



# Anthropogenic influences on mercury in Chinese soil and sediment revealed by relationships with total organic carbon<sup>☆</sup>

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## ARTICLE INFO

### Article history:

Received 25 July 2019

Received in revised form

24 August 2019

Accepted 4 September 2019

Available online 5 September 2019

### Keywords:

Mercury

Topsoil

Surface sediment

Anthropogenic influences

Hg:TOC

China

## ABSTRACT

Rapid industrialization has led to high levels of mercury (Hg) releases from anthropogenic sources in China. When deposited to terrestrial ecosystems, Hg has a high affinity for natural organic carbon. This means that Hg concentrations will vary naturally as a function of the total organic carbon (TOC) content of different soils and sediment. Thus, Hg to TOC ratios in topsoil and surface sediment provides a useful normalized tracer of the anthropogenic impact on Hg contamination. We compiled literature-documented Hg and total organic carbon (TOC) data for topsoil ( $n = 957$ ) and surface sediment ( $n = 1142$ ) in China. Topsoil samples ( $n = 100$ ) were also collected in this study to broaden the spatial coverage. We found large differences in Hg:TOC ratios among topsoil from background sites, agricultural and urban areas, and mining sites and surface sediment from fluvial, coastal, and marine environments. Specifically, a significant increase in Hg:TOC ratios occurred between soils from background sites (median: Hg:TOC = 21.1; Inter-Quartile Range (IQR): 9.67 to 40.7) and agricultural areas (median: 34.1; IQR: 22.1 to 58.7), urban areas (median: 62.1  $\text{ng g}^{-1}$ ; IQR: 34.2 to 154) and mining sites (median: 2780; range: 181 to 43500). Urban and mining sites show the largest increase in Hg:TOC ratios, reflecting elevated anthropogenic Hg inputs in these areas. Fluvial sediment showed higher Hg:TOC ratios (median: 197; IQR: 109 to 389) than coastal (median: 88.3; IQR: 46.8 to 168) and marine sediment (median: 89.7; IQR: 53 to 138), indicating decreased anthropogenic Hg input from rivers to coastal and marine regions. Results of our study suggest Hg:TOC ratios are a useful normalized indicator of the influence of anthropogenic Hg releases on Hg enrichment in topsoil and surface sediment.

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## 1. Introduction

Mercury (Hg) is a naturally occurring heavy metal that undergoes a complex biogeochemical cycle on the Earth's surface. Gaseous elemental Hg ( $\text{Hg}^0$ ) is the dominant species of Hg in the atmosphere (Selin, 2009).  $\text{Hg}^0$  has a long atmospheric lifetime of 0.5–1 years and can be transported both regionally and globally

prior to deposition (Amos et al., 2013). Once deposited to terrestrial and aquatic ecosystems, Hg is subject to microbial methylation, which produces a bioaccumulative neurotoxin known as methylmercury (MeHg) (Gilmour et al., 1992). MeHg biomagnifies through aquatic food chains and reaches concentrations that are  $10^6$  to  $10^7$  times higher in fish compared to the surrounding water (Engstrom, 2007; Ullrich et al., 2001). Elevated levels of MeHg have been observed in many aquatic organisms, even in remote regions without significant point sources of Hg (Mason et al., 2012; Zhang et al., 2014).

The global biogeochemical cycle of Hg is thought to be tightly coupled with the carbon cycle (Couic et al., 2018). Mercury is released to the environment from both natural (e.g., volcanoes,

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hydrothermal vents, biomass burning) and anthropogenic sources (e.g., fossil fuel combustion, non-ferrous metal refining, waste incineration, cement production) (Pirrone et al., 2010). Many of these sources are significant emitters of carbon dioxide (CO<sub>2</sub>) into the atmosphere (De Simone et al., 2015; Sundquist, 1993). Once emitted into the atmosphere, substantial amounts of CO<sub>2</sub> are converted to organic matter via primary production by photosynthetic plants (Prentice et al., 2001). Plants also accumulate a large amount of atmospheric Hg<sup>0</sup> via the uptake by stomata and porous cell walls (Enrico et al., 2016; Jiskra et al., 2018). On a global basis, the atmospheric Hg<sup>0</sup> uptake by vegetation is estimated to be 1000–1100 Mg per year, which enters into soil through litterfall (Wang et al., 2016a; Jiskra et al., 2018). Atmospheric Hg<sup>0</sup> can also react with oxidants such as bromine to form divalent mercury (Hg<sup>II</sup>), which is highly water-soluble and deposits quickly (Amos et al., 2013). Precipitation effectively scavenges Hg<sup>II</sup> and particulate mercury (Hg<sub>p</sub>) from the atmosphere (Åkerblom et al., 2015). A recent study have suggested that Hg concentrations in precipitation are closely linked with total organic carbon (TOC) and consistent Hg:TOC ratios in precipitation have been reported across Sweden (Åkerblom et al., 2015).

