Research Network (CERN). For each plot, 1–2-kg of soil samples of the O-horizon soil and 0–20 cm of mineral soil were collected. After natural drying, the soil samples were ground in an agate mortar and then sifted through a 200-mesh (74-μm) sieve. The total Hg concentration of each soil profile was measured with an RA-915 + multifunctional mercury analyzer (Lumex, Mission, BC, Canada). Total C and N and S concentrations were measured by the vario MACRO cube (Elementar, Hanau, Germany). Hg, C, N, and S measurements were performed in duplicates, and accepted variations of replicated measurements were less than 5%. Standard samples were measured in every ten samples, with recoveries ranging from 95 to 105%. GBW07405 (GSS-5) was used as the soil Hg standard, IVA99994 as the soil C and N standards, and AR-4018 as the soil S standard.

3. Results and discussion

3.1. Variations in soil Hg concentration

From 2007 to 2013, the mean soil Hg concentration was 257 ± 14 ng g^{-1} in the O-horizon and 248 ± 15 ng g^{-1} in the A-horizon in the EB forest (Fig. 2, Table S1). The soil Hg concentration did not differ significantly between the two soil layers (paired *t*-test; $P > 0.05$). The mean Hg concentration in the O-horizon exhibited a slowly decreasing trend over 2007–2013 ($R^2 = 0.42$, $P < 0.05$, Fig. S1). This may be related to decreased precipitation and litterfall production from 2007 to 2013, as the correlation coefficient was 0.73 between soil Hg concentrations of the EB forest and its annual precipitation, and 0.66 between the O-horizon Hg concentration and annual litterfall production (Fig. S2). Observations over a longer time period are needed to verify this hypothesis because the results were not statistically significant ($P > 0.05$). No significant difference in soil Hg concentration was found between the rainy and dry seasons for the two soil layers, indicating that surface runoff and vertical Hg transport may not be significant. This may be attributed to high soil C contents (19.6% for the O-horizon and 10.8% for the A-horizon, Table S1) in the soil organic matter, which can strongly bind Hg in upper soil profiles (Skyllberg et al., 2006).

In the MC forest, the mean Hg concentration was 94 ± 27 ng g^{-1} in the O-horizon and 70 ± 11 ng g^{-1} in the A-horizon (Fig. 2). The Hg concentration in the O-horizon was significantly higher than that in the A-horizon (paired *t*-test; $P < 0.05$), suggesting that the Hg sequestration process in the MC forest differed from that in the EB forest. Different from the EB forest, the Hg concentration in the A-horizon during the rainy season was significantly higher than for the dry season in the MC forest (paired *t*-test; $P = 0.02$, Fig. 2), which indicates vertical Hg transport. Furthermore, the Hg accumulation rate during 2007–2013 in the two soil layers of the MC forest showed opposite trends over time (Fig. S3), further verifying the above hypothesis. The C, N, and S concentrations in the A-horizon in the EB forest were 1.3–3 times higher than those in the MC forest (Table S1), which indicates that there is greater organic matter accumulation in the EB forest. Therefore, we propose that the difference in the seasonal cycle of soil Hg between the two types of forest mainly resulted from the impacts of organic matter.

Although the dominant tree species were similar in these two forests, the soil Hg pools in the two soil layers in the EB forest were 3 times higher than those in the MC forest (Fig. 3). A previous study in the EB forest has shown that Hg concentrations in the bottom soil are between 20 and 30 ng g^{-1}, indicating that the Hg in the O/A horizons resulted from atmospheric inputs and the weathering of litterfall (Zhou et al., 2013). The foliage Hg pool of the dominant tree species was 350 mg ha^{-1} in the EB forest, which was 3.2-times higher than that in the MC forest (Fig. 3) and is in agreement with

