Published in final edited form as:

Environ Sci Technol. 2018 June 19; 52(12): 6945–6956. doi:10.1021/acs.est.8b01319.

Impact of Water-Induced Soil Erosion on the Terrestrial Transport and Atmospheric Emission of Mercury in China

Maodian Liu^{†,‡}, Qianru Zhang[†], Yao Luo[†], Robert P. Mason[‡], Shidong Ge[†], Yipeng He[‡], Chenghao Yu[†], Rina Sa[§], Hanlin Cao^{||}, Xuejun Wang^{*,†}, and Long Chen^{*,‡}

[†]Ministry of Education Laboratory of Earth Surface Process, College of Urban and Environmental Science, Peking University, Beijing 100871, China

[‡]Department of Marine Sciences, University of Connecticut, 1080 Shennecossett Road, Groton, Connecticut 06340, United States

[§]College of Geographical Sciences, Inner Mongolia Normal University, Hohhot, Inner Mongolia 010022, China

^{||}Finance Department, Guanghua School of Management, Peking University, Beijing 100871, China

^{*}Key Laboratory of Geographic Information Science (Ministry of Education), East China Normal University, Shanghai 200241, China

Abstract

Terrestrial mercury (Hg) transport, induced by water erosion and exacerbated by human activities, constitutes a major disturbance of the natural Hg cycle, but the processes are still not well understood. In this study, we modeled these processes using detailed information on erosion and Hg in soils and found that vast quantities of total Hg (THg) are being removed from land surfaces in China as a result of water erosion, which were estimated at 420 Mg/yr around 2010. This was significantly higher than the 240 Mg/yr mobilized around 1990. The erosion mechanism excavated substantial soil THg, which contributed to enhanced Hg(0) emissions to the atmosphere (4.9 Mg/yr around 2010) and its transport horizontally into streams (310 Mg/yr). Erosion-induced THg transport was driven by the extent of precipitation but was further enhanced or reduced by

*Corresponding Authors: Tel: +86-10-62759190; xjwang@urban.pku.edu.cn (X.W.). Tel: +86-21-54341246; chenlong@geo.ecnu.edu.cn (L.C.).

ORCID

Xuejun Wang: 0000-0001-9990-1391

Long Chen: 0000-0001-9574-7307

Notes

The authors declare no competing financial interest.

ASSOCIATED CONTENT

Supporting Information

The additional information includes the. The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.8b01319.

Land elevation and the distribution of permanent streams (Figure S1); land cover (Figure S2); distributions of precipitation and average temperature (Figure S3); a map of Chinese river basins (Figure S4); distributions of water-induced soil removal fluxes (Figure S5); land covers in three typical regions (Figure S6); Loess Plateau and Karst rocky desertification (Figure S7); the results of uncertainty analysis in this study (Figure S8); soil THg concentration data (Table S1); the erosion grade, erosion modulus, and sediment delivery ratio (Table S2); and summaries of THg fluxes associated with water erosion and deposition (Tables S3 and S4) (PDF)

vegetation cover and land use changes in some regions. Surface air temperature may exacerbate the horizontal THg release into water. Our analyses quantified the processes of erosion-induced THg transport in terrestrial ecosystems, demonstrated its importance, and discussed how this transport is impacted by anthropogenic inputs and legacy THg in soils. We suggest that policy makers should pay more attention to legacy anthropogenic THg sources buried in soil.

Graphical Abstract

INTRODUCTION

Methylmercury (MeHg), a potent neurotoxin, is widely distributed in the biophysical environment and threatens the health of wildlife and humans.^{1–4} The global release of total mercury (THg) to the environment is estimated to have increased 1.5- to 3-fold since the industrial revolution.^{5,6} Quantification of the THg amount released into the environment is essential to assess Hg global biogeochemical cycling accurately, and such evaluations have been performed at the global scale.^{7–11} Nevertheless, the mechanism of horizontal terrestrial transport of THg is still poorly quantified. Previous studies have already indicated that terrestrial discharges of inorganic Hg, natural organic matter and nutrients, can significantly enhance MeHg accumulation in aquatic biota.^{12,13} THg in natural soil is contributed from both background sources and previous anthropogenic THg emission and deposition. The soil THg contributed from the latter is the so-called legacy anthropogenic source THg that recycles through the biosphere.¹⁴ At the global scale, THg contributed from natural processes (including background and legacy anthropogenic sources) to the aquatic environment cannot be ignored.¹¹ Quantification of these processes is particularly important, since the terrestrial THg released to the aquatic environment can directly enhance the MeHg level in aquatic biota.