Large fractions of Hg in terrestrial and aquatic ecosystems are associated with organic matter due to the strong binding affinity between Hg and thiol (-SH) groups in organic matter (Skylberg et al., 2000). Positive correlations have been observed between Hg and TOC in soil and sediment from different sites around the world (Chakraborty et al., 2015; Kirk et al., 2011; Obrist et al., 2011; Yin et al., 2016b; Yu et al., 2014). The organic matter content in soil and sediment is an important environmental driver governing Hg concentration, Hg:TOC ratios are thought to be relatively variable both spatially and temporally. For instance, anomalously high Hg:TOC ratios have been observed in modern sediments relative to pre-industrial sediments before 1800s, reflecting both an increase in anthropogenic Hg deposition and changes in organic matter inputs to some ecosystems (Leipe et al., 2013). Anomalously high Hg:TOC ratios have also been observed in sedimentary rock profiles during the Earth's extinction events caused by large historical volcanic Hg emissions (Grasby et al., 2017; Them et al., 2019). As such, many studies have established long-term changes in Hg:TOC ratios at a local scale (Chakraborty et al., 2015; Kirk et al., 2011; Obrist et al., 2011; Yin et al., 2016b). The factors driving the variation in Hg:TOC ratios across larger spatial scale; however, remain poorly understood.

China is regarded as the world's largest emitter of anthropogenic Hg, accounting for ~27% of the total global anthropogenic Hg emissions (Pacyna et al., 2010). The extent of Hg emissions and pollution; however, is not evenly distributed across China. For instance, > 90% of China's population and Gross Domestic Product (GDP) occurs over < 50% of China's land surface in eastern and central China (Dijk, 2011). On the other hand, western China hosts one of the most remote and pristine areas on the Earth surface (Wang et al., 2016b). In particular, the Tibetan Plateau (TP) covers ~26% of the total land surface and <1% of the total population of China (Yin et al., 2016a). In this study, we compiled literature Hg and TOC concentrations of topsoil and surface sediment from China and characterized Hg:TOC ratios across different ecosystems and land use types. A nationwide investigation on Hg:TOC ratios of topsoil and surface sediment across China may provide new insights that reflect the degree of anthropogenic activities releasing Hg in the environment, compared to Hg concentration alone. Given the spatially variable Hg sources and the extent of anthropogenic activities, we aim to fully characterize Hg:TOC ratios across topsoil and surface sediment in China and elucidate factors governing varying Hg:TOC ratios across ecosystems and land use types.

## 2. Materials and methods

### 2.1. Literature data

We synthesized data collected in the literature on Hg and TOC concentrations of soil and sediment in China to calculate Hg:TOC ratios. In general, the dataset includes a total of 957 topsoil and 1142 surface sediment (Table A1, with Hg and TOC data paired). The geographical location of the samples is illustrated in Fig. 1. The depth of the samples was summarized in supplementary Table A1. Only the results of topsoil (depth: 0–10 cm) and surface sediment (depth: 0–5 cm) were used in this study.

However, we caution that different methods were used to measure the TOC (Walkley-Black vs %LOI vs EA) and Hg (acid digestion vs combustion), which may result in slightly different values and eventually bring deviations to the study. According to the land use information reported in the literature, soil samples were classified into four types: Background ( $n = 262$ ), agricultural ( $n = 496$ ), urban ( $n = 95$ ), and mining ( $n = 104$ ; Table S1); Sediment were classified into three types: Fluvial ( $n = 97$ ), coastal/nearshore ( $n = 698$ ), and marine/offshore ( $n = 347$ ; Table S1), according to the sampling locations reported in the literature. These classifications are used to report Hg and TOC concentrations, and Hg:TOC ratios for soil and sediment.

As shown in Fig. 1, the background soil concentrations reported in the literature were mainly collected from the TP. Agricultural soil data were mainly available from central and eastern China from areas with active production of various agricultural crops including rice, wheat, and corn. Urban soil data were reported from Beijing and Shanghai, which reflect the most typical urbanized sites in China. Mining soil data were reported from the Wanshan Hg mine in Guizhou Province, the Xunyang Hg deposit in Shanxi Province, and the Xikuangshan Sb-Hg-Au deposit in Hunan province, which are known as large mining sites in China. Fluvial sediment data were from the Songhua, Chao River, and Hai River. Coastal sediments were from the Pearl River Estuary, Yangtze River Estuary, and Yellow River Estuary, which are the most industrialized river channels in China. Marine sediments were reported from the Bohai Sea, Yellow Sea, East China Sea, and South China Sea.

### 2.2. Measured data

To improve the spatial coverage of the data synthesis, we collected additional topsoil (0–10 cm) samples and analyzed them for Hg and TOC concentrations. The locations of new sampling sites are shown in Fig. 1. Briefly, we obtained urban soil ( $n = 65$ ) from 13 cities of Shenzhen, Guangzhou, Wuhan, Chengdu, Nanjing, Qingdao, Ningbo, Zhengzhou, Dalian, Kunming, Fuzhou, Lanzhou, and Tianjin. These cities are among the top 30 largest cities in China in terms of both GDP and population. As the background and agricultural soil reported from the literature were only from the TP and southern China, respectively, we obtained background soil ( $n = 10$ ) from Ningxia and Inner Mongolia in western China and agricultural soil ( $n = 25$ ) from northern China. At each site, soil was collected using a pre-cleaned spade, placed in a plastic bag, and delivered to the State Key Laboratory of Ore Deposit Geochemistry, Institute of Geochemistry, Chinese Academy of Sciences. In the laboratory, all soil samples were freeze-dried at  $-78^{\circ}\text{C}$  for 72 h, ground, homogenized, and sieved to 100  $\mu\text{m}$  mesh, prior to analysis.

