Mercury distribution in surface soil of China is potentially driven by precipitation, vegetation cover and organic matter

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Zhao-Yang Zhang
Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences

Gang Li
Institute of Urban Environment, Chinese Academy of Sciences

Lei Yang
Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences

Guo-Xin Sun
Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences

Corresponding Author
gxsun@rcees.ac.cn
ORCiD: https://orcid.org/0000-0002-9397-3164

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Abstract

Background
Mercury (Hg) distribution in surface soil in China is quite uneven with relatively high concentrations in southeastern China and low concentrations in northwestern China. The reason for this is inconclusive so far, especially on the continental scale. In the present study we used the multiple linear regression model to evaluate the relative importance of these different factors and elucidate the contribution on soil Hg of major factors, such as dry and wet precipitations, vegetation cover, soil organic matter and solar radiation.

Results
Wet and dry deposition associated with precipitation and vegetation cover, and emissions influenced by soil organic matter (SOM), are key factors controlling Hg distribution in surface soil. In southeast China, high wet deposition associated with south Asia monsoon and dry deposition, enhanced by vegetation canopies, together with low emission caused by high vegetated surface and solar radiation, are responsible for high Hg in soil (> 0.08 mg/kg). In northeast China, medium wet Hg deposition and high dry deposition via throughfall and litterfall, low emission due to weak solar radiation and high SOM are responsible for high Hg accumulation in soil. In northwest China, low wet deposition, together with high emission by low vegetation cover (bare soil), SOM and strong solar radiation contributed to low Hg in surface soil (< 0.03 mg/kg).

Conclusions
We suggest that wet deposition derived from Asian monsoon, dry deposition linked to vegetated surfaces and Hg emission associated with vegetation cover, SOM and solar radiation play key roles in Hg balance in other terrestrial environments worldwide, especially in those regions with significantly high wet and dry deposition and high vegetation cover.

1. Background
Mercury (Hg) is an extremely toxic element to humans [1, 2] and ecological receptors [3]. Some atmospheric Hg species (e.g. elemental Hg) produced by anthropogenic activities are relatively stable and can be subjected to long-distance transport to remote regions, and Hg in the air ultimately found their way (wet and dry deposition) into terrestrial or aquatic systems [4]. It was reported that soil Hg generally account for > 90% of Hg stored in terrestrial ecosystems and most Hg in soil (> 80%) is
from atmospheric inputs [5].

At large geographical scales, the spatial distribution of Hg and related main affecting factors in the terrestrial environment is poorly understood [6], although soils and terrestrial sediments have been indicated to act as net Hg sinks on century timescales [7]. Most investigations focus on the transformation of Hg species, biogeochemical cycling of Hg among the atmosphere, terrestrial system and/or ocean [8], or Hg contamination in soil at regional scale [9]. It has been reported that the flux of Hg deposition and emission from soil were controlled by various environmental factors, such as solar radiation, temperature, Hg speciation and content, soil porosity, soil moisture, pH, soil organic matter content, ambient Hg concentration, vegetation cover, and precipitation [10–15]. It is generally accepted that at the small scale (local or regional scale), mining activities or combustion of Hg-bearing coal are among the most important factors affecting Hg distribution in soils [9]. However, at the continental and sub-continental scale such as in China, it remains largely elusive as to why Hg distribution in surface soil is so uneven (Fig. 1). It is still unclear at present whether the spatial variability of soil Hg distribution at such a large scale is affected by other factors besides lithology.

China is one of the world’s largest sources of Hg to air [17–19]. The Hg distribution in surface soil was mainly from Hg deposition of atmosphere [16]. The first soil Hg map in China was obtained in National Soil Background Survey, which was initiated in late 1980s and issued in 1994 (Fig. 1A) [20]. Considering the possible influence of rapid industrialization and dramatic urbanization occurred in China in past decades on the elements distribution in surface soil, the soil Hg map (Fig. 1B) was updated by new geochemical mapping project named the National Multi-Purpose Regional Geochemical Survey (NMPRGS) of China, which was initiated in 1999 and completed in 2014 [2, 20]. Comparing the old and new soil Hg maps on the national scale, the spatial distribution of Hg in soil were consistent with each other, regardless of the difference of Hg concentration in the same region, although local scale contamination of soil Hg via gold production or fossil fuels combustion were observed because of unprecedented economic growth in China during the past two decades [21]. Hg distribution in surface soil in China was uneven before the Chinese economic boom (Fig. 1A), with relatively high concentration in southeast and northeast, and much lower concentration in northwest.
China (Fig. 1). The primary driving factors for Hg distribution in surface soil at large scale are elusive.