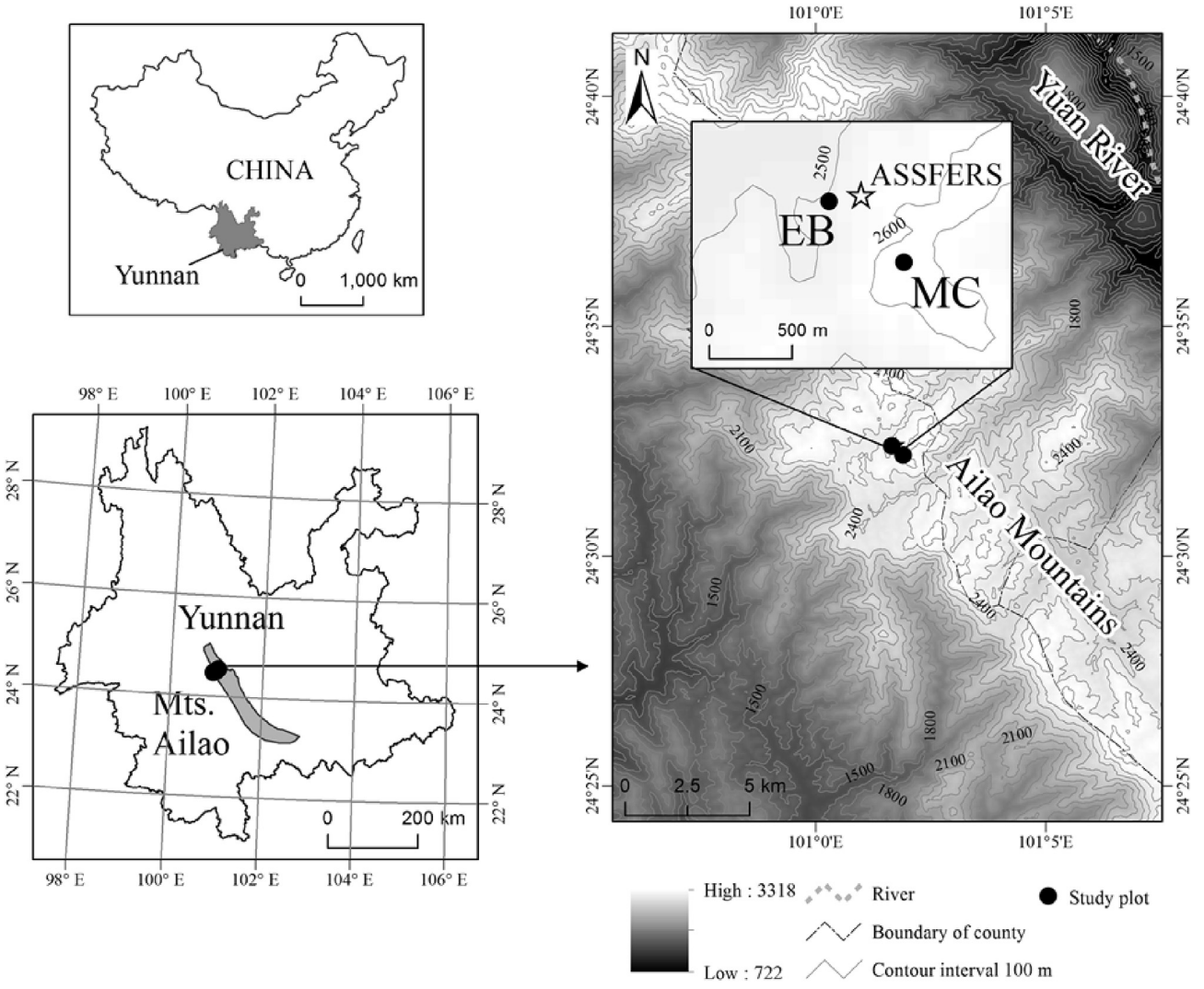

Fig. 1. Location of the study site.

the pattern found in soil Hg pools (Liu et al., 2002; Zhou et al., 2013).

Hg concentrations in the EB forest were higher than those reported for temperate forests in North America, Europe, and North China. Obrist et al. (2011) reported 126–134 ng g^{-1} for the O-horizon and 8–90 ng g^{-1} for the A-horizon in 14 temperate forests in the USA. Navratil et al. (2009) reported 142–258 ng g^{-1} for the O-horizon and 32–93 ng g^{-1} for the A-horizon in five forest sites in Central Europe. Luo et al. (2014) found concentrations of 44 ± 11 ng g^{-1} in the O-horizon in nine forests in Northeast China. The high soil Hg pool at the ASSFERS is probably caused by higher Hg deposition from litterfall, which is 75 μg m^{-2} yr^{-1} and can be up to one order of magnitude greater than that of temperate broadleaf forests (Blackwell and Driscoll, 2015; Demers et al., 2007; Ericksen et al., 2003; Juillerat et al., 2012; Larssen et al., 2008; Lee et al., 2000; Obrist et al., 2005, 2012, 2011; Zhou et al., 2013). Since China has a large area of subtropical forests (Zhang et al., 2016), information on Hg cycles in these forests could provide new insights into Hg pools in China.