The terrestrial ecosystem is a net sink of THg globally, since the terrestrial ecosystem receives substantial atmospheric THg deposition.^{2,15} Rainfall-runoff events that induce soil erosion occur naturally, but they can be accelerated by human cultivation and deforestation activities and result in the enhancement of terrestrial material transport, as shown for carbon.^{16–18} This likely also transports THg, which can then be buried in the redepositional landscape environment, transported into rivers, and eventually delivered to lake and marine ecosystems, especially after flood events.^{16,19} Limited data from field measurements in other countries have suggested that erosion of Hg-contaminated soil is an important source of Hg contamination to the aquatic environment.^{20–22} For example, the extensive human-

induced deforestation taking place in the Amazon Basin in Brazil has released approximately 500 g/km²·yr of THg from soil into the nearby aquatic environment.²¹ This suggests that terrestrial THg transport induced by water erosion following human activities constitutes a major disturbance of the natural Hg cycle. Existing measurements from selected small watersheds cannot represent other regions. Clearly, estimates over different regions should be obtained based on soil THg measurement and detailed field surveys of water erosion to allow for an accurate assessment of this potential flux.¹⁷ Here, we provide such estimates for China and examine their impacts on Hg fate and transport in the region.

China occupies a large land area of approximately 960×10^4 km², and its land elevation varies from sea level in the coastal region to approximately 8800 m in Tibet and includes plains, deserts, and mountains, etc. (Figures S1 and S2, Supporting Information, SI). China covers several different climatic zones, ranging from the subtropical zone in southern China to the cool-temperate zone in northern China.²³ The annual precipitation varies from 20 mm in the dry area of the northwest region to 2600 mm in the south (SI Figure S3). Consequently, complicated water erosion processes occur in different regions of China. Scientists have made extensive efforts to quantify atmospheric THg emissions from direct anthropogenic sources and natural processes in China.^{24,25} In total, it has been estimated that 530 Mg of THg was emitted into the air from direct anthropogenic sources in China in 2014,²⁴ while inputs from natural processes (including background source and reemission of legacy anthropogenic source) were 470 Mg/yr.²⁵ In recent papers, we have quantified that 110 Mg of THg was released from direct anthropogenic sources into the aquatic environment in China in 2015.^{26,27} While the terrestrial ecosystem is an important source for the aquatic environment, to our knowledge, however, quantification of the THg amount released from natural processes into the aquatic environment has not been adequately studied in China.

In this study, we first quantify the THg removal from soil induced by water erosion in China during the last 20 years, based on abundant soil THg measurement data and detailed national field surveys of water erosion in different periods during this time frame. We then quantify the THg release into the aquatic environment and atmosphere caused by these erosion processes in China. This study presents a new understanding about the potentially significant role of terrestrial THg transport in the global THg cycle and asks a question that has not been investigated in any detail elsewhere. Our evaluation could therefore help to identify its impacts on the environment and will assist in the implementation of the Minamata Convention in China and elsewhere.

MATERIALS AND METHODS

Soil THg Removal Induced by Water Erosion

To quantify the soil THg removal induced by water erosion, we applied a database of water-induced soil erosion in China from the *Ministry of Water Resources of China*, which can be downloaded at <http://162.105.205.87/chinaerosion/>.¹⁷ The database was created based on two detailed national survey data sets of water erosion in 1995–1996 and 2010–2012. These two national surveys combined remote-sensing images and field survey data to provide the spatial distribution of water erosion for a total of 2359 counties in China in the two periods.

