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Distribution of Mercury in Foliage, Litter and Soil Profiles in Forests of the Qinling Mountains, China

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Abstract: Forest ecosystems have been confirmed to be a sink of the global mercury (Hg) in the biogeochemical cycle. However, few studies have investigated the distribution of Hg in forest ecosystems on a regional scale in China. This work aimed to investigate the concentrations, distribution and influential factors of Hg in the Qinling Mountains forests in central China. Foliage, litter and soil profile samples were

collected at 24 sampling sites across the Qinling Mountains forests. The results of the present study showed that the concentrations of Hg in foliage, litter, organic soils and mineral soils were maintained at relatively low levels compared with those in subtropical forests of Southwest China. The average Hg concentrations followed the order litter (74 ± 34 ng·g^{-1}) > organic soil (71 ± 37 ng·g^{-1}) > mineral soil (34 ± 21 ng·g^{-1}) > foliage (31 ± 15 ng·g^{-1}). Mercury in foliage showed no obvious spatial pattern, likely due to differences in tree species and ages across the sampling sites. Higher concentrations of Hg in litter were observed on the southern slope (low altitude), while the distribution of Hg in organic soils was the opposite. Both the tree species and environmental parameters (altitude, temperature and precipitation) controlled the Hg concentrations in litter by regulating the decomposition rate of the litter. There were significantly positive correlations between the Hg concentrations and soil organic carbon, nitrogen and sulfur in all soil layers, indicating that organic matter has a high geochemical affinity for Hg in soils. Because of the lower turnover rate and the higher accumulation of organic matter in high altitude and low temperature areas, Hg loss from biogeochemical cycling processes was effectively reduced. The spatial distribution of Hg in forests soil can be shaped by the distribution of organic matter at the regional scale.

Keywords: Mercury, Qinling Mountains, Forest ecosystems, Soil profile, Organic matter

1. Introduction

Mercury (Hg) is a highly toxic heavy metal and a well-known global environmental contaminant (Tchounwou et al., 2003). Unlike other heavy metals, Hg can remain in the atmosphere for months or years, and the monsoon circulation causes Hg pollution on a global scale (Schroeder and Munthe, 1998). Atmospheric Hg is derived from both natural sources (e.g. volcanic activities and rock weathering) and anthropogenic sources (e.g., fossil fuel combustion, gold and mercury mining, nonferrous metal smelting and cement production) (Du et al., 2019; Pirrone et al., 2010). The global Hg model estimated that humans emit 2500 ± 500 Mg of Hg to the atmosphere per year, and approximately 3600 ± 3200 Mg of atmospheric Hg is deposited on terrestrial surfaces each year (Du et al., 2019; Outridge et al., 2018).

Forest ecosystems account for approximately 31% of the Earth's area and are one of the most important ecosystems in remote areas (Keenan et al., 2015). Based on mass balance, forest ecosystems are considered to be net sinks of the atmospheric cycle of Hg on a global scale (Graydon et al., 2012; Obrist, 2007; Selin, 2010). Forest leaves take up atmospheric Hg (gaseous elemental Hg) though stomata, or adsorb atmospheric Hg (particulate Hg and reactive gaseous Hg) on leaf surfaces (Gong et al., 2014; Graydon et al., 2009). Mercury in leaves is considered to originate mainly from atmospheric Hg, and leaves are usually used as a biomonitor for the study of the long-range atmospheric transport of Hg (Davidson et al., 2000; Gong et al., 2014). Mercury in leaves ultimately enters forest soils in the form of litter deposition (dry deposition). Studies have shown that litter deposition exceeds precipitation and throughfall (wet

deposition), and accounts for approximately 70% of the Hg input flux to forests in Southwest China (Fu et al., 2010; Zhou et al., 2013; Zhou et al., 2015). The deposited Hg is mainly adsorbed by soil organic matter and is preserved in forest soil (Gong et al., 2014; Obrist et al., 2009; Pokharel and Obrist, 2011). Moreover, more than 95% of Hg resides in the upper organic soil layers (Grigal, 2003; Juillerat et al., 2012). Of course, the Hg preserved in soil can rerelease into the atmosphere due to the volatile nature of Hg, or be leached into runoff and soil solution (Kerblom et al., 2008; Pokharel and Obrist, 2011). Although the people have paid attention to the importance of forest ecosystems to global Hg cycling, relevant basic data are still lacking.

The Qinling Mountains (QMs) are China's most important north-south geographical dividing line, and have a significant blocking effect on the movement of north–south airflow. In summer, humid ocean air can hardly penetrate northwestern China, and in winter, cold currents have difficulty migrating to southern China, due to the barrier formed by the OMs. As an ecological barrier, the QMs also play many functions, such as regulating climate, conserving water and soil and maintaining biodiversity. The blocking effect of the QMs on the atmosphere may increase the deposition and accumulation of atmospheric Hg in forest systems, which will pose a potential threat to environmental quality and biological health in those regions. However, information on the regional spatial distribution of Hg in forest ecosystems is quite limited in China, and most of the studies mainly focus on the Tibetan Plateau and subtropical forests of the Southwest China (Du et al., 2019; Fu et al., 2010; Gong et al., 2014; Lu et al., 2016; Wang et al., 2017; Zhou et al., 2016). To the best of our knowledge,

there are still few studies on the concentration levels and distribution of Hg in the OMs forest systems.