The high atmospheric Hg concentration (2–5 ng m^{-3}) in South China (Fu et al., 2015) has been suggested to enhance Hg uptake by foliage, which would lead to elevated Hg deposition from litterfall. However, the mean Hg concentration in leaf litter of 17 subtropical evergreen forests in South China is 51 ± 39 ng g^{-1} (Niu et al., 2011), which is not significantly high compared to the mean value (45 ± 11 ng g^{-1}) for North American and European forests (Blackwell and Driscoll, 2015; Demers et al., 2007; Ericksen et al., 2003; Juillerat et al., 2012; Larssen et al., 2008; Lee et al., 2000; Obrist et al., 2005, 2012, 2011). This could be because besides atmospheric Hg concentration, other factors (e.g., solar irradiation, plant species, leaf lifespan, etc.) also significantly influence foliage Hg uptake (Blackwell and Driscoll, 2015; Ericksen and Gustin, 2004; Ericksen et al., 2003; Laacouri et al., 2013; Zhu et al., 2016). The combined effects from the other factors likely offset foliage Hg accumulation from elevated atmospheric Hg concentration in South China. An alternative explanation for the higher litterfall Hg inputs to soil could be greater total litter biomass (2–10 times the quantity of that in T/B forest) produced by subtropical evergreen forests (Ni and Song, 1998; Wang et al., 2009; Zhou et al., 2007, 2013). This calls for a re-assessment when more data are available.

3.2. Hg stoichiometry in relation to C, N, and S

The mean Hg/C ratios for the EB forest were 1.3 ± 0.3 and 2.4 ± 0.3 mg kg^{-1} in the O-horizon and A-horizon, respectively (Fig. 4a–4b). For the MC forest, the mean Hg/C ratio was 0.5 ± 0.2