Hg concentrations were determined by a DMA-80 Hg analyzer, following an established method (Liu et al., 2013a). Standard reference material (GSS-5, soil) and sample replicates were included for quality assurance and quality control. Recoveries of Hg ranged between 90 and 116% ( $n = 6$ ) for GSS-5, and the relative standard deviations of sample duplicates were all within 10%. The

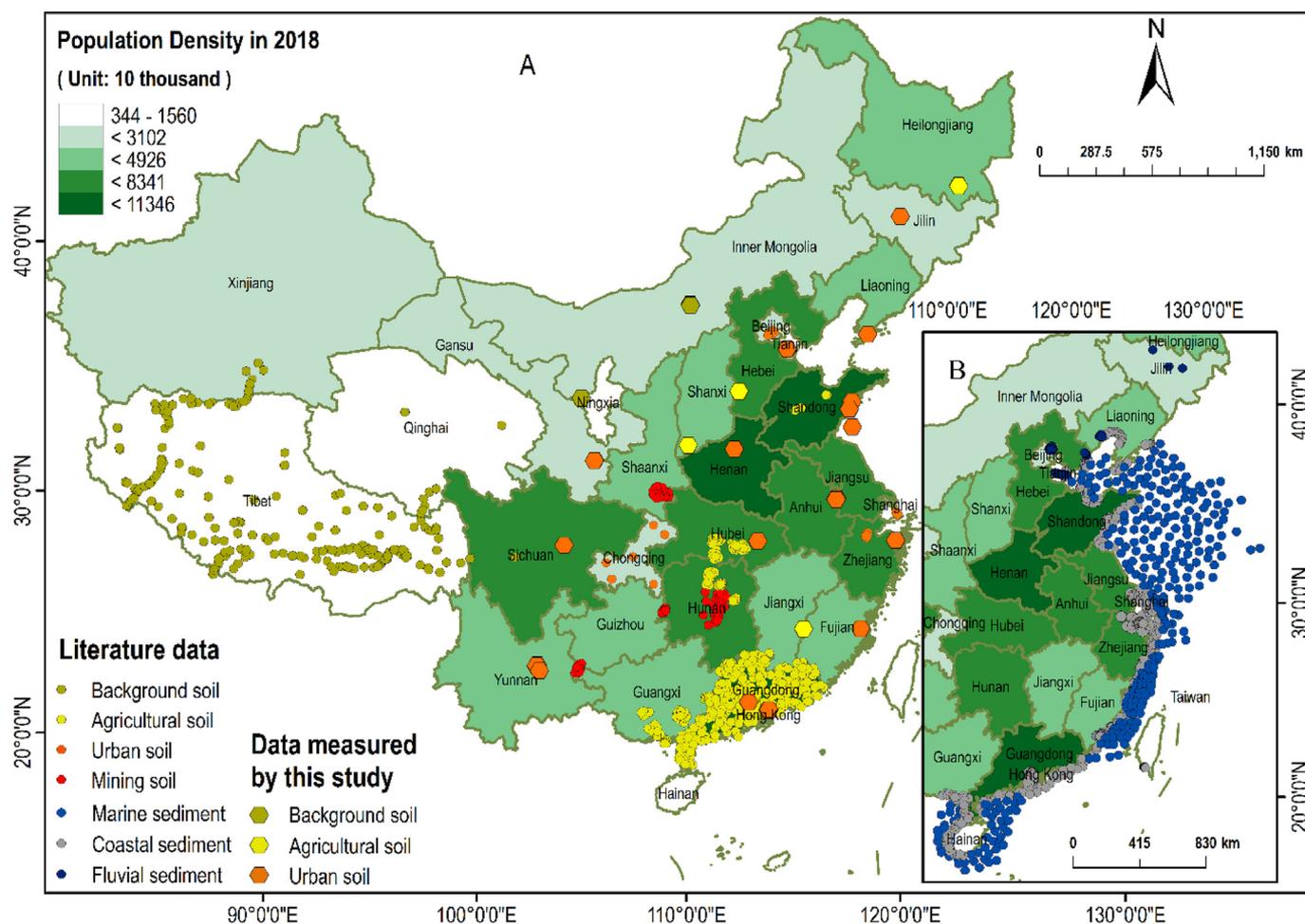


Fig. 1. Map of sample locations with Hg:TOC data synthesized in this study. The population density data in this figure was based on China Statistical Yearbook (2018).

TOC concentration (in %) was measured following the Walkley-Black procedure (Nelson and Sommers, 1996). Duplicate measurements were performed, and the relative standard deviations of sample duplicates were all below 5%. Results are reported in Table A1.