In this work, a comprehensive study on the concentrations of Hg in, litter and soil profiles across the QMs forests was carried out and the sampling sites spanned 700 km in longitude and 200 km in latitude. This study aims to (1) reveal the Hg concentration levels and spatial distribution in foliage, litter and soil profiles in the QMs forests and (2) understand the controlling factors and cycling of Hg in the QMs forests.

2. Materials and methods

2.1 Study area

The QMs span five provinces, cover a distance of approximately 200 kilometres from north to south and 700 kilometres from east to west, and are China's most important geographical dividing line between north and south.. The QMs are also a climate transition zone in China, where the warm temperate climate gradually tends towards the semihumid zone and typical subtropical climate from south to north and continuously tends towards the humid zone from east to west (Deng et al., 2019). The average annual precipitation of the QMs is approximately 825 mm, and approximately 70% of the precipitation is recorded during the monsoon period from May to September (Meng et al., 2021). The soil in the study area is typically mountain yellow brown earth. The vegetation is dominated by deciduous broad-leaved forests (such as *Quercus variabilis* Bl. and *Populus davidiana* Dode) on the southern slope, and are by coniferous and mixed broadleaf-coniferous forests (such as *Pinus massoniana* Lamb. and *Koelreuteria paniculata* Laxm.) on the northern slope of the OMs (Table 1S). There are

obvious differences in climatic factors between the northern and southern slopes of the QMs, with a dry and cold climate on the northern slope and a moist and warm climate on the southern slope.

2.2 Sample collection and processing

Foliage, litter and soil profiles were collected at 24 sites surrounding the QMs forests in August, 2020; and 12 of the sampling sites were located on the southern slope, and the remaining sites were located on the northern slope (Fig. 1). The altitude, average annual temperature and average annual precipitation of the sampling sites ranged from 290–1600 m, 6–15℃ and 640–990 mm, respectively (Table 1S). Each sampling site had three replicate plots, and each the plots were typically spaced more than 200 m apart. The sampling plots were chosen randomly and were more than 100 m away from main roads and buildings. In each sampling plot, undamaged fresh foliage was collected from the dominant tree species at heights of approximately 3–6 m above the ground using an averruncator, and the shrub foliage was collected by hand, and then mixed together. The corresponding surface litter (undecomposed and partially decomposed, approximately 1–3 cm) was collected by hand. Soil vertical profile samples were collected from top to bottom (each sample had a thickness of 5 cm) using stainless-steel hand-shovels after the surface litter was removed. The collection depth was 40 cm, and 8 soil samples were collected from each profile. The first 5 cm depth of the profile represented the organic horizon (strongly decomposed litter and humic substances), and the 5–40 cm depth of the profile represented the mineral horizon. The mercury concentrations in the mineral soils were the average concentrations of Hg in the 5–40

cm soil layers. Foliage and litter samples were stored in nylon mesh bags, and soils were stored in cloth bags. Coordinate and altitude information was recorded by GPS. In addition, litterbags were installed at the 6 sampling sites from August 2020 to July 2021 to collect freshly fallen leaves and to calculate the annual litterfall (Table 2S).

The soil samples were air-dried at room temperature (20–25℃), crushed with a wooden hammer and sieved through a 2 mm sieve (No. 10 mesh size). In the laboratory, the foliage and litter samples were washed in deionized water and then oven-dried at 40°C. Finally, the soils, foliage and litter were homogenized to 200 mesh size before chemical analysis.