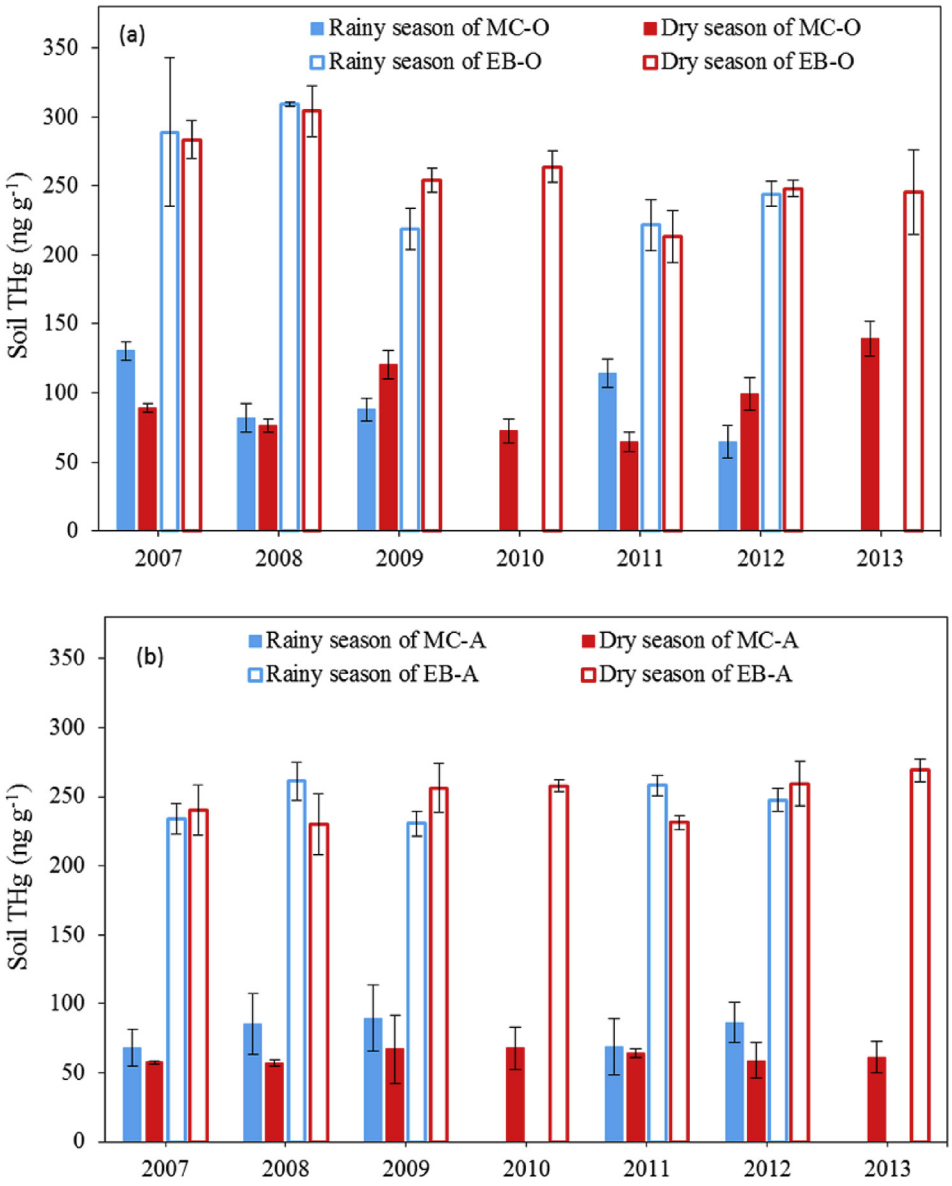

Fig. 2. (a) Hg concentration (means ± SEs) in O horizon during 2007–2013, and (b) Hg concentrations (means ± SEs) in A horizon during 2007–2013. EB is evergreen broadleaf forest, and MC is mossy coppice forest. 2010 rainy season data are missing due to bad weather conditions.

and 0.9 ± 0.2 mg kg^{-1} in the O-horizon and A-horizon, respectively (Fig. 4b). All the Hg/C ratios in soil were 3–14 times higher than those in foliage (Zhou et al., 2013). Temperate forests in Central Europe show comparable Hg/C ratios of 1.7 mg kg^{-1} in the O-horizon and 2.4 mg kg^{-1} in the A-horizon (Navratil et al., 2014). Obrist et al. (2011) reported Hg/C ratios of 0.7 mg kg^{-1} in the O-horizon and 3.1 mg kg^{-1} in the A-horizon in forests in North America. Two Swedish forest sites have Hg/C ratios of 0.63 and 0.93 mg kg^{-1} in the O-horizon and A-horizon (Akerblom et al., 2008), respectively, which are comparable to the ratios of the MC forest in the present study. The Hg/C ratios were higher in humus soil than in foliage at both the EB and MC forest sites, suggesting that during C mineralization, Hg accumulates in residual soil organic matter. The highest Hg/C ratios in mineral soil were attributed to the long-term mineralization of C and Hg accumulation in organic matter, as well as the adsorption of inorganic soil compounds (e.g., Al and Fe secondary minerals) (Akerblom et al., 2008). The EB forest has higher Hg/C ratios than the MC forest mainly because of higher atmospheric/foliar Hg inputs, i.e. higher leaf litter production.

The soil Hg concentration was significantly associated with the C concentration for the two different soil layers (O- and A-horizons) of the MC forest (Fig. 5a). However, there was no correlation between the Hg and C concentrations in the O-horizon of the EB forest. The absence of a relationship between the Hg and C concentrations does not necessarily suggest that Hg was not bound by soil organic matter in the EB forest. One potential explanation is that more inorganic C could physically mix with organic matter and that a significant relationship only reflects Hg absorbed by soil organic matter in the two layers of the MC forest and the A-horizon of the EB forest. This explanation is supported by the following observations: (1) during litter decomposition, the Hg concentration did not exhibit a positive correlation with the C concentration (Pokharel and Obrist, 2011); and (2) the soil organic C sink in the EB forest was 1–3 times greater than that in the MC forest (Liu et al., 2002).