### 2.3. Statistical analyses

All statistical analyses were performed using the SPSS software (version 24). As shown in Fig. 2A–C, Hg and TOC concentrations and Hg:TOC ratios showed large variations. However, the majority of the data (i.e., the middle 50%) showed narrow ranges. Therefore, the median value and interquartile range (IQR) were more robust indicators of the statistical dispersion of the data. Data for Hg and TOC concentrations and Hg:TOC ratios are parametrically distributed. Pearson correlation coefficients ( $r$ ) are used to representing the degree of correlation between variables, and  $p < 0.05$  is used as a criterion to assess statistical significance. One-way analysis of variance (ANOVA) was performed to compare whether Hg and TOC concentrations, and Hg:TOC ratios differed significantly between land use types and ecosystems.

## 3. Results and discussion

### 3.1. Hg and TOC concentrations and Hg:TOC ratios in soil

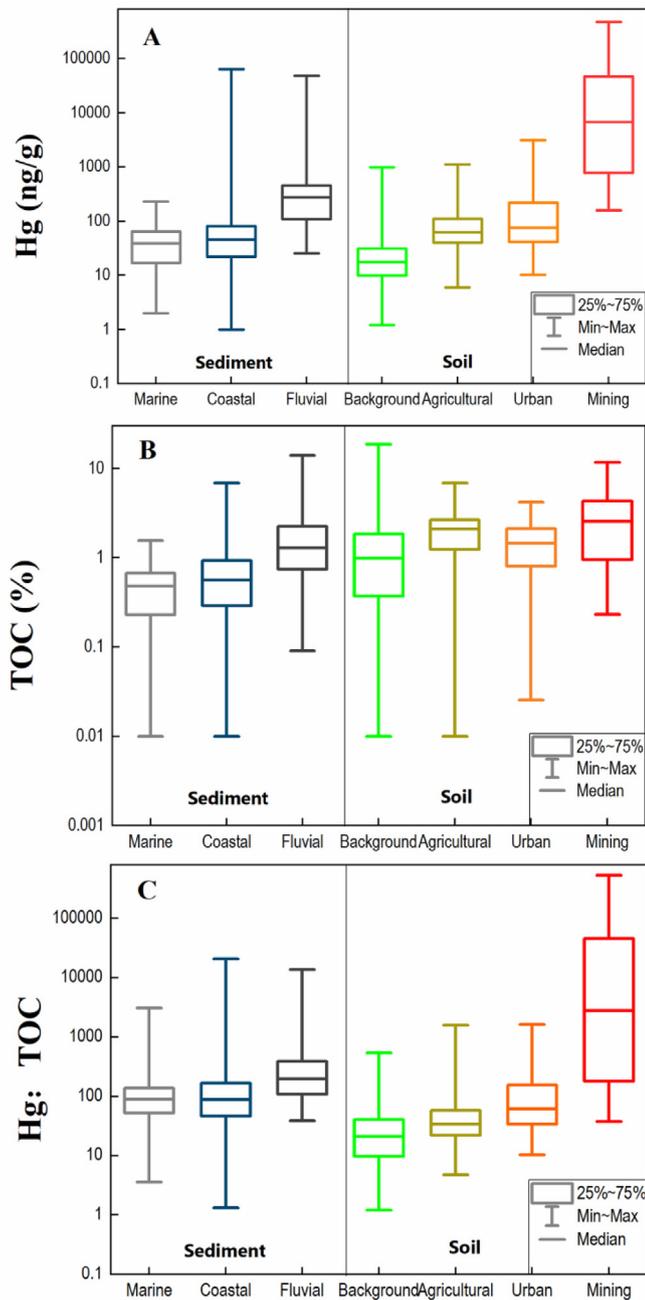
Soil Hg concentrations were significantly different among land

use types (Fig. 2A). Background soil displayed the lowest median Hg concentration of  $17.6 \text{ ng g}^{-1}$  (IQR:  $9.91\text{--}31.2 \text{ ng g}^{-1}$ ;  $n = 257$ ), which is lower than that of agricultural soil (median:  $63 \text{ ng g}^{-1}$ ; IQR:  $40.5\text{--}112 \text{ ng g}^{-1}$ ;  $n = 501$ ), urban soil (median  $76 \text{ ng g}^{-1}$ ; IQR:  $44.4\text{--}218 \text{ ng g}^{-1}$ ;  $n = 170$ ) and mining soil (median:  $6800 \text{ ng g}^{-1}$ ; IQR:  $780\text{--}46250 \text{ ng g}^{-1}$ ;  $n = 104$ ). TOC levels in soil from different regions are illustrated in Fig. 2B. The IQR of background soil was  $0.37\%\text{--}1.87\%$  ( $n = 257$ ), with a median value of  $0.98\%$ , which is lower than those of agricultural (median:  $2.08\%$ ; IQR:  $1.23\text{--}2.65\%$ ;  $n = 501$ ), urban (median:  $1.44\%$ ; IQR:  $0.82\text{--}2.09\%$ ;  $n = 170$ ), and mining soil (median:  $2.55\%$ ; IQR:  $0.95\text{--}4.32\%$ ;  $n = 104$ ).