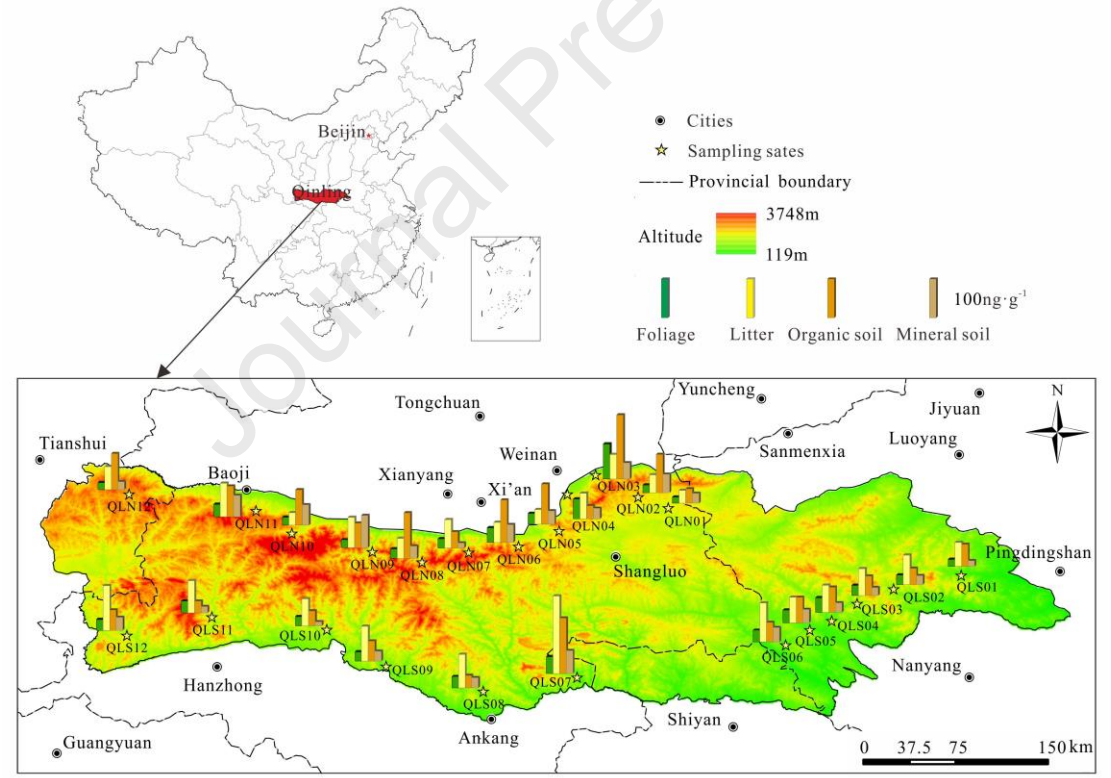


Fig. 1. Location of the study area, the distribution of the sampling sites and the concentrations of Hg in foliage, litter, organic soils and mineral soils.

2.3 Analytical methods and quality control

The chemical indexes of all samples were determined at the Chengdu Comprehensive Testing Center of Rocks and Ores, Sichuan Bureau of Geology and

Mineral Resources. A soil sample of approximately 0.1 g was extracted and digested using 5 mL aqua regia (3:1 HCl/HNO_3) at 100 °C for 1 h in a silicic borate beaker. After cooling, 5 mL of a mixed solution of thiourea and ascorbic acid (50 g/L) was used as the reducing agent. Foliage and litter samples were digested by a mixture of 10 mL HNO_3 and 2 mL H_2O_2 in a clean tank of the microwave digestion apparatus. After digestion, 0.5 mL of $HCLO_4$ was added to the solution, which was then diluted to 50 mL with deionized water. The total Hg concentration was measured by atomic fluorescence spectrometry (AFS; AFS-3000, Beijing Haiguang Instrument Co, China). The soil organic carbon (SOC) was measured using the volumetric method with potassium dichromate ($K_2Cr_2O_7$). Total sulfur (S) was measured using the volumetric method with iodine standard liquid. Total nitrogen (N) was measured using the distillation method.

The quality of the chemical analysis (the accuracy and precision of the data) was checked with blank samples, repetitive samples and standard reference materials (GSS-30, GSS31, GSS33 and GSS34 for soil samples and GSB-2 and GSB-3 for plant samples) during the analytical process. The detailed description can be seen in Supplementary file.

2.4 Statistical analysis

The mercury concentrations of the foliage, litter, organic soils and mineral soils were compared using one-way analyses of variance (ANOVAs) to determine whether the Hg concentrations differed between the southern and northern slopes. Pearson's correlation matrix was used to determine the relationship between two variables. The

data were analysed using SPSS 19.0 software.

3. Results and discussion

3.1 Hg concentrations in foliage, litter and soils

Prior to Hg determination in foliage, litter, and soils samples, method validation was made and the results is presented (Supplementary file). The results indicated that there is good agreement between the measured and certified values of the reference materials, the passing rate for accuracy was 100%, and the qualified rate of repeatability inspection exceeded 93%.

The average Hg concentrations in the different components generally showed a decreasing order of litter, organic soils, mineral soils and foliage (Fig. 1 and Table 1). The mercury concentrations in the foliage of the QM forests ranged from 16 $ng \cdot g^{-1}$ to 91 $ng \cdot g^{-1}$, with an average of 31 ± 15 $ng \cdot g^{-1}$. Compared to the Hg concentrations in the foliage, litter (ranging from 32 $ng \cdot g^{-1}$ to 200 $ng \cdot g^{-1}$, with an average of 74 ± 33 $ng \cdot g^{-1}$) was more enriched in Hg. Higher Hg concentrations were measured in the litter than in the fresh foliage, similar to previous studies (Huang et al., 2020; Obrist et al., 2011). On the one hand, during the process of decomposition after leaf fall, Hg in foliage is immobilized by organic matter and mainly resides in the residual fraction, and Hg is enriched with the reduction of foliage mass; therefore, the Hg concentration in the litter was higher than that in the fresh foliage (Huang et al., 2020; Pokharel and Obrist, 2011; Wang et al., 2016). On the other hand, litter has a longer exposure time than foliage and can continuously accumulate Hg in the atmosphere. The mercury concentrations in organic soils (ranging from 30 $ng \cdot g^{-1}$ to 166 $ng \cdot g^{-1}$, with an average of 71 ± 36 $ng \cdot g^{-1}$)