2. Materials And Methods

The map of soil mercury distribution in China (Fig. 1A) was printed on the atlas titled “The atlas of soil environmental background value in the People’s Republic of China” 1994, page 10-11, China Environmental Science Press; and digitized with ArcGIS 10.2 (Environmental Systems Research Institute Inc, Redlands, California, USA). The 2014 map (Fig. 1B) was cited from published literature [2], and the dataset for this map was from a new survey of geochemical mapping project [20]. Precipitation data was provided by China Meteorological Data Sharing Service System. The precipitation distribution map was modified from our previous study [22] using ArcGIS 10.2. The normalized difference vegetation index (NDVI) data and the map of soil organic matter (SOM) were from National Earth System Science Data Center, National Science & Technology Infrastructure of China (http://www.geodata.cn). The solar radiation distribution map was downloaded from SOLARGIS (https://solargis.com/maps-and-gis-data/download/China).

We converted the soil mercury distribution, mean annual precipitation, NDVI, and soil organic matter content into raster format and then randomly created 3000 points at the national scale in ArcGIS10.2. All corresponding values of 3000 points in different datasets were extracted by spatial analysis tools in ArcGIS. The error points would be filtered out by the “dplyr” package in the statistical software program “R” (R-Core-Team) and a total of 2827 points were obtained. We used the subsampling data to eliminate biases caused by uneven data density for different intervals of subsets (sampling bin). The precipitation dataset was separated into 19 subsets (sampling bin) with every 100 mm/year interval in R, and the last subset contains all points with precipitation amount higher than 1800 mm/year (Fig. 2C). In order to obtain the relationship (Fig. 2B) between the Hg concentration in surface soil and precipitation, one hundred sample points were randomly selected from each subset of precipitation and the average value of those points was taken as the representative of this subset.

The NDVI dataset was divided into 9 subsets based on NDVI value from 0 to 0.9 at intervals of 0.1; 200 sample points was randomly obtained from each sampling bin and the average value of these 200 sample points stands for this subset (Fig. 2E&2F). For the dataset of soil organic matter (SOM), it
was split into 10 subsets at interval of 1% from 0, and the last subset contained all values > 10%, the average of the 200 randomly sampling points represents this subset (Fig. 2H&2l). The data of solar radiation were processed in the same way as other factors. It was split into 8 subsets, the first subset includes those points with the value of solar radiation less than 1000 kWh m$^{-2}$, and the last subset includes all datas higher than 1800 kWh m$^{-2}$.

3. Results And Discussion
3.1 Soil mercury distribution in China
Mercury is a naturally occurring element, with an average crustal abundance of ~ 0.05 mg/kg [23]. Volcanoes and rock weathering can introduce Hg into the soil with significant local variations. In China, the average natural abundance of Hg in the Earth's crust is estimated to be 0.065 mg/kg [24], slightly higher than that in other parts of the world (~ 0.05 mg/kg). However, the Hg concentrations in surface soils are in the range of 0.08–0.2 mg/kg in southeast China, much higher than the national background value (0.065 mg/kg) (Fig. 1A). After economic soar in the past several decades, soil Hg content is substantially raised (Fig. 1B). In some regions such as Yangtze River Delta and Pearl River Delta located in southeast China, the average soil Hg concentrations were up to 0.59 and 0.50 mg/kg, respectively [25]. In some individual regions Hg concentration is even higher than 1 mg/kg, and this probably due to the local anthropogenic Hg emission and deposition during rapid industrial development in the past several decades [26]. The amount of Hg partitioning to runoff is considered to be a small fraction (200 Mg/year) of the amount of Hg stored in soil ($1.15 \times 10^6$ Mg/year) [27]. This means that the spatial variability of soil Hg distribution is affected by other factors besides lithology, which plays an important role in elevating Hg concentrations in surface soil.