There were positive correlations between the Hg and TOC concentrations in the background, agricultural, and urban soil ( $r^2 = 0.2$  to  $0.4$ ;  $p < 0.05$  for all; Fig. 3A) but not in the mining soil ( $r^2 < 0.1$ ;  $p > 0.05$ ). As illustrated in Fig. 2C, the background soil showed the lowest median Hg:TOC ratio (median:  $21.1$ ; IQR:  $9.67$  to  $40.7$ ). There was a large increase in Hg:TOC ratios from agricultural soil (median:  $34.1$ ; IQR:  $22.1$  to  $58.7$ ), urban soil (median:  $62.1 \text{ ng g}^{-1}$ ; IQR:  $34.2$  to  $154$ ) to mining soil (median:  $2780$ ; range:  $181$  to  $43,500$ ). This suggests that anthropogenic activities have drastically increased soil Hg:TOC ratios at agricultural, urban and mining sites across China.

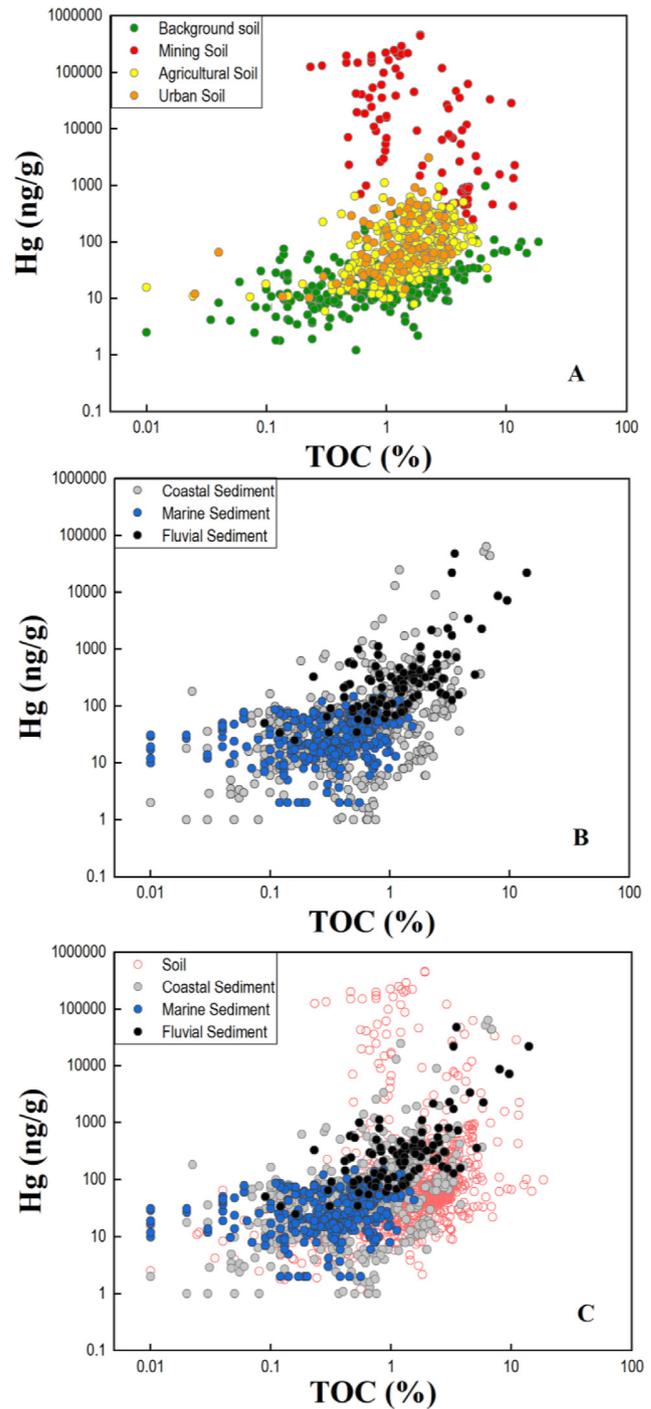
### 3.2. Concentrations and Hg:TOC ratios in sediment

As shown in Fig. 2A, fluvial and coastal sediment showed median Hg concentrations of  $277 \text{ ng g}^{-1}$  (IQR:  $110\text{--}454 \text{ ng g}^{-1}$ ;  $n = 97$ )



**Fig. 2.** The frequency of A) Hg concentration, B) TOC concentration, and C) Hg:TOC ratio for Chinese soil and sediment. Fluvial sediments have significantly higher Hg, TOC and Hg:TOC ratios than other type of sediment ( $p < 0.05$ , ANOVA). Mining soil have significant higher Hg, TOC and Hg:TOC ratio than other type of soil ( $p < 0.05$ , ANOVA). Background soil have significant lower Hg, TOC and Hg:TOC than other type of soil ( $p < 0.05$ , ANOVA).

and  $46 \text{ ng g}^{-1}$  (IQR:  $22\text{--}81.59 \text{ ng g}^{-1}$ ;  $n = 698$ ), respectively. Marine sediment displayed much lower median Hg concentration of  $39.4 \text{ ng g}^{-1}$  (IQR:  $17\text{--}64.2 \text{ ng g}^{-1}$ ;  $n = 347$ ). Elevated Hg concentrations in fluvial and coastal sediment were also reported for sediments collected from Minamata Bay, Japan ( $86\text{--}3460 \text{ ng g}^{-1}$ , Li et al., 2009), the Northeastern U.S. coast (Maine, Massachusetts, Rhode Island, Connecticut and New Jersey;  $221\text{--}2962 \text{ ng g}^{-1}$ ) (Kwon et al., 2014), and the California Coast ( $87\text{--}3870 \text{ ng g}^{-1}$ , Davydov et al., 2018), which are known to be influenced by historical and recent anthropogenic Hg sources.