were similar to those measured in the litter, but the average concentration was 209% higher than the Hg concentrations in the mineral soils (ranging from 10 $ng \cdot g^{-1}$ to 83 $ng \cdot g^{-1}$, with an average of 34 ± 17 $ng \cdot g^{-1}$). Decreasing Hg concentrations from organic horizons to mineral horizons in forest soil have been widely reported (Obrist et al., 2009; Zhou et al., 2016). The enrichment of Hg in organic soils confirmed that the Hg mainly originated from litter deposition and was hardly affected by the geological background (Obrist, 2012; Obrist et al., 2011). SOC and Hg interact strongly with each other and can inhibit mobility and transformation.

Table 1 Comparison of Hg concentrations in foliage, litter, organic soils and organic soils with other forest regions

Site	Forest type	Foliage		Litter		Organic soil		Mineral soil		References
		Range	Mean ± SD	Range	Mean ± SD	Range	Mean ± SD	Range	Mean ± SD	
Qinling, China	Mixed broadleaf-conifer forest	16 – 91	31 ± 15	32 – 200	74 ± 33	30 – 166	71 ± 36	10 – 83	34 ± 17	This study
Tieshanping, Southwest, China	Mixed broadleaf-conifer forest	/	/	109 – 208	157 ± 24	54 – 462	191 ± 65	23 – 160	80 ± 31	(Zhou et al., 2015)
Ailaoshan, Southwest, China	Evergreen broadleaf forest			/	/	213 – 310	257 ± 14	230 - 269	248 ± 15	(Lu et al., 2016)
Gongga, Southwest, China	Mixed broadleaf-conifer forest	/	28 ± 4	/	/	120 – 260	/	/	/	(Fu et al., 2010)
Eastern Tibetan Plateau, China	Coniferous forest	/	/	15 – 81	35 ± 16	/	70 ± 37	/	/	(Wang et al., 2017)
Himalayas and Tibetan Plateau	/	/	/	/	/	24 – 255	63 ± 47	/	/	(Huang et al., 2019)
14 forests of the USA	Mixed broadleaf-conifer forest	8 – 48	/	22 – 83	/	46 – 420	/	/	60	(Obrist et al., 2011)
Junsele, Northern, Sweden	Coniferous forest and dwarf shrubs	/	/	79 – 163	/	188 – 313	/	17 - 31	/	(Jiskra et al., 2015)
Tapajos National Forest, Brazil	/	/	/	/	/	190 – 293	245	/	/	(Figueiredo et al., 2018)
Northeast China	Mixed broadleaf-conifer forest	/	/	39 – 73	51 ± 11	49 – 101	68 ± 20	19 - 67	60	(Luo et al., 2014)

The results from the QM forests were compared with those from other forest regions of the world (Table 1). The mercury concentrations in the litter and organic soils were obviously lower than those measured in Tieshanping (average 157 ± 24 $ng \cdot g^{-1}$ for litter, average 191 ± 65 $ng \cdot g^{-1}$ for organic soils), Aliaoshan (average 257 ± 14 $ng \cdot g^{-1}$ for organic soils) and Gongga (range 120–260 $ng \cdot g^{-1}$ for organic soils) forests in

southwestern China (Fu et al., 2010; Lu et al., 2016; Zhou et al., 2015). These results may be attributed to the differences in atmospheric Hg concentrations, as high atmospheric Hg concentrations result in an increase in dry and wet depositions of Hg. Studies have shown that nonferrous metal smelters have a significant impact in Southwest China, which contribute to 7.5% and 7.1% of wet deposition and dry deposition of Hg, respectively (Wang et al., 2014). In addition, the difference in litter biomass production can significantly affect the input of atmospheric Hg to the forest floor (Wang et al., 2017). The mercury concentrations in the organic soils in this study were lower than those measured in America (range 46–420 $ng \cdot g^{-1}$), Sweden (range 188–313 $ng \cdot g^{-1}$) and Brazil (range 190–293 $ng \cdot g^{-1}$) (Figueiredo et al., 2018; Jiskra et al., 2015; Obrist et al., 2011). However, the Hg concentrations in the organic soils were comparable to those measured on the Tibetan Plateau (average 63 ± 47 $ng \cdot g^{-1}$ and 70 ± 37 $ng \cdot g^{-1}$) and in northeastern China (average 68 ± 20 $ng \cdot g^{-1}$) (Huang et al., 2019; Luo et al., 2014; Wang et al., 2017), and the Hg concentrations in the litter were higher than those measured on the Tibetan Plateau in China (average 35 ± 16 $ng \cdot g^{-1}$), China (Wang et al., 2017). Our results showed that the Hg levels in the litter and organic soils of the QM forests were relatively low, which may reflect the relatively low atmospheric Hg concentration and Hg deposition flux in this area.