3.2 Mercury input from wet precipitation
One source of Hg in soil beyond parent materials could be the atmospheric deposition, including dry and wet deposition [28, 29]. Significant linear relationship ($R^2 = 0.91$) between soil Hg level and precipitation were observed (Fig. 2B). This trend is also supported by European results (Figure S4A).

Soil Hg in Europe is markedly linearly increased ($R^2 = 0.92$) accompanied by precipitation (< 800 mm/year). However, higher precipitation (> 800 mm/year) did not enhance soil Hg accumulation,
and soil Hg accumulation keeps constant at high levels (if last point is omitted due to geological high Hg background) (Figure S4A). Obviously, high wet deposition was one of main contributors to high surface soil Hg. These results suggested that precipitation derived from East Asian monsoon is one of the important factors controlling soil Hg distribution in China. In central China, surface soil Hg concentration is relatively lower than that in southeast China because the wet deposition in this region is relatively low (400–800 mm). In northwest China, wet deposition is much less (annual precipitation less than 200 mm), in some regions annual precipitation even less than 100 mm, such as the sand and Gobi desert as no influence from the east Asian summer monsoon. The low Hg contents in surface soils (< 0.03 mg/kg) were observed (Fig. 1), and these values were significantly lower than the background soil Hg (0.065 mg/kg). This indicates that significant Hg loss occurred in this region, but the reason is still elusive.

Natural and anthropogenic sources contributed approximately equally to the total atmospheric Hg [30]. East China as an important source region of atmospheric Hg in China [31, 32], constituting approximately 20% of the total emissions [33]. The highest annual surface air total gaseous Hg of > 3 ng/m$^3$ as simulated in a model [34] and the highest concentration of total gaseous Hg being 3.3 ng/m$^3$ were observed in this region [18, 33]. The high mean Hg concentration in southeast China such as the Yangtze River Delta region was mainly caused by occasional long-distance transport from domestic source regions and regional anthropogenic emissions due to the rapid economic development which relied heavily on coal combustion for electricity generation [8, 33]. Relatively high level of particulate bound Hg (0.13–0.18 ng/m$^3$) is readily removed through wet deposition [33]. Eastern Chinese soil will be largely influenced by anthropogenic Hg emissions from surrounding areas due to its close proximity to the largest anthropogenic Hg emissions. Elevated Hg pollution in the southeast atmosphere can be expected to have higher deposition in the local soil through precipitation. Most Hg deposition occurred during the monsoon season with 83% of oxidized and particulate bound Hg [35]. Higher Hg concentrations were observed in sediment samples collected in the Indian estuarine regions during post-monsoon than pre-monsoon of Indian summer monsoon [36],
confirming wet deposition derived from monsoon is one of major Hg contributors.

The Asian monsoon system controls wet deposition over mainland Asia including China. During summer monsoons, a strong land-sea pressure gradient is produced by high insolation over continental regions, forcing oceanic winds to converge over the southeast China, bringing oceanic moisture and causing abundant rainfalls in this region [37]. The mean annual precipitation decreases gradually from 2,000 mm from the Southeast coast to 400–800 mm in central China, and then decreases to less than 200 mm in most of the northwest (Fig. 2A).

The Hg concentration is substantially linearly enhanced due to the increased precipitation in China ($R^2 = 0.91$) (Fig. 2B, 2C, Table 1), suggesting that the dominant transport routes of Hg are highly governed by the monsoons in southeast Asia [38]. Total wet deposition of Hg in the regions of southeast China was simulated by Global/Regional Atmospheric Heavy Metals models (GRAHM), and consistent with high amounts of precipitation [34]. Monsoon has increased total Hg levels in some regions of the Tibetan Plateau by carrying Hg enriched air from south Asia [39]. Another example is that high wet Hg deposition has been observed in some coastal regions of USA, such as Porto Rico [40] and Florida [41]. The distribution of total Hg wet deposition is similar to the total annual precipitation in USA (Figure S1), indicating that wet deposition is one of the major contributors to soil Hg content as well. It is widely accepted that a large pool of gaseous oxidized Hg existed in the upper free troposphere and lower stratosphere, which was oxidized from the global Hg pool [13, 40, 42].