**Fig. 3.** Relationship between Hg and TOC concentrations for different types of soil and sediment. Panel A) shows soil classified by land use type, Panel B) shows sediment classified by ecosystem type, and C) soil and sediment.

As shown in Fig. 2B, TOC concentrations decreased from the fluvial (median: 1.29%; IQR: 0.74–2.24%), coastal (median: 0.60%; IQR: 0.29–0.93%), to marine sediment (median: 0.48%; IQR: 0.23–0.67%). The Hg:TOC ratios in general exhibit the similar decreasing trend, from the fluvial (median: 197; IQR: 109 to 389), coastal (median: 88.3; IQR: 46.8 to 168), to marine sediment (median: 89.7; IQR: 53 to 138). The magnitude of Hg:TOC ratios observed in this study is similar to the ranges of those reported from other regions. The coastal sediment of New York/New Jersey

Harbor, known to be contaminated by modern wastewater discharges and legacy Hg during the Industrial Revolution, had a relatively elevated median Hg:TOC ratio of 122 (4.11–194, Hammerschmidt et al., 2008).

We observed significant positive relationships between the Hg and TOC concentrations among all sediment types ( $r = 0.4$  to  $0.7$ ;  $p < 0.05$  for all; Fig. 3B), suggesting that TOC is an important factor influencing Hg accumulation in sediment. This is consistent with many previous studies that observed positive correlations between Hg and TOC in sediments on a local-regional scale, such as Hudson River, Chesapeake Bay and Bay of Fundy ( $r = 0.55$ ; according to Heyes et al. (2006)), coastal Florida ( $r = 0.58$ , according to Kannan et al. (1998)), and New York/New Jersey Harbor ( $r = 0.4$ , Hammerschmidt et al. (2008)). The linear relationships between sediment Hg versus TOC concentrations for this study were also within the ranges of the background and mining soil and similar to agricultural and urban soil of this study (Fig. 3C).

### 3.3. Factors governing Hg concentration and Hg:TOC ratios in soil

Soil Hg concentrations synthesized in this study increase in the order of background, agricultural, urban, and mining soil (Fig. 2A). This suggests that the extent of anthropogenic Hg influence is the main factor governing Hg levels across different soil types. The lowest median Hg concentration ( $17.7 \text{ ng g}^{-1}$ ) was observed in the background soil and those predominantly collected from the TP, indicating limited anthropogenic influence in this region. The higher median Hg concentration in the agricultural soil ( $63 \text{ ng g}^{-1}$ ) compared to the background soil ( $17.6 \text{ ng g}^{-1}$ ) can be explained by the fact that most agricultural soils locate in eastern and central China. In these populated areas, atmospheric deposition and the use of industrial and domestic sludge and wastewater irrigation might contribute to the high concentrations in agricultural soil significantly (Davydov et al., 2018; Pillay et al., 2007). In the urban soil, the elevated median Hg concentration ( $76 \text{ ng g}^{-1}$ ) can be explained by Hg emissions from industrial activities (e.g., electric producing, coal combustion and other Hg-related factories), domestic biomass burning, sludge incineration, and vehicle emissions (Jiang et al., 2006). In mining areas, the highest median Hg concentration ( $6800 \text{ ng g}^{-1}$ ) in the soil is directly related to the extraction and processing of Hg-rich ores. Mining and refining result in the extensive release of Hg to the surrounding environment as illustrated by previous studies (Li et al., 2012; Rytuba, 2003).

Chen et al. (2015) recently reported variable Hg levels in soil from different Chinese provinces. Another recent study by Kwon et al. (2018) used a global scale atmospheric chemistry transport model (GEOS-Chem) to simulate present-day total atmospheric Hg deposition across different Chinese provinces. Fig. 4 shows that in eastern and central China, where high atmospheric Hg deposition

occurs, the soil is associated with significantly elevated Hg concentration. Apart from Hg-contaminated sites and regions proximity to point mercury sources, soil primarily receives Hg from the atmosphere through wet deposition, dry deposition, and vegetative uptake (Demers et al., 2013). Soil Hg levels have long been used to estimate the impacts of locally and regionally emitted atmospheric Hg in many regions (Yu et al., 2014). It should be noted that the soil chemistry is complicated (Duan et al., 2018; Zhang et al., 2019). Although soil is a sink of Hg, the accumulation of Hg in soil are also influenced by a number of factors (vegetation type, soil type, precipitation, temperature, sulfur content, etc), and many of these factors are changing rapidly in the last decades (Obriest et al., 2018; Gruba et al., 2019).