3.2 Spatial distribution of Hg in foliage

Spatial distribution studies can provide important information regarding the role of the regional contributions and environmental effects on the observed Hg loads in remote regions (Gong et al., 2014). The absorption of atmospheric Hg by stomata is

considered to be the predominant pathway of Hg accumulation in fresh foliage (Laacouri et al., 2013). Foliage is usually used as a biomonitor of pollutants, which has provided evidence for the long-rang atmospheric transport of pollutants from sources to remote forest regions (Davidson et al., 2000; Gong et al., 2014). For fresh foliage, we found that the sampling sites S03 (91 $ng \cdot g^{-1}$), S04 (51 $ng \cdot g^{-1}$) and S06 (38 $ng \cdot g^{-1}$) near southeastern Xi'an, a large industrial city in China, had relatively higher Hg concentrations (Fig. 1 and Table 1S). Xu et al. (2017) reported that the total gaseous Hg concentration in Xi'an was 5.66 $\pm$ 2.73 $ng \cdot m^{-3}$, which was at a moderate level compared with other cities in China. The foliage in the QM forests may have been polluted by atmospheric Hg from Xi'an under the influence of the northwestern monsoon. There were no obvious spatial distribution patterns of foliar Hg concentrations in the study area of the QM forests (Fig. 2) and no significant correlations with altitude, temperature and precipitation. Previous studies have shown that the leaf stomata and leaf lifespan are important factors that influence Hg accumulation in foliage (Blackwell and Driscoll, 2015; Laacouri et al., 2013; Wang et al., 2016). A higher stomatal conductance leads to higher air-foliage Hg^0 exchange, and a longer lifespan of foliage results in a higher accumulation of atmospheric Hg (Graydon et al., 2006; Wang et al., 2016). The variation in the tree species and age of each sample may confound the potential spatial distribution information in the QM forests.

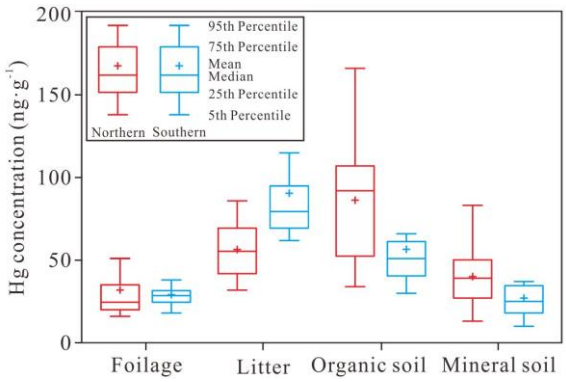


Fig. 2. The concentrations of Hg in foliage, litter, organic soils and mineral soils on the northern and southern poles.

3.3 Spatial distribution of Hg in litter and soils

As shown in Fig. 1, the highest Hg concentrations in the litter (200 ng·g^{-1}) and organic soils (143 ng·g^{-1}) were measured at sampling site S07, which demonstrated the input of litter Hg to organic soil. However, there was no synergistic overall variation trend between the Hg concentrations in the litter and organic soils. Because the southern and northern slopes of the QMs have significantly different forest types and climate characteristics, we attempted to analyse the difference in the Hg concentrations in the foliage, litter, and organic and mineral soils between these slopes. The results showed that the Hg concentrations in the litter ($p < 0.05$, according to one-way ANOVA) and organic soils ($p < 0.05$, according to one-way ANOVA) differed significantly between the southern and northern slopes of the QMs. Interestingly, the Hg concentrations in the litter on the southern slope (average 91 ± 36 ng·g^{-1}) were higher than those on the northern slope (average 57 ± 17 ng·g^{-1}), while the Hg concentrations in the organic soils (average 57 ± 28 ng·g^{-1} for the southern slope and 86 ± 37 ng·g^{-1} for the northern slope) and mineral soils were the opposite (average 27 ± 15 ng·g^{-1} for the southern slope and 40 ± 24 ng·g^{-1} for the northern slope) (Fig. 2). Previous studies have shown that Hg concentrations in organic soils mainly originate from litter Hg deposition, which causes