The mean Hg/N ratios for the EB forest were 20.2 ± 3.6 and

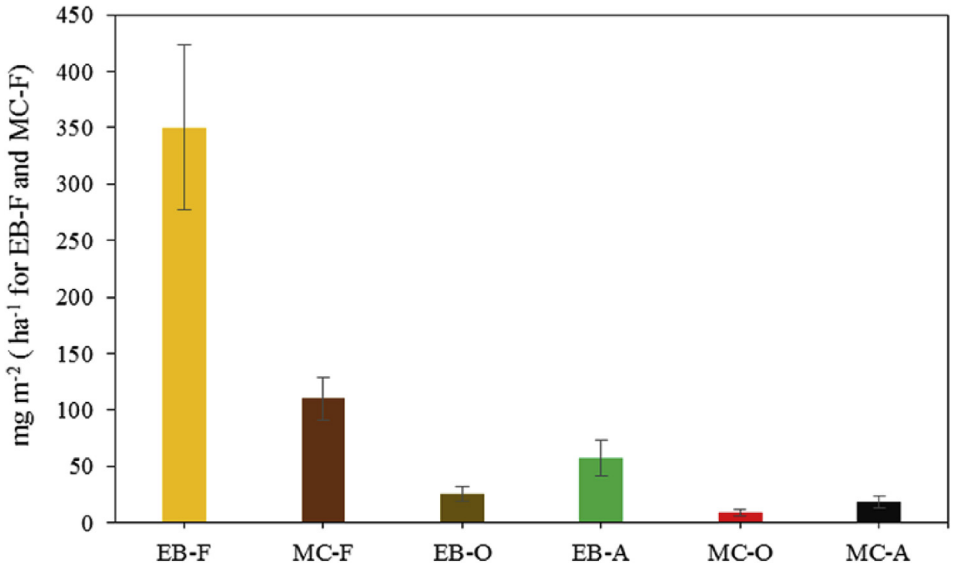

Fig. 3. Soil/foliage Hg pools (mg m^{-2}) for the mossy coppice (MC) forest and the evergreen broadleaf (EB) forest. F stands for foliage, O stands for soil O-horizon, and A stands for soil A-horizon.