Hg:TOC ratios increased in order from the background soil (21.1), agricultural soil (34.1), urban soil (62.1) to the mining soil (2780). This supports the premise that anthropogenic activities are responsible for increasing Hg inputs to the soil at varying degrees. The consistently low Hg:TOC ratio, as illustrated in Fig. 3A, occurs in the background soil. Even the high TOC soil collected from the TP showed low soil Hg concentration (Fig. 3A). While vegetation is known to play an important role in delivering both Hg (via litterfall, throughfall) and carbon (via primary production) to soil, we speculate that the low annual average temperature in the TP ( $-2.9$ – $3$  °C; Xu et al., 2006) may be leading to low decomposition rates of soil organic matter. This can prevent efficient sequestration of Hg to soil in addition to low direct atmospheric mercury deposition to soil via wet and dry deposition (Fig. 4). Similarly, high TOC contents and low Hg concentration were observed in the Arctic peat bogs and permafrost soil due to low organic matter decomposition rates at low temperature (Enrico et al., 2016; Schirrmeister et al., 2011).

In the case of the agricultural soil, in which the magnitude of vegetative Hg and carbon delivery (litterfall, throughfall, primary production) to soil is high and anthropogenic activities can facilitate artificial TOC input through sewage irrigation and fertilizer applications, we observed relatively low and narrow ranges in TOC concentration in the agricultural soil. Unlike many North American and European countries, the majority of biomass, including both grain and straw, are harvested from Chinese farmlands. Straw burning remains an important energy source for home heating and cooking in small Chinese villages. In fact, previous studies have observed significant TOC reduction in Chinese agricultural soil caused by long-term straw harvesting (Jurgensen et al., 1997; Laird and Chang, 2013). Even without the elevated TOC levels in the agricultural soil, atmospheric deposition of Hg via wet and dry deposition, irrigation via wastewater, and high bedrock Hg concentration (Kwon et al., 2018) explain increased Hg concentrations as well as Hg:TOC ratios in the agricultural soil. In the case of the urban and mining soil, the relatively narrow ranges in TOC with elevated Hg concentration are due to changes in land cover caused

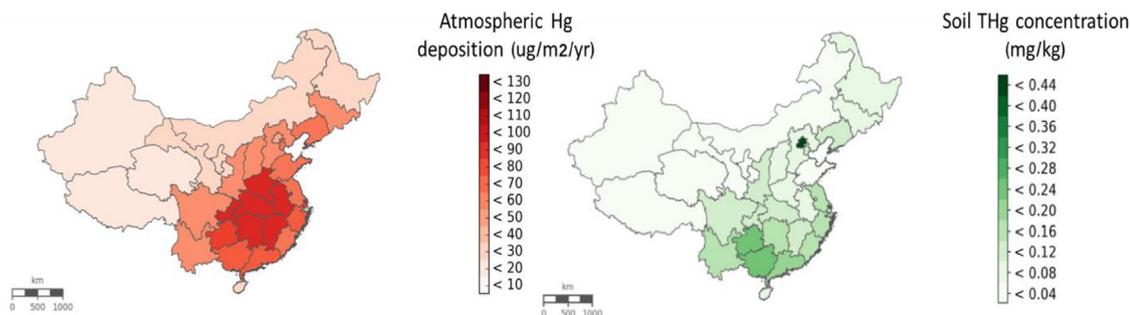


Fig. 4. Map of Chinese provinces showing present-day atmospheric Hg deposition (based on Kwon et al. (2018)) and soil Hg concentration (based on Chen et al. (2015)).

by urbanization and direct excavation for mining activities, leading to the removal of topsoil organic matter. Particularly for the mining soil, a substantial amount of Hg directly released from mining activities appears to overwhelm the natural Hg:TOC ratio.

In summary, our study suggests that anthropogenic activities have significantly altered the natural ratio of Hg and TOC in surface soil across China. Modern anthropogenic activities result in elevated Hg inputs (i.e., direct atmospheric deposition, mining activities). Land use types govern the TOC level in soils (i.e., straw burning, urbanization, excavation) and determine the balance between Hg concentrations and % TOC in the soil and the observed Hg:TOC ratios across China. The role of vegetation in controlling both Hg and carbon input to the soil is likely only important in remote regions such as the TP, where there is a sufficient amount of biomass for Hg<sup>0</sup> uptake by foliage and primary production by vegetation (Wang et al., 2016c). Mercury is readily bound to soil organic matter due to the strong binding affinity between Hg and thiol (-SH) groups in organic matter (Skylberg et al., 2000).