elevated concentrations of Hg in forest soils (Obrist, 2012; Obrist et al., 2011; Zhou et al., 2018; Zhou et al., 2015). However, the distribution patterns of Hg in the organic soils and litter in this study were inconsistent. Of course, simple Hg concentrations in litter cannot reflect the litter deposition flux of Hg. Subtropical broad-leaved forests generally have greater annual litterfall flux than temperate coniferous forests. The annual deposition fluxes of Hg via litterfall on the southern slope (48.3 $\mu g \cdot m^{-2} \cdot year^{-1}$) were higher than those on the northern slope (17.7 $\mu g \cdot m^{-2} \cdot year^{-1}$) of the QM forests (Table 2S). Furthermore, Zhou et al. (2018) showed that the annual deposition fluxes of Hg via litterfall in coniferous forests (40.5 $\mu g \cdot m^{-2} \cdot year^{-1}$) were lower than those in broad-leaved forests (90.9 $\mu g \cdot m^{-2} \cdot year^{-1}$) in Southwest China. Therefore, the distribution of Hg in organic soils in forests may not simply be driven by litterfall deposition flux, but also by the accumulation and biogeochemical cycling of Hg.

3.4 Factors influencing the Hg concentrations in litter

Differences in tree species on the northern and southern slopes of the QM forests may be a factor underlying the distribution pattern of Hg in litter. As described in the previous section, the sampling sites on the southern slope are all deciduous broad-leaved forests, while the northern slope is dominated by broadleaf-coniferous mixed forests. The mass of litter gradually decreases during the process of decomposition, but the mass of Hg had no obvious variation or slightly increased, which led to an increase in the Hg concentrations in litter (Demers et al., 2007; Heyes et al., 1998; Pokharel and Obrist, 2011; Zhou et al., 2018). However, the litter of the broad-leaved forest had a higher decomposition rate than that of the coniferous forest. For example, Zhou et al.

(2018) found that the half-life of litter was 0.96 years for broad-leaved forests and 1.31 years for coniferous forests. Over time, the decomposition rate of litter in the broad-leaved forest exceeded that in the coniferous forest, resulting in relatively higher Hg concentrations in the litter on the southern slope. Demers et al. (2007) showed that Hg concentrations in litter increased by 127% and 109% in a deciduous forest and coniferous forest, respectively, after two years of decomposition. Hall and St Louis, (2004) reported that Hg concentrations in litter increased by 476% and 149% in a deciduous forest and coniferous forest, respectively, during an 800-day decomposition period.

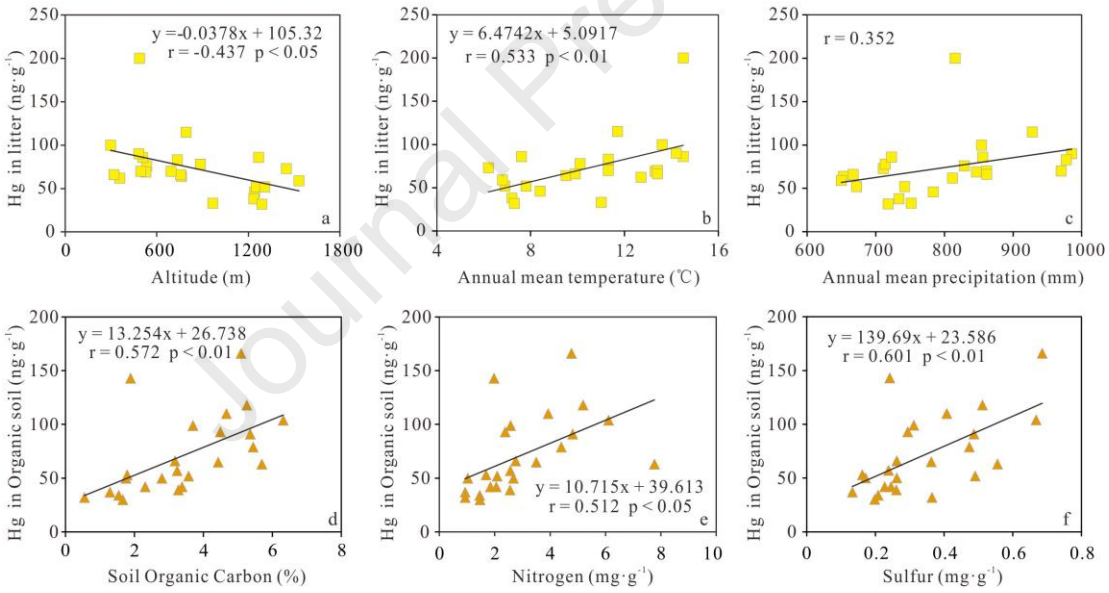


Fig. 3. The relationship between Hg concentrations in litter with altitude (a), annual mean temperature (b) and annual mean precipitation (c). The relationship between Hg concentrations in organic soils between soil organic carbon (d), nitrogen (e) and sulfur (f) concentrations in organic soils.