Gaseous oxidized Hg is soluble in cloud droplets or readily scavenged by wet deposition [7], and is a persistent Hg source in precipitation. Thunderstorms, deep convective rainstorms, are particularly effective at scavenging gaseous oxidized Hg from boundary layer into the cloud base, and resulted in nearly 50% increase of Hg deposition in comparison with non-convective rainfall events [43]. The thunderstorm frequency is also highly contributed to Hg wet deposition. The average annual days of thunderstorms was high (> 80 days) in southern and southwestern regions of China [44], indicating more Hg wet deposition occurred in these regions, which is consistent with high soil Hg (Fig. 1). Wet deposition associated with east Asian summer monsoon has been considered as a major contributor to the distribution of trace elements such as volatile element selenium [22] and arsenic [45, 46].
Excessive Hg input to soil was occurred in the monsoon season than that in other seasons [35, 47]. Wet deposition typically accounts for 50–90% of the atmospheric Hg entering the environment in some regions [48, 49]. The east Asian summer monsoon generates wind patterns in summer that sweep atmospheric Hg derived from the South China Sea and transport through the atmosphere and deposit to terrestrial ecosystem of southeast China (Fig. 2A).

3.3 Mercury input from dry precipitation
Besides wet deposition, Hg in air also enters terrestrial ecosystems through dry deposition, such as gravity deposition, atmospheric diffusion and other processes [29, 50, 51]. The precipitation in central China, such as northeastern China is moderate (400–800 mm). However, soil Hg levels is relatively high (0.04–0.20 mg/kg, Fig. 1A). This confirming the dry deposition is also an important contributor to soil Hg accumulation. The significantly linear relationship ($R^2 = 0.94$) between soil Hg levels with NDVI at the national scale indicated the vegetation cover has significant effects on Hg accumulation in soil (Fig. 2D, 2E, 2F). Litterfall represents dry deposition of gaseous elemental Hg and throughfall reflects wash-off of dry deposited gaseous oxidized Hg and particulate Hg. Dry deposition of Hg, followed by throughfall and litterfall, is an important source of Hg to forest ecosystem because deposition of gaseous elemental Hg through the vegetation uptake occurred throughout the year [52]. It has been demonstrated that upland forest soil act as strong sinks for Hg and dry deposition of Hg through litterfall (71.2 µg/m$^2$/yr) was the major pathway for Hg loading to the forest catchment in southwestern China, which is much higher than corresponding wet deposition (5.2 µg/m$^2$/yr) [53, 54]. In temperate, forested regions like northeast China, dry Hg deposition is widely accepted to be about two times of wet Hg deposition [40, 55] and uptake of Hg by forest canopy played a predominant role in soil Hg accumulation [56]. The global dry Hg deposition via litterfall is 2–6 times higher than Hg emission from corresponding soil [15], although soil Hg emission could counteract part of soil Hg deposition. About 72–90% of total Hg deposition in forest areas of China was contributed by dry deposition [38]. Especially, the large deposition through litterfall suggests a strong accumulation of Hg in soil [54]. Vegetation is known to contribute to atmospheric Hg as a sink through direct uptake from the air.
through stomata on the leaf surface [11, 57] with Hg species of nanoparticulate or bis-thiolate complex [58]. It has been demonstrated that almost all of the Hg in foliar tissue originated from the atmosphere [50]. Evidence from stable isotope signatures of vegetation and soil Hg suggested that 50–80% of soil Hg net deposition at terrestrial sites derived from plant elemental Hg uptake [59, 60]. In the autumn litterfall would represent a new Hg input to terrestrial ecosystems when deciduous trees enter dormancy and leaves senesce, e.g. forests in northeast China. Vegetated ecosystems increase atmospheric deposition compared to their barren counterparts through additional litterfall deposition [5, 10, 55, 61]. Deposition of atmospheric Hg via uptake by vegetation leaf and plant depositions during leaf-off periods were recognized to be a major pathway for the deposition of atmospheric Hg [29] and Hg accumulated in soils [59, 62]. Typical dry deposition velocities for elemental Hg over vegetated surfaces are substantially higher than that over non-vegetated surfaces (Fig. 2D, Table 1) [51].