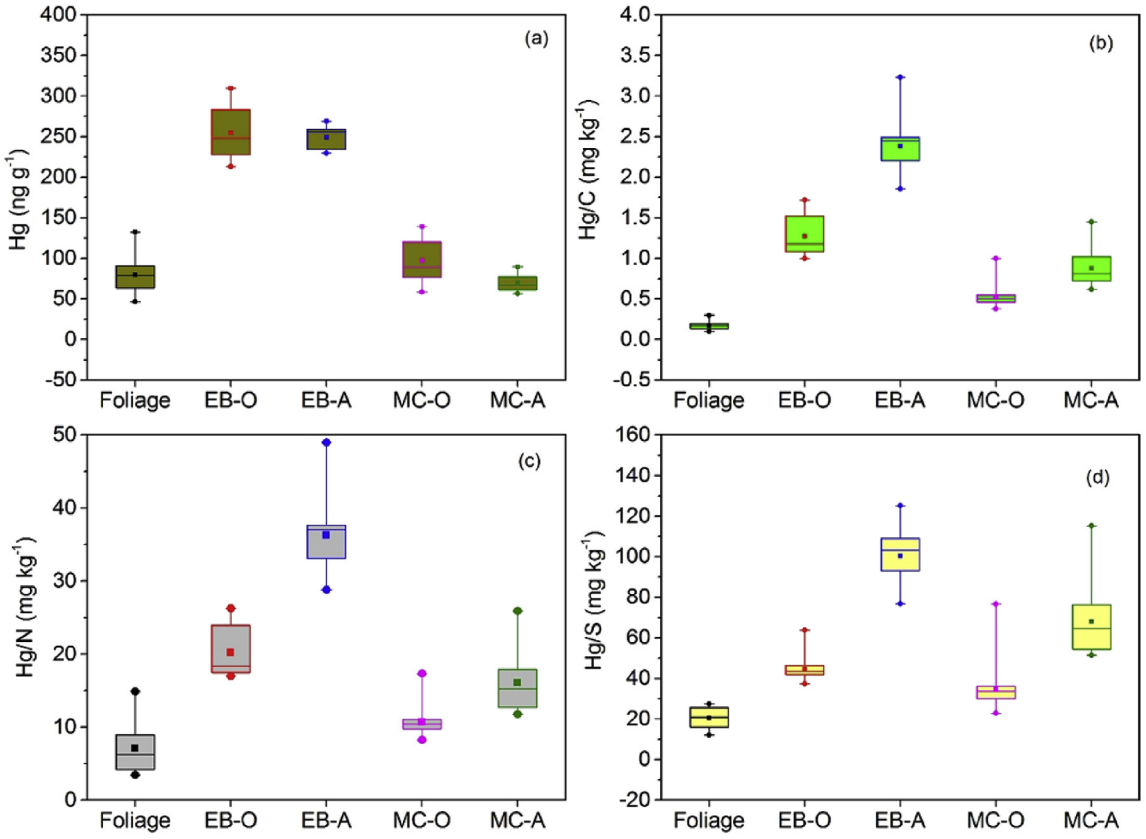

Fig. 4. Boxplots for the Hg concentration (a) and Hg/C (b), Hg/N (c), and Hg/S (d) ratios. The box boundaries represent the 5^{th}-95^{th} percentiles. The squares represent mean values. EB, evergreen broadleaf forest; MC, mossy coppice forest; O, O-horizon; A, A-horizon.

36.3 ± 5.2 mg kg^{-1} in the O-horizon and A-horizon, respectively (Fig. 4c). For the MC forest, the mean Hg/N ratios were 10.7 ± 2.3 and 16.1 ± 4.4 mg kg^{-1} in the O-horizon and A-horizon, respectively (Fig. 4c). The relationships between Hg and N concentrations were similar to those between Hg and C concentrations. In addition, the C/N ratio has been used as a tracer for the age of C (Obrist et al., 2011). C/N ratios in soil (the O-horizon and A-horizon) were lower than that of foliage (Fig. 6a), while Hg/C ratios increased with decreasing C/N ratios, indicating that Hg accumulated during soil development (Figs. 5a and 6b). The above trends are consistent with earlier studies in North America and Europe, and they have been explained as the internal (the original Hg in litter) and