Linear regression analysis based on Pearson's correlation coefficient indicated that the Hg concentrations in the litter were dependent on the altitude, temperature and precipitation (Fig. 3). The mercury concentrations in the litter increased with increasing temperature ($r = 0.533$, $p < 0.01$) and precipitation ($r = 0.352$) and decreasing altitude

308 ($r = -0.437$, $p < 0.05$) (Fig. 3). An increase in temperature may result in an increase in
309 microbial activity and a higher decomposition rate of litter (Gorecki et al., 2021; Sierra
310 et al., 2015). These studies show that environmental parameters can affect Hg
311 concentrations in litter by controlling the decomposition rate of the litter. High
312 temperature and precipitation accelerated the decomposition rate of the litter, leading
313 to higher Hg concentrations in the litter on the southern slope. The difference in tree
314 species and environmental parameters (such as temperature and precipitation), which
315 affected the litter decomposition rate, were the main reasons for the higher Hg
316 concentrations in the litter on the southern slope than on the northern slope.

317 **3.5 Factors influencing the Hg concentrations in soils**

318 Soil organic matter is considered to be an important factor controlling the
319 distribution of Hg in terrestrial ecosystems, and C—, N— and reducible S— in organic
320 molecules have a strong geochemical affinity for divalent Hg (Obrist et al., 2011). The
321 correlation between Hg and SOC, N and S in organic soils in the QM forests were
322 analysed, and the results showed that there were significant positive correlations
323 between Hg and SOC ($r = 0.572$, $p < 0.01$), N ($r = 0.512$, $p < 0.05$) and S ($r = 0.601$, p
324 < 0.01) (Fig. 3). Soil organic carbon, N and S are the main constituents of organic matter
325 in soil. Organic matter is a key variable that affects the spatial distribution of Hg in
326 terrestrial ecosystems. Based on the relationship between Hg and SOC, N and S in
327 organic soils, the distribution pattern of Hg can be explained by analysing the
328 distribution of SOC, N and S.

329 The concentrations of SOC, N and S on the northern slope were 1.5-2 times higher

than those on the southern slope, which indicated that there was greater organic matter accumulation on the northern slope. This is consistent with our knowledge of the storage of organic matter. As the altitude increases and temperature decreases, the turnover rate of C decreases, which increases the C residence time (Blackwell and Driscoll, 2015; Obrist et al., 2011). The concentrations of SOC in organic soils were positively correlated with altitude ($r = 0.534$, $p < 0.01$) and were negatively correlated with temperature ($r = -0.628$, $p < 0.01$) and precipitation ($r = -0.485$, $p < 0.05$), confirming the above conclusion (Fig. 1S). Higher runoff and evasion flux of Hg were measured in low-altitude areas than in high-altitude areas (Zhou et al., 2015). We believe that the higher organic matter accumulation on the northern slope effectively reduced the Hg loss from runoff and evasion in the biogeochemical cycling processes.

This view seems to be a good explanation for the distribution pattern of Hg in the soils of the QM forests. However, runoff and the evasion flux of Hg only account for a small part of annual atmospheric Hg deposition (Harris et al., 2007; Hintelmann et al., 2002). Because of the large number of adsorption sites in the soils (Obrist et al., 2011), even low-organic soils can retain the main portion of atmospheric Hg deposition. he continuous accumulation of Hg may be more important for the accumulation of Hg in soils (more details are discussed in the following sections).

3.6 Distribution of Hg in vertical soil profiles

The highest Hg concentrations were measured in the upper soil layer (organic soils, 0–5 cm), and Hg concentrations decreased with increasing depth (Fig. 4). The concentrations of SOC, N and S showed vertical distribution characteristics similar to

those of Hg, and the highest concentrations of SOC, N and S were also found in the organic soil layers (Fig. 4). These observations are generally consistent with the results reported in previous studies (Du et al., 2019; Gong et al., 2014; Obrist et al., 2011). Because of the high geochemical affinity of organic matter for Hg, soil organic matter has been proven to be one of the most important factors influencing the distribution of Hg (Gong et al., 2014; Obrist et al., 2011; Wang et al., 2016; Zhou et al., 2015). The correlation between Hg and SOC and N and S in each soil layer were analysed, and the results suggested strong relationships between SOC, N and S and the Hg concentrations across all soil layers (Table 3S). In addition, the average concentrations of Hg in each layer of the vertical soil profiles on the northern slope were higher than those on the southern slope, which may be attributed to the migration of Hg in the soil (Fig. 4).