In China, soil Hg concentrations are quite different among land cover types with the order: forested upland (0.055 mg/kg) > planted/cultivated (0.050 mg/kg) > herbaceous upland/shrubland (0.036 mg/kg) > barren soils (0.015 mg/kg), with average 3.6 times higher in forests than that in barren locations (Fig. 3A). The percentages of Hg contents in surface soil with different land vegetation are quite different as well (Fig. 3B). Atmospheric Hg deposition fluxes in western China, such as the Tibetan Plateau and Xinjiang province ranged from 0.74 to 7.89 µg/m²/yr, was at least one order of magnitude (71.2 µg/m²/yr) lower than that of the east Asia (forest cover) [32]. Result in this study showed different vegetation cover matched the soil Hg distribution in China. In general, Hg concentrations in forest & crops covered soil (Figure S3A) were higher than these in grassland (Figure S3A). The desert region in northwest China exhibited the lowest Hg concentrations in soil because of the barren soils. In western United States soil Hg distribution is strongly linked to vegetation greenness with the same order of land covers in China, and forested land showed more than 2.5 times higher soil Hg level of barren soils [5]. The merged map of vegetation cover and soil Hg concentration in the US (Figure S2) showed that high soil Hg level roughly matched the forest cover. It
is probably due to the facts that the average carbon density of forested upland (89 t C/ha) is much higher than that of grasslands (21 t C/ha) and croplands (5 t C/ha) [63]. Much high carbon fixation capability of forests (20-100 times) in comparison with that of cropland [63] suggested high uptake of gaseous elemental Hg in forested lands considering similar oscillations of Hg uptake by vegetation and net CO$_2$ respiration in the northern Hemisphere [60]. It was estimated that net Hg sequestration via foliage and litterfall is 1000-1200 Mg per year [15, 64], which is half of the annual primary anthropogenic emissions [60], and most of which was transferred to soil.

The forests in China are mainly distributed in the northeastern, southwestern and southern regions (Figure S3A). It has been reported that at Changbai Mountain area in northeast China, annual dry depositional flux was up to 16.5 µg/m$^2$/yr; significantly higher than corresponding wet depositional flux (8.4 µg/m$^2$/yr) [12]. In the Arctic tundra which is a globally important Hg sink [65], the gaseous elemental Hg was taken up by aboveground vegetation and subsequently transferred to tundra soils, leading to high soil Hg concentration, especially in the vegetation growing season in which gaseous elemental Hg deposition were amplified by tundra vegetation [52]. The alpine tundra on Changbai Mountain of northeastern China is the only arctic vegetation with the best-reserved tundra ecosystems in northeast Eurasia [66]. High soil Hg concentrations in this region (Fig. 1) indicated that alpine tundra might be an important Hg sink as well, as Arctic tundra [52]. Higher seasonal dry depositional flux was observed in spring and fall, which is related with forest canopies, than that in winter [12]. At Gongga Mountain area located at high soil Hg region in southwest China, annual wet and dry deposition fluxes of Hg were 26.1 and 66.4 µg/m$^2$/yr [67], respectively. It has been demonstrated that evergreen broadleaf forest in south China exhibited one order of magnitude greater dry deposition via litterfall (75.0 µg/m$^2$/yr) than those in boreal forest in north China [54]. It is acceptable that high soil Hg accumulation in south China was observed because of forest cover.