#### 3.4. Factors governing Hg concentrations and Hg:TOC ratios in sediment

As with soil, the decreasing trend in Hg concentrations from the fluvial, coastal, to marine sediment suggests that the extent of anthropogenic Hg input is the most important factor leading to Hg concentration variation across the sediment type. Marine sediment typically receives atmospherically deposited Hg from long distances and from coastal regions (Mason and Sheu, 2002; Selin et al., 2008). In contrast, fluvial sediment are subject to anthropogenic Hg input via historical and recent industrial activities and wastewater discharge (Donovan et al., 2016; Liu et al., 2013b; Shi et al., 2005). Runoff of Hg bound to soil organic matter has also been suggested to be an important Hg input process for fluvial sediment in a number of forested and agricultural areas (Branfireun et al., 2005; Grigal et al., 2000; Jonsson et al., 2014). Coastal sediment receives Hg from various sources and processes such as industrial runoff, long-range and local anthropogenic atmospheric deposition, and fluvial transport of both legacy and recent Hg sources (Balcom et al., 2004; Kwon et al., 2014; Yin et al., 2015). This explains the widest range in Hg concentrations observed in coastal sediment. Many previous studies have reported a linear relationship between Hg and TOC concentrations in fluvial, estuarine, and coastal sediment (Hammerschmidt et al., 2008; Heyes et al., 2006; Kannan et al., 1998; Schartup et al., 2013). These studies have collectively shown significant linear relationships between sediment Hg and TOC concentrations, suggesting that TOC, regardless of location, plays an important role in the amount of Hg accumulation and retention in aquatic sediment.

The decreasing trend in the Hg:TOC ratios from the fluvial, coastal to marine sediment provide insights into sources, transport, and fate of Hg and TOC across the aquatic environment. Terrestrial runoff from the background, agricultural, urban and mining soil is likely the dominant input pathway for both Hg and TOC to the fluvial sediment. The median THg concentration of the fluvial sediment was within the range of that for the four types of soil (Fig. 2A). The fluvial sediment also showed similar median TOC concentrations and Hg:TOC ratios with these soil (Fig. 2B and C). Our observations are consistent with a previous study by Liu et al. (2016), who estimated that over 80% of Hg detected in Chinese freshwater is originated from natural sources such as leaching, runoff, and erosion processes from land as well as legacy anthropogenic sources including erosion of Hg-contaminated land surfaces, resuspension of Hg-contaminated river bed sediment, and land and water management practices (Liu et al., 2016). These processes are also responsible for the input of TOC into the fluvial

sediment. Moreover, our fluvial sediment data were collected from northeastern China (near Beijing), a region that is known for rapid economic development and associated Hg and TOC emissions and releases via industrial activities (Zhang et al., 2015). Once Hg is introduced to river channels, sediments with high organic matter content are effective in scavenging and retaining Hg from the water column (Conaway et al., 2003; Sunderland et al., 2006; Taylor et al., 2012). This is consistent with the highest Hg:TOC ratios observed in our fluvial sediment compared to other sediment type reported in this study.

The decreasing patterns in Hg concentration and TOC concentration, and Hg:TOC ratio in our fluvial, coastal to marine sediment are consistent with a previous study that suggested that fluvial Hg transport is generally associated with the movement of organic matter in both dissolved and particulate form (Liu et al., 2012). Mercury that remains in the water column, or that has resuspended from the fluvial sediment, is subsequently transported to coastal and marine regions via riverine transport. Amos et al. (2014) estimated that riverine transport accounts for 11–48% of the total Hg input to coastal regions globally. Generally, the majority (~70%) of Hg bound to particulate phases, rather than dissolved phases, in the riverine water column is deposited to coastal sediment (Amos et al., 2014). Meanwhile, it appears that both Hg and TOC are diluted in coastal and marine sites, which experience relatively little direct input of Hg and TOC. Marine sediment typically displays low TOC levels due to lower primary productivity compared to terrestrial ecosystems. The magnitude of TOC input to marine sediment, responsible for sediment mercury retention, is also thought to be limited both seasonally and spatially and governed by biological decay and carbon remineralization processes (Sunderland et al., 2009). Anthropogenically derived Hg and TOC released from industrial, agricultural and domestic activities to fluvial sediment, in combination with coastal and marine transport, explain the observed large spatial scale pattern of Hg:TOC ratios across Chinese sediment presented here.

#### 4. Environmental implications

This study, for the first time, characterized the nationwide Hg:TOC ratios in different types of topsoil and surface sediment over China. Our results suggest that the correlation between Hg and TOC in topsoil and surface sediment not only occurs in specific sites but also on a national scale, suggesting that TOC is a major factor affecting Hg accumulation in soil and sediment. In this regard, this study demonstrates that Hg:TOC ratios can trace anthropogenic Hg influence in a more clear way, compared to the use of Hg concentrations alone. The Hg:TOC ratios have dramatically shifted to much higher values in different types of soil (especially agricultural, urban and mining soil) and sediment (especially fluvial and coastal sediment), due to anthropogenic emissions of Hg into the environment. Notably, it is believed this scenario also occurs across the world due to the rapid emission of Hg, especially since the rapid industrialization and globalization in the 1950s (Amos et al., 2013; Yin et al., 2018).

Furthermore, Hg:TOC ratio also provide insight to sources and processes that cause Hg input and transport across geochemical reservoirs in the modern age. Upon changes in anthropogenic Hg emissions, as well as human activities altering land use, the observed Hg:TOC ratios could change quickly. Changes in atmospheric Hg emissions and deposition due to the implementation of the Minamata Convention on Mercury could be tracked in topsoil and surface sediment by using their Hg:TOC. We suggest that the establishment of such baseline Hg:TOC ratios could be an important part of global monitoring and effectiveness evaluation by different countries for understanding sources and processes

leading to changes in Hg input.

## Acknowledgments

This work was supported by the Natural Science Foundation of China (41873047).

## Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.envpol.2019.113186>.

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