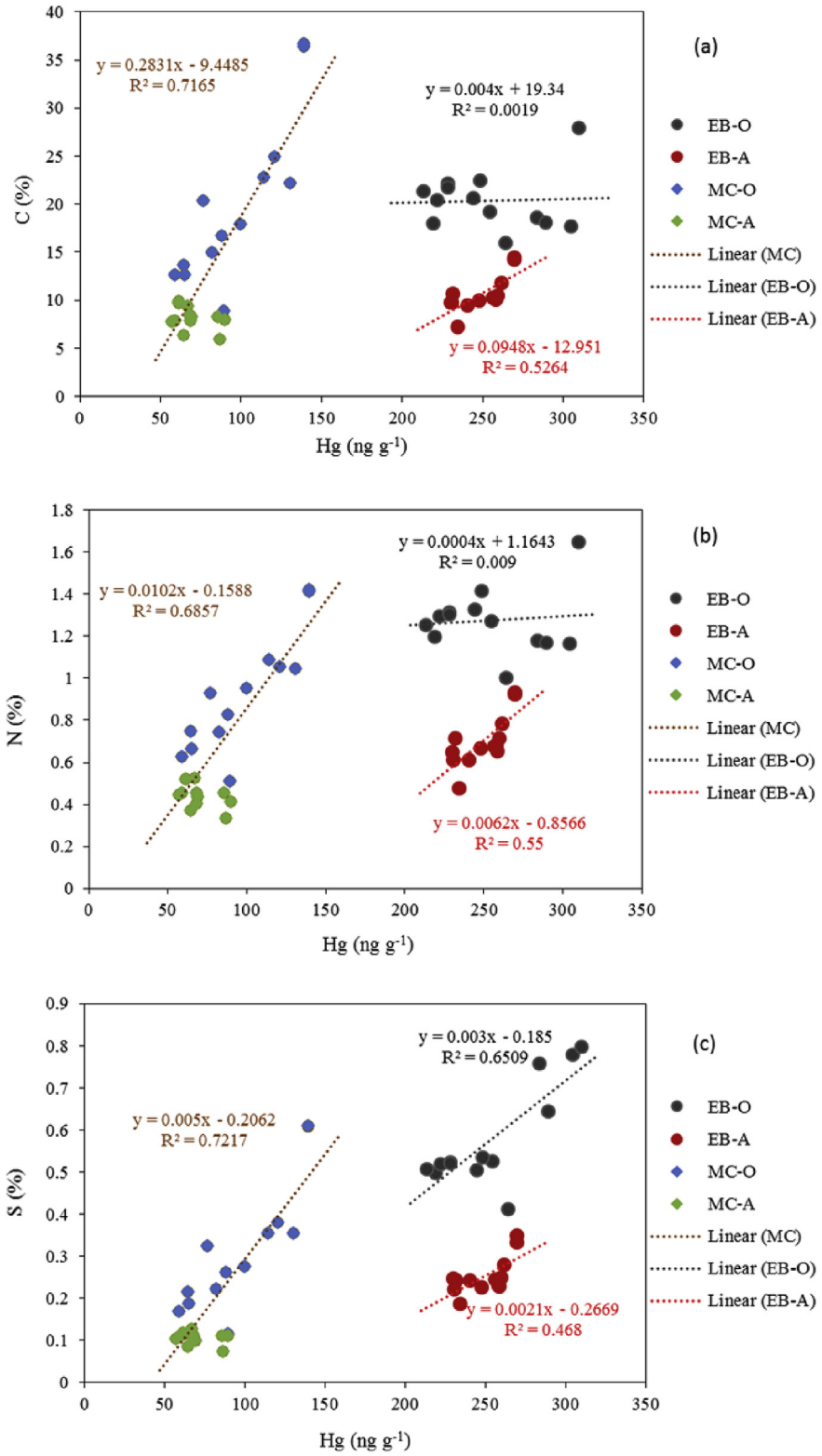

Fig. 5. Soil Hg concentration in relation to C (a), N (b), and S (c) concentrations in the mossy coppice (MC) and evergreen broadleaf (EB) forest soil profiles (O-horizons and A-horizons). All $P < 0.05$ except for the relationships between Hg and C and between Hg and N in EB-O.

external Hg (Hg uptake from the environment during litter decomposition) bound by soil organic matter during long-term atmospheric Hg exposure (Demers et al., 2007; Navratil et al., 2014; Obrist et al., 2011).

The mean Hg/S ratios of the EB forest soil were 44.7 and 100.5 mg kg^{-1} in the O-horizon and A-horizon, respectively (Fig. 4d), and in the MC forest, they were 34.8 and 67.9 mg kg^{-1} in the O-horizon and A-horizon, respectively (Fig. 4d). The Hg/S ratios