We then combined the water erosion information with two databases of soil pollution surveys in which 4095 and 38 393 samples were collected in different locations for all types of soil that reflected different land uses or functional zones (such as cropland, urban area and unused land) in two periods of 1986–1990 and 2005–2013, respectively (Table S1, SI).^{28–30} Sampling and measurements for the pollution surveys were conducted by the major scientific research and national monitoring institutions in China, such as the Chinese Academy of Sciences and major universities. Sampling methods for these two databases were based on the grid (or cell) sampling method, i.e., the sampling sites are randomly distributed within a regular grid of $n \times n$ km² (approximately 50×50 and 15×15 km² for the first and second pollution surveys, respectively) and have at least one sampling site in each grid.³⁰ Samples were first digested with a concentrated acid mixture (HNO₃–HF–HClO₄) and then analyzed by Cold Vapor Atomic Absorption Spectrometry (CVAAS) or Cold Vapor Atomic Fluorescence Spectrometer (CVAFS), with greater than 80% sample recovery.^{28–30} Hence, the results of these two national surveys should not have large systemic errors. The surface soil THg concentration data (0–20 cm) from these databases were used in this study because THg usually accumulates in the surface soil, and soil erosion happens mainly in this layer.^{17,25}

A Monte Carlo method was applied to simulate the probabilistic distributions of all results in the form of a statistical distribution.²⁷ The erosional component of soil THg was derived using the following equation:³¹

$$E_{\text{THg},i}(x) = \sum_{jk} (C_{\text{THg},j}(x) \times M_k \times A_{jk} \times K) \quad (1)$$

where $E_{\text{THg},i}(x)$ is the probabilistic distribution of the flux of eroded soil THg induced by water erosion (Mg/yr) in each river basin (or province) i . For this, we classified all 2359 counties in China into 58 secondary basins, following a previous study (SI Figure S4).²⁶ We also modeled the flux of eroded soil THg induced by water erosion in 31 provinces based on the administrative division in China (except Taiwan Province, Hong Kong and Macao).²⁷ In eq 1, $C_{\text{THg},j}(x)$ is the probabilistic distribution of the surface soil THg concentration (ng/g) in county j . The concentrations of soil THg followed log-normal distributions that were considered in the uncertainty analysis based on the Monte Carlo method in this study.^{28–30} Also, M_k is the erosion modulus (i.e., the mass of soil removed induced by water erosion, Mg/km²·yr) for erosion grade k , which followed the uniform distribution based on the *Criterion of Classification of Soil Erosion* (SL190–2007).³² In eq 1, A_{jk} is the water erosion area (km²) of erosion grade k in county j . We divided water erosion areas into five grades based on the criterion,³² as was done in a previous study (SI Table S2).¹⁷ Following previous studies, a uniform distribution with a fixed coefficient of deviation was assumed for the water erosion area data (5%), since the data were derived from official statistics.^{26,27} Finally, K is the unit conversion factor for THg (10^{-9} in this study).

THg Atmospheric Deposition in China

A previous study simulated the atmospheric THg deposition in China using the GEOS-Chem chemical transport model (version 9.02, <http://geos-chem.org>), based on an anthropogenic emission inventory from 2010 in China.³³ In this study, we reran the model to separate the distribution of wet and dry deposition of THg in 2010 in China, since there is still a lack of monitoring data of THg deposition across some regions of China. We also separated the deposition into three Hg forms, i.e., elemental Hg(0), gaseous soluble Hg(II) and nonvolatile particulate Hg(P). Details of the simulation method can be found in the previous study.³³

To convert the raster data ($1/2^\circ \times 2/3^\circ$ horizontal resolution) of THg deposition into fluxes for each county in China, the kriging interpolation method was applied in this study. The kriging interpolation method provides a method of estimation based on the variogram function and spatial structure analysis. In this study, we used the ordinary kriging method to depict the spatial variability distribution of the atmospheric THg deposition, and the simulation was accomplished using IDRISI version 17.0. Standard errors of the interpolation results ($\pm 4.1\%$) were considered in the uncertainty analysis based on the Monte Carlo method.

Erosion-Induced Horizontal Terrestrial THg Transport

We quantified the amount of THg released into streams from the confluence of erosion-induced (water-erosion-induced) THg erosion and wet deposition of atmospheric THg in China, based on mass balance principles.³⁴ The flux of THg released into streams was derived using the following equations, as used in previous studies:^{8,17}

$$S_{\text{THg},i}(x) = S_{\text{PHg},i}(x) + S_{\text{DHg},i}(x) \quad (2)$$

$$S_{\text{PHg},i}(x) = \sum_{jk} [(E_{\text{THg},jk}(x) + \text{WD}_{\text{PHg},j} + \text{WD}_{\text{DHg},j} \times R_{1,i}) \times \text{SDR}_k] \quad (3)$$