The presence of any soil cover has been found to decrease atmospheric Hg fluxes in comparison with bare soils, and even leads to a shift from a net atmospheric emission to a net deposition from the atmosphere [57, 68]. For example, studies were carried out within various types of forest systems
across a vast region from the south (South Carolina) to the north (Maine) in the USA during the same season. Mercury emission associated with the litter covered forest floor was extremely low [68], fluctuating around 0 ng/m²/h, with many deposition rather than emission occurring at both daytime and nighttime, irrespective of the forest type, soil type, and variations in weather conditions [11, 68]. The influence of soil cover on Hg emission has been evaluated systematically by integrating a large amount of published data, indicating soil covers substantially reduced Hg emission compared to bare soils in the following order: plant/leave surfaces (0.12 ng/m²/h) < soil-plus vegetation (0.00 ng/m²/h) < soil covered by snow (0.40 ng/m²/h) < soil covered by litter (0.70 ng/m²/h) < bare soil (1.07 ng/m²/h) [69]. Canopy covers in forests and grasslands/shrublands are partially expected to reduce Hg emissions by limiting soil warming and solar loads via shading the forest floor.

3.4 Mercury sequestration by soil organic matter

Soil organic matter contributes to the mobility and emission of Hg in soil. Soil Hg concentrations were positively related to soil organic carbon [63, 65] and Hg emission is inversely correlated with organic content in soils because organic carbon has a high affinity to Hg in soil through functional groups, in particular with thiols [5, 70]. In general, with the increase of SOM in soil, the Hg concentration in soil substantially enhanced (Fig. 2G, 2H, 2I). Similar relationship between soil organic matter and Hg concentration was observed in Europe as well (Figure S4B). In aqueous system, Hg(II) is reduced to volatile elemental Hg by organic acids and these reactions appear to be strongly induced by sunlight [71]. These processes do not occur in soils because sunlight is rather difficult to radiate soil, especially with vegetation cover. Binding of Hg with organic matter inhibited the reduction of Hg(II) to the elemental Hg, and subsequent emission from soil. Strong correlation between soil Hg concentrations and organic carbon has been observed in the western United States [5]. In northeast and southwest China, the distribution of Hg in these regions is geographically coincident with the contents of soil organic matters, especially in northeast China (Figs. 1&2H) [22]. High Hg levels in northeast regions are observed although wet deposition is relatively low (400–800 mm). Soils covered by forests, and enrichment in organic matters enhanced Hg deposition by litterfall and throughfall,
simultaneously depleted Hg emission. At high organic region (peat bog ecosystem) it is estimated that dry deposition of gaseous elemental mercury is dominated (~ 80%), rather than wet deposition (~ 20%) [72].

3.5 Mercury emission from soils
Mercury emission from soil is one of the most important processes of Hg losses in terrestrial environments [6, 73]. It is commonly accepted that gaseous elemental Hg is the major volatile Hg species emitted to the air from soil [6, 73]. It has been indicated that biotic processes have a relatively constant and lower influence on the Hg emission from the soil and the variable abiotic processes is regarded as a significant pathway of Hg removal in uplands [74]. Solar radiation, which induces photoreduction, is the most common and pronounced factor for gaseous elemental Hg emission. Many studies have identified the positive relationship which is attributed to photochemically mediated reduction that converts soil Hg(II) to volatile element Hg [75], although other biotic (microorganisms) and abiotic (organic matters) reduced Hg as well [76]. A strong negative correlation ($R^2 = 0.89$) was observed between the concentration of soil Hg and solar radiation (Fig. 4B), which indicated that increasing solar radiation would accelerate the emission of Hg from soil to atmosphere. The solar radiation intensity in China declines from northwest to southeast (Fig. 4A) [77], indicating that more Hg emission from soil would be occurred in northwest than southeast regions. Previous study shows that the maximum amount of solar radiation reaching the ground in winter is $116 \text{w} \cdot \text{m}^{-2}$, while in winter it is $312 \text{w} \cdot \text{m}^{-2}$. Vegetation cover could decrease the role of solar radiation on Hg emission due to indirect radiation in comparison with direct radiation on bare soil [34]. Direct solar radiation due to poor vegetation cover in northeast China was one of the contributors to low soil Hg level.

In addition, the positive association between soil clay content and Hg concentration [63] suggested that atmospheric Hg was hardly sequestrated into deserted and sandy soils with low clay content in northwest China, which has arid and semi-arid climate [22]. Soil and air temperature are other key factors influencing Hg emission [69]. High temperatures, which are collinear to radiation, generally stimulate Hg evasion from surface soil [78, 79]. Desertified lands (sand desert and Gobi desert) in
northwest China belong to bare soils in arid and semi-arid regions of northwest China. Due to low or no vegetation covers on desertified lands in northwest China, the amount of heat and solar radiation are easy to reach the soil surface which sequentially enhance the Hg emission.