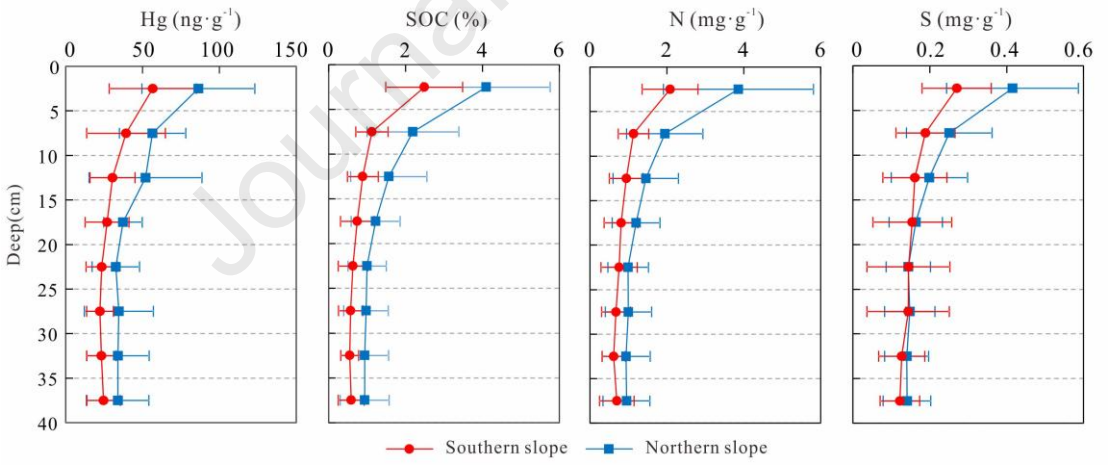
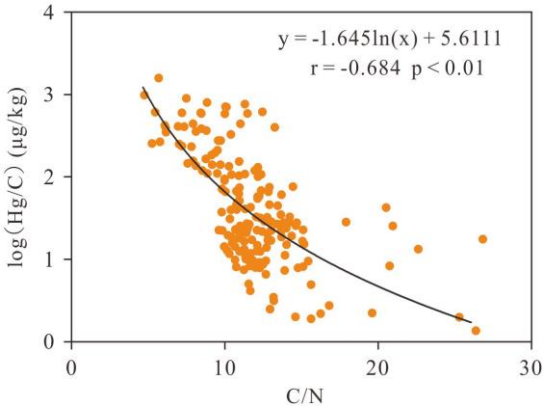


Fig. 4. Vertical distribution of mercury (Hg), soil organic carbon (SOC), nitrogen (N) and sulfur (S) in soil profiles

The soil Hg/C ratios were 0.24–7.94 ng/gC, and the average values for the northern and southern slopes were 2.95 ng/gC and 3.53 ng/gC, respectively. The C/N ratios are indicative of the degree of decomposition, and lower C/N ratios generally represent older, decomposed fractions (Gong et al., 2014; Obrist et al., 2009). The C/N ratios in

soils ranged between 4.7 and 26.8, with an average of 11.8. We analysed the relationship between the Hg/C ratios and C/N ratios in the soil samples (Fig. 5). The results showed a significantly negative correlation between Hg/C and C/N ($r = -0.684$, $p < 0.01$), which suggested that older, highly decomposed soils have high levels of Hg. In other words, during mineralization, the Hg/C ratio increases with C loss. The continuous accumulation of atmospheric Hg in soils under the adsorption of organic matter has led to the distribution pattern on a regional scale.

This study emphasized the influence of organic matter accumulation and Hg retention on the soil Hg concentration at the regional scale. However, higher concentrations of Hg have been measured in the subtropical southeastern and southwestern areas in China with a moist and warm climate compared with northeastern China with a cold and dry climate (Wang et al., 2021). This seems to contradict the results of our study. However, the southeastern coastal region of China is the most highly developed area in China, and southwestern China has a long history of nonferrous metal smelting. High atmospheric Hg concentrations lead to an increase in Hg deposition (wet/dry deposition), which also has a strong influence on the distribution of Hg in soil on a regional scale.



388 **Fig. 5**. The relationship between Hg/C ratio and C/N ration in soil

389 ## 4. Conclusions

390 This study investigated the Hg concentrations and distribution in the foliage, litter
391 and soil profiles at 24 sampling sites across the QMs forests. These QMs forests had
392 relatively lower Hg concentration levels than southwestern Chinese and American and
393 European forests, which reflects the lower atmospheric Hg concentration and Hg
394 deposition in this area. Environmental parameters can affect Hg concentrations in litter
395 by controlling the litter decomposition rate. High temperature and precipitation
396 accelerated the decomposition rate of the litter, leading to higher Hg in the litter on the
397 southern slope at low altitudes. The highest Hg concentrations were measured in the
398 organic soils, and litter deposition is an important source of Hg to organic soils.
399 However, the distribution of Hg in soils was not completely controlled by the litter
400 deposition flux. A lower C turnover rate and higher organic matter accumulation in
401 high-altitude areas effectively reduced the Hg loss from biogeochemical cycling
402 processes. Moreover, continuous accumulation of atmospheric Hg in soil under the
403 adsorption of organic matter has led to the distribution pattern on a regional scale.

404

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Declaration of Interest Statement

We declare that we have no competing financial interests or personal relationships that could have appeared to influence.