$$S_{\text{DHg},i}(x) = \sum_j [(W_{\text{DHg},j}(1 - R_{1,i}) \times R_{2,i}] \quad (4)$$

where $S_{\text{THg},i}(x)$ is the probabilistic distribution of THg released into a stream (Mg/yr) of a river basin (or province) i . We divided $S_{\text{THg},i}(x)$ into particulate ($S_{\text{PHg},i}(x)$) and dissolved ($S_{\text{DHg},i}(x)$) phases following Amos et al.⁸ $\text{WD}_{\text{PHg},j}$ and $\text{WD}_{\text{DHg},j}$ are PHg and DHg in wet deposition of atmospheric THg (Mg/yr) in county j ; SDR_k is the sediment delivery ratio (%) of erosion grade k , which is defined as the ratio of the mass of sediment yield (Mg/yr) at the outlet of a small catchment to the mass of soil eroded (Mg/yr) in the catchment. The SDR_k is positively correlated with erosion severity.³⁵ Ranges of SDR for different erosion grades were derived following a uniform distribution from 10% to 100% and are provided in SI

Table S2.^{17,35} We have assumed that THg is from water erosion of each county's releases into local streams, as assumed in a previous study,¹⁷ since more than 98% of the counties in China have permanent streams (SI Figure S1). In the equations above, $R_{1,i}$ is the ratio reflecting whether W_{DHg} is absorbed ($R_{1,i} > 0$) or released ($R_{1,i} < 0$) by the eroded soil in basin i ; $R_{2,i}$ is the fraction (%) of precipitation discharged into streams in river basin i , ranges from 85% to 90% in different primary river basins in China, and is derived from the annual precipitation data and riverine water capacity.³⁶ To our knowledge, there is a lack of studies that focus on the mechanism that details how THg is absorbed to eroded soil from wet deposition. Zheng et al. indicated that the enrichment behavior and transport mechanisms of THg are more closely related to soil particle transport than those associated with organic matter in surface layers, based on rainfall-runoff experiments.³⁷ Following Amos et al., we estimated $R_{1,i}$ based on the water-particulate partition coefficient (K_D) of THg as follows:⁸

$$\text{Log}_{10}K_D = \text{Log}_{10}\left(\frac{C_{\text{PHg}} \times 1000}{C_{\text{DHg}}}\right) \quad (5)$$

$$C_{\text{THg}} = C_{\text{PHg}} \times \text{TSS} + C_{\text{DHg}} \quad (6)$$

$$R_1 = \frac{C_{\text{DHg}} - C_{\text{DHg}'}}{C_{\text{DHg}}} \quad (7)$$

where C_{PHg} and C_{DHg} are the initial PHg (ng/g) and DHg concentrations (ng/L) in the precipitation; C_{THg} is the concentration of THg (ng/L) in the temporary runoff (precipitation mixed with eroded soil); TSS is the total suspended sediment concentration (g/L) calculated by the mass of eroded soil (Tg) and precipitation (km^3); and $C_{\text{DHg}'}$ is the DHg concentration (ng/L) in the temporary runoff. C_{DHg} can be derived from eqs 5 and (6) as follows:

$$C_{\text{DHg},j(x)} = \frac{C_{\text{THg},j(x)} \times 1000}{\text{TSS}_j \times K_D + 1000} \quad (8)$$

In this study, the $\text{Log}_{10}K_D$ value in the runoff was set as 4.7 ± 0.30 (mean $\pm$ standard deviation), which was calculated by a previous study, based on the abundance of measurement data from the published literature.⁸ Hence, C_{DHg} in eq 8 can be replaced by $C_{\text{DHg}'}$ in the eq 7. We considered the standard deviation of the $\text{Log}_{10}K_D$ value in the uncertainty analysis. We preliminarily estimated the contributions of total anthropogenic THg (contributed from direct and legacy anthropogenic sources) and background sources to the erosion-induced THg release into streams, based on the fully coupled, seven-reservoir box model developed by Amos et al. and updated in our previous study.^{15,38} In this study,

we defined the surface soil of China as a single reservoir, and reran the model at a millennium scale. We separated the contributions of atmospheric deposition from direct anthropogenic emission, previous anthropogenic emission, and background sources based on the model. Details of the inventories used in the model and modeling methods are described in previous studies.^{15,38}