3.6 Factors controlling soil Hg distribution in China
We hypothesize that spatial variability of soil Hg levels in China is potentially caused by the balance between wet depositions associated East Asian summer monsoon, dry deposition associated with forest/vegetation cover and inhibited emission associated with soil organic matter and solar radiation. The model of distribution of soil Hg was setup by multiple linear regression as follows:

$$\log(Hg) = -2.809 + 9.398 \times 10^{-4} \times P + 1.1774 \times 10^{-1} \times NDVI$$
$$+ 8.879 \times 10^{-2} \times \log(SOM) - 9.897 \times 10^{-5} \times (SR),$$

$$P < 0.01, R^2 = 0.51$$

where Hg is the concentration of soil Hg (mg/kg) in the 0–20 cm surface soil, P is average annual precipitation (mm), SOM is soil organic matter (%) and SR is solar radiation (kWh m\(^{-2}\)). Both sample regression analysis of soil Hg with first three variable and multiple linear regression analysis showed positive relationships and with solar radiation showed negative relationship between soil Hg level and those variables. Statistical analysis shows the relative importance of four variables is ranked as: precipitation (49.4%) > NDVI (23.4%) > solar radiation (17.9%) > SOM (9.3%). Precipitation has direct impact on soil Hg through wet deposition, and vegetation cover has direct impact on soil Hg through dry deposition. Considering that precipitation has significant positive effects on vegetation cover [80], it could be reasonable that precipitation has more contribution than NDVI to the soil Hg distribution. In southeast China, high Hg input occurred from 1) high regional atmospheric Hg level caused by anthropogenic emission, 2) high wet deposition (> 800 mm) linked with Asian summer monsoon, 3) high dry deposition linked with vegetated canopies, especially forest and 4) weak solar radiation reach to the surface soil. Vegetation cover, high soil organic matter and moderate solar radiation in
this region limited Hg emission from soil. Although soil Hg emission offsets the Hg increase to some degree in soil by deposition, high deposition fluxes substantially enhance the soil Hg level. In northwest China, Hg deposition is lower than that of the southeast China. Low vegetation cover and high solar radiation, together with low organic matter, indicating much less Hg was accumulated in soil. It is a plausible explanation to low Hg (< 0.03 mg/kg, lower than the national background value 0.065 mg/kg) in surface soil. In northeast and southwest China, high Hg in soil (> 0.04 mg/kg) geographically coincides with the high organic matter, forests cover and weak solar radiation. Elevated Hg is accumulated in soil due to high forest cover and soil organic matter.

4. Conclusions
In conclusion, we suggest that wet deposition associated with summer Asian monsoon, dry deposition associated with vegetation cover and inhibited emission linked with soil organic matter and photoreduction due to solar radiation are the key factors in controlling Hg redistribution in terrestrial environments in other regions of the world. Vegetation cover, soil organic matter and solar radiation are key factors controlling Hg air/soil exchange, such as Hg emission. Monsoon circulations followed by precipitation are critical to the global transport of volatile elements like Hg. There are several major monsoon systems around the world such as Australian summer monsoon, north and south American monsoon, west African monsoon. Monsoon rainfall concurrent wet Hg deposition may affect corresponding spatial distribution of soil Hg. Vegetation cover, especially forest cover is vital factor controlling Hg deposition and its distribution in terrestrial environment, forest/shrubs is a crucial component of the Earth’s surface covered ~ 30% of the world’s land area [81], including tropical seasonal rainforest, tropical montane rainforest, monsoon forest, etc. Future work would focus on a comprehensive quantification of soil Hg heterogeneity and spatial variability not only in China but also at the global scale, especially monsoonal regions and forest covered lands. This would be helpful to understand its potential ecosystem and human health risks in these regions.

Abbreviations
Hg: mercury; SOM: soil organic matter; NDVI: normalized difference vegetation index; NMPRGS: National Multi-Purpose Regional Geochemical Survey; P: average annual precipitation; SR: solar
radiation

Declarations

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Authors’ contributions

Z.Y.Z., G.X.S. designed research and wrote the paper; G.L. and L.Y. contributed data analysis.