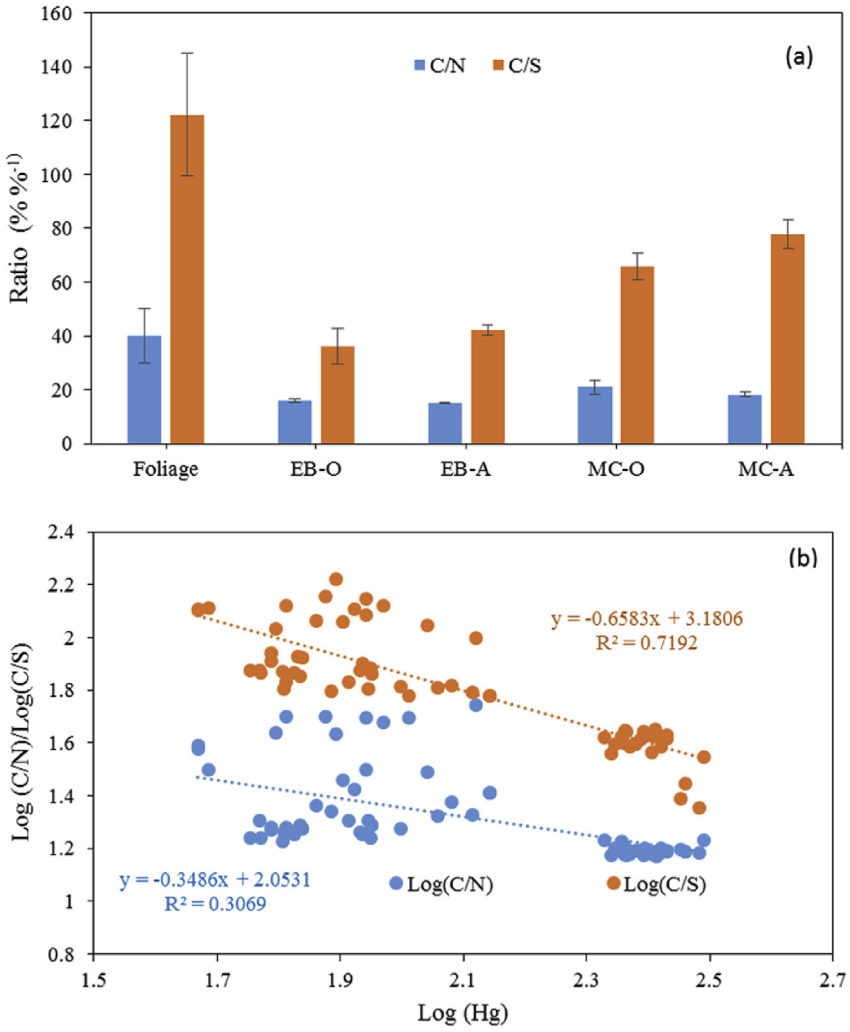

Fig. 6. C/N and C/S ratios in individual samples (a). Values of Hg as a function of the C/N and C/S ratios in all samples (b). EB, evergreen broadleaf forest; MC, mossy coppice forest; O, O-horizon; A, A-horizon. All $P < 0.05$, suggesting significant linear relationships at 95% confidence level.

in these two forests were lower than those in forests from North America and Europe. Demers et al. (2013) reported a Hg/S ratio of 210 mg kg^{-1} for upland forest humus soil in North America. Navratil et al. (2014) found Hg/S ratios ranging from 83 to 250 mg kg^{-1} for humus and mineral soil in European forests. Nasr and Arp (2011) found Hg/S ratios ranging from 108 to 220 mg kg^{-1} in humus and mineral soil. The relatively low Hg/S ratios in the subtropical forests in the present study may be linked to high S deposition (Fu et al., 2011). Additionally, Hg concentration was associated with the S concentration in all soil profiles of the two forests (Fig. 5c). Further, the Hg concentration was negatively related to C/S ratio in all samples across seasons and years. S mineralization in soil was slower than that of C (Houle et al., 2001; Ribeiro et al., 2002). Hg/C ratios increased with decreasing C/S ratios, suggesting that the dynamics in soil Hg concentration is closely linked to S concentration (Fig. 6b). Faster mineralization of C may lead to diminished production of reduced organic S functional groups, which have high affinity binding sites for Hg, thereby resulting in soil Hg accumulation (Zhang and Lindberg, 1999).

Compared to temperate forests, the timescale of C and other nutrient cycles in subtropical forest is typically shorter (Vitousek, 1984). C, N, and S in the surface soil are primarily originated from the decomposition of the litter. Field data have shown that total Hg mass in litter can increase by 37–147% after 1–2 years of litter decomposition in temperate forests (Demers et al., 2007; Hall and Louis, 2004; Pokharel and Obrist, 2011). To date, however, the fate of Hg during litter decomposition in subtropical forests has not been documented. Due to the uniqueness of subtropical forests in terms of soil microbial diversity, water circulation, and carbon cycle (Tan et al., 2012), the processes of Hg accumulation and sequestration in subtropical forests may be different from temperate and tropical forests. Further studies focusing on these issues will greatly improve the understanding of Hg accumulation in subtropical forests, and the contribution of the subtropics to the global Hg cycle.

3.3. Implications

We reported an enhanced Hg accumulation in forest floor soil in subtropical China, which has become the largest Hg emitter in the world because of rapid economic development during the past few decades. To date, atmospheric Hg inputs into forests in China were 1–60 times higher than those in North American and European forests (Fu et al., 2010; Wang et al., 2009; Zhou et al., 2013). Our results suggest that subtropical forests in South China are accumulating a high amount of Hg because of their high litter production and the interaction between Hg and S in the surface soil. However, these forests are facing threats from human activities, and their areas are decreasing. If deforestation occurs, soil Hg

evasion could increase by an order of magnitude, thereby accelerating soil Hg cycles and enhancing natural Hg emissions (Carpi and Lindberg, 1998; Gustin et al., 1999, 2000). Additionally, the change in land use from forests to croplands would deplete soil C, leading to runoff of Hg bound to organic matter in rainwater. The intensive Hg runoff could provide abundant Hg sources for the methylation of inorganic Hg, thereby further increasing ecological risks. Overall, we emphasize the importance of subtropical forests in South China in absorption and accumulation of Hg. Further studies are needed to assess the negative impacts of human disturbances and land use change on regional and global Hg cycles.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.jenvman.2016.08.073.

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