THg Emission to the Atmosphere from Water Erosion-Induced Soil Turnover

It is commonly accepted that erosion induces a source for CO₂ in the erosional area due to mixing during transport, since part of the surface soil (0–20 cm in this study) can be mixed into the subsurface soil layer (>20 cm) during precipitation and erosion processes and because the increased decomposition of the new surface soil organic carbon (SOC) provides an additional CO₂ source.^{16,39} Increases in Hg(0) emissions from natural soil due to precipitation events have also been observed in field studies.^{40–42} However, there is still a lack of studies on the case of Hg(0) emissions to the atmosphere from surface soil in the erosion impacted area caused by water erosion on a large scale. While Wang et al. provided a comprehensive estimation of the emission of Hg(0) from natural surfaces in China based on mechanistic models,²⁵ we further quantified the contribution of Hg(0) emissions from the erosional area from erosion to the total emissions of Hg(0) in the surface soil in China, using the approach outlined below:^{17,39}

$$ES_{\text{re, Hg(0)}, i}(y) = \sum_{jk} \left(ES_{\text{Hg(0)}, j}(y) \times \frac{A_{jk}}{A_{\text{total}, j}} \times (1 - \text{SDR}_k) \times R_{3,d} \times R_4 \right) \quad (9)$$

where $ES_{\text{re, Hg(0)}, i}(y)$ is the probabilistic distribution of the amount of Hg(0) emissions from the erosion area to the atmosphere (Mg/yr) due to water erosion in the river basin (or province) i ; and $ES_{\text{Hg(0)}, j}(y)$ is the probabilistic distribution of the amount of total Hg(0) emissions from surface soil (Mg/yr) in county j . We applied the ordinary kriging method to depict the spatial variability distribution of the Hg(0) emission in each county as mentioned above. $A_{\text{total}, j}$ is the total area (km²) of county j ; $(1 - \text{SDR}_k)$ is the fraction of soil that is redeposited for erosion grade k . Following previous studies,^{17,35} we assumed that all of the eroded soil THg is redeposited within the same county. Jing et al. found that the amount of eroded soil accumulated within its source watershed was higher with smaller SDR.³⁵ Yue et al. verified this phenomenon based on comparing observed SDR values and erosion grades in representative areas.¹⁷ In eq 9 above, $R_{3,d}$ is the turnover rate of soil (yr⁻¹) at depth d and can be calculated as follows:³⁹

$$R_{3,d} = R_{3,0} \times e^{(-2.6 \times d)} \quad (10)$$

where $R_{3,0}$ is the turnover rate (yr⁻¹) at depth 0 cm in the erosion site, and e is the natural base. A previous study found that $R_{3,0}$ is 0.03 yr⁻¹ (ranges from 0.02 to 0.04 yr⁻¹) in China.¹⁷ On the basis of eq 10, 56 to 73 years is needed for the subsurface soil from 20 to 30 cm to

become the surface soil, caused by precipitation and erosion processes.³⁹ R_4 in eq 9 is the enhancement ratio of Hg(0) flux observed after a precipitation event, which ranges from 0 to 16 times (average 5.8) greater, depending on the soil water content.^{41,42} We assumed that the subsurface soil is initially dry before its turnover into the surface layer due to water erosion.⁴¹ All ranges were considered in the uncertainty analysis.

Finally, we quantified the probabilistic distribution of the amount of THg redeposition after being eroded from the surface soil in a river basin (or province) i ($D_{\text{THg},i}(x)$), as detailed below:¹⁷

$$D_{\text{THg},i}(x) = \sum_j [(E_{\text{THg},j}(x) + \text{WD}_{\text{Phg},j} + \text{WD}_{\text{DHg},j} - S_{\text{THg},i}(x)] \quad (11)$$