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Availability of data and materials

The datasets used and analyzed during the current study are available from the corresponding author on reasonable request.

Ethics approval and consent to participate

Not applicable.

Consent for publication

Not applicable.

Competing interests

The authors declare no competing financial interest.

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Tables
Table 1 Summarized Hg wet deposition, dry deposition, emission and net input (deposition minus emission) in soil of different regions, China.

| Precipitation (mm/yr, i.e. L/m²/yr) | < 400 | 400-800 |
|------------------------------------|-------|---------|
| Regions                            | Northwest | Northeast |
| Ecosystem                          | Bare soil or grassland | Forest and shrub |
| Hg wet precipitation (µg/m²/yr)     | 1.08-4.63 | 8.40 |
| Hg dry precipitation (µg/m²/yr)     | - | 16.5-20.2 |
| Hg emission (µg/m²/yr)              | 9.37 | -1.05 |
| Net Hg input                       | -8.29 ~ -4.74 | 26.0 ~ 29.7 |
| References                         | [32], [67], [69] | [12], [69], [82] | [6], [4], [5] |

Figures
A  Year 1994

Soil Hg (mg/kg)
- $<0.01$
- $0.01-0.02$
- $0.02-0.04$
- $0.04-0.08$
- $0.08-0.20$
- $>0.20$
- No data

B  Year 2014

Soil Hg (mg/kg)
- $0.03$
- $0.06$
- $0.09$
- $0.12$
- $0.15$
- $0.18$
- $0.21$
- $0.24$
- $0.90$
Figure 1

Spatial distribution of soil mercury in China (mg/kg) surveyed at different periods: (A) map surveyed in the late 1980s from a book titled “The Atlas of Soil Environmental Background Value in the People’s Republic of China” 1994, page 50–51, China Environmental Science Press (modified using ArcGIS Geographic Information Systems software version 10.2, Environmental Systems Research Institute Inc, Redlands, Calif) and (B) updated map surveyed in 2000s [2]. More details are presented in reference [16]. Note: The designations employed and the presentation of the material on this map do not imply the expression of any opinion whatsoever on the part of Research Square concerning the legal status of any country, territory, city or area or of its authorities, or concerning the delimitation of its frontiers or boundaries. This map has been provided by the authors.
The maps of factors and relationships of Hg levels in surface soil with these factors. Spatial distribution of (A) wet deposition in China; (D) NDVI (created using ArcGIS Geographic Information Systems software version 10.2, Environmental Systems Research Institute Inc, Redlands, Calif); (G) soil organic matter (created using ArcGIS Geographic Information Systems software version 10.2, Environmental Systems Research Institute Inc, Redlands, Calif). Significant relationships between soil Hg level and (B) precipitation, (E) NDVI and (H) SOM. All data including soil Hg level, precipitation, NDVI and SOM were corrected for variation in sampling intensity by subsampling. Median, range, upper quartile, and lower quartile of soil Hg for each interval of (C) precipitation, (F) NDVI and (I) SOM. Note: The designations employed and the presentation of the material on this map do not imply the expression of any opinion whatsoever on the part of Research Square concerning the legal status of any country, territory, city or area or of its authorities, or concerning the delimitation of its frontiers or boundaries. This map has been provided by the authors.
Median, range, upper quartile, and lower quartile of soil Hg concentrations in different vegetation cover types (A). Large differences exist among different land cover types. The percentages of Hg in Chinese surface soil with different land cover type (B).
The maps of solar radiation in China and relationship of Hg levels in surface soil with solar radiation. Spatial distribution of (A) solar radiation in China; Significant relationships between soil Hg level and solar radiation (B); Median, range, upper quartile, and lower quartile of soil Hg for each interval of solar radiation (C). Note: The designations employed and the presentation of the material on this map do not imply the expression of any opinion whatsoever on the part of Research Square concerning the legal status of any country, territory, city or area or of its authorities, or concerning the delimitation of its frontiers or boundaries. This map has been provided by the authors.

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