Soil Mass Balance Model

To quantify the THg flux from direct anthropogenic sources into surface soil and complete the map of THg cycling in the environment in China, we developed a preliminary surface soil mass balance model, based on mass balance principles.³⁴ The THg fluxes in the mass balance model were expressed by an input–output equation, as follows:^{15,31}

$$\text{input}_{\text{THg}} + \Delta_{\text{THg}} = \text{output}_{\text{THg}} \quad (12)$$

$$\text{WD}_{\text{THg}} + \text{DD}_{\text{THg}} + D_{\text{THg}}(x) + \Delta_{\text{THg}} = E_{\text{THg}}(x) + \text{ES}_{\text{Hg}(0)}(y) + \text{EV}_{\text{Hg}(0)} \quad (13)$$

where DD_{THg} is the dry deposition of atmospheric THg (Mg/yr). Δ_{THg} is the type of accumulation or depletion of THg in the model, which could lead to the increase ($\Delta_{\text{THg}} < 0$) or decrease ($\Delta_{\text{THg}} > 0$) of soil THg concentration.⁴³ $\text{EV}_{\text{Hg}(0)}(z)$ is the amount of Hg(0) emission from vegetation, which was considered as a part of the mass balance model, since plant litter would become part of the surface soil with its decomposition.^{15,44} Overall, the uncertainties of chemistry-and meteorology-related models are difficult to quantify; thus, following previous studies, we set 30% as the uncertainty range for THg deposition results from the GEOS-Chem model.^{45,46} Landfill was not included in the model, since the soil THg concentration databases do not contain landfill information.

Other Databases Used in This Study

Land cover data were extracted from maps of 300-m annual global land cover in 1992 and 2010 from the European Space Agency using ArcGIS version 10.3 (web site: <https://www.esa-landcover-cci.org/>) to compare changes in land cover during this period in China. Annual precipitation and average temperature data (SI Figure S3) were collected from the China Meteorological Administration (web site: <http://www.cma.gov.cn/>).

Statistical Analyses

All statistical analyses and fitting models presented in this study were conducted in R version 3.3.3 (R Project for Statistical Computing). Significant levels were determined at the $P < 0.05$ level (*) and < 0.01 (**). No statistical methods were used to predetermine the sample size in this study.

Uncertainty Analysis

A Monte Carlo simulation method was applied and performed in 10 000 runs to analyze the robustness of THg fluxes in this study.^{26,27} Details of parameter settings are provided above. Median values and the 60% confidence intervals (ranging from 20% to 80%) of the statistical distributions were modeled to quantify the THg fluxes and to characterize the uncertainty ranges.^{26,27}

RESULTS AND DISCUSSION

The Soil THg Level Changed in ~20 Years

The average surface soil THg concentration was 70 ± 68 (mean $\pm$ standard deviation) ng/g around 2010 in China and has increased from 44 ± 43 ng/g around 1990 ($P < 0.01$ **).^{28–30} This means that vast quantities of THg are stored in the surface soil layer in China, which was 1.0×10^5 Mg (range, 8.4×10^4 to 1.2×10^5 , using a 60% confidence interval (CI) based on the Monte Carlo simulation) around 2010, and substantially increased from 6.3×10^4 Mg (5.2×10^4 to 7.6×10^4) around 1990. The increase indicates that terrestrial ecosystems in China have received extensive inputs of anthropogenic THg (including direct and legacy sources) in the last several decades.^{27,47} A previous study found that 530 Mg of anthropogenic THg was emitted into the atmosphere in 2014 in China and had increased rapidly from 250 Mg in 1990, consistent with our estimates on the increasing rate.²⁴ However, the THg release from direct industrial sources into surface soils still lacks adequate quantification in China.

Soil THg levels in most provinces have increased significantly from 1990 to 2010 due to the rapid increase in human activities and sources, such as wastewater release and increased traffic densities during the period.^{27,48} The significant relationship between soil THg concentration and population density (an effective index that reflects regional economic development) (Figure 1b, c), as well as similar slopes of the correlation plots ($k = 0.25$, logarithmic transformation) over time also indicates that human activity can significantly enhance soil THg contamination levels.⁴⁹ For example, higher soil THg levels are found in Guizhou and Guangxi provinces due to continuous Hg mining activities in Southwest China.⁵⁰ In Jiangsu Province, the soil THg concentration decreased (54% in the 20 years), but the reason for it is unclear. Soil THg concentrations were lower in some provinces, such as Xinjiang, which has the largest administrative area but a low population density. Therefore, the influence of human activity was less significant (Figure 1a). Soil THg concentration was also lower in Tibet around 1990 (24 ± 16 ng/g, lower than remote regions such as in Arctic),⁵¹ reflecting that Tibet is one of the cleanest areas in China. The concentration in Tibet around 2010 was 2.6-fold higher than that around 1990 due to the increase in human activity

in this area; for example, it has the highest per capita THg release from domestic sewage among all provinces in China.²⁷

Flux of Soil THg Removal Induced by Water Erosion

In total, 420 Mg/yr (340 to 520 Mg/yr) of THg was removed from soil induced by water erosion around 2010 and significantly increased from 240 Mg/yr (190 to 300 Mg/yr) around 1990. Area-weighted average soil THg removal rates were 25 and 44 g/km²·yr around 1990 and 2010 in China, respectively. The upstream of the Yangtze River (Changjiang) basin (including the Jialing, Yalong, Wu, and Min River basins, SI Figure S4) contributed most of the flux around 1990, which was 80 Mg/yr (with a 97 g/km²·yr removal rate), followed by the main stream of the Yellow River (Huanghe) basin (43 Mg/yr with a 48 g/km²·yr removal rate, Figure 2a). The most intensified soil erosion regions are concentrated in the Loess Plateau (middle of the Yellow River basin, (SI Figure S5)).¹⁷ In total, 1400 and 1600 Tg/yr of soils were eroded from the upstream of the Yangtze River and Yellow River basins around 1990, respectively, which are both Cambisols, according to the classification of the Food and Agriculture Organization (FAO) (<http://www.fao.org/>). High soil THg concentrations in the upstream of the Yangtze River basin, which was approximately 2-fold higher than that in the Yellow River basin around 1990, shifted the ranking of these two river basins, which was different from that found for SOC removal in China.¹⁷

The case of erosion-induced soil THg removal around 2010 showed different patterns, compared with that around 1990 (Figure 2b, c), and this was due to changes in erosion levels and/or changes in soil THg. The upstream of the Yangtze River basin contributed 120 Mg/yr (with a 160 g/km²·yr removal rate), followed by the Xi River (Xijiang) basin (78 Mg/yr with a 260 g/km²·yr removal rate) and the main stream of the Yellow River basin (38 Mg/yr with a 42 g/km²·yr removal rate, Figure 2b). For the upstream region of the Yangtze River basin, the mass of water-induced soil removal decreased 22% from 1990 to 2010, while the average soil THg concentration increased 68% (Figure 2d). The Wu River (Wujiang) basin, which is located in Guizhou province (southeast of the upstream of the Yangtze River basin, SI Figure S4), has the highest erosion-induced soil THg removal rate (320 g/km²·yr) around 2010. As mentioned above, the soil THg concentration in Guizhou province was higher than most other provinces in China due to continuous Hg mining activities.⁵⁰ Upstream of the Yellow River basin, from 1990 to 2010, water-induced soil removal decreased 50% (Figures 2d and S6) due to the implementation of the “returning farmland to forests and grassland” initiative, which is a huge national-scale program for soil conservation in Northern China that was implemented after 1990, especially for the Loess Plateau regions (SI Figure S7).⁵² However, different from what happened for SOC removal,¹⁷ the effect of the program on THg removal (SI Figure S5) has been negated in the Yellow River basin, due to the increase in soil THg concentration in this basin (Figure 2d), which is associated with the increase in human activities, such as coal burning and nonferrous metal smelting.²⁴

The Xi River basin (the largest secondary basin of the Pear River basin in South China, SI Figure S4), which occupies 3.1% of the land territory of China, contributed 19% of the total erosion-induced soil THg removal in China around 2010. The Xi River flows through Yunnan, Guizhou, Guangxi, and Guangdong Provinces (located in Southwest and South

China regions), where soil THg levels were high (Figure 1a). The Xi River then flows into the South China Sea. The mass of soil removal in the Xi River basin increased rapidly from 1990 to 2010 (240%), which was a sharp contrast to the case in the main stream of the Yellow River basin (Figure 2d). Karst rocky desertification (SI Figure S7), induced mainly by human activities (mostly agricultural cultivation), has transformed this natural soil-covered karst area into a rocky landscape.⁵³ Karst rocky desertification has happened in the Southwest and South China regions in recent years, especially in Guizhou province (SI Figure S5).⁵³ The sharp increase in erosion-induced soil THg removal in the Xi River basin from 1990 to 2010 (6-fold increase, Figure 2d) could therefore be attributed to the increase in agricultural cultivation, plus high soil THg concentration, wet climate conditions (1800 to 2300 mm of annual precipitation, SI Figure S3) and the extensive karst landscapes in the river basin.

Fluxes of soil THg removal in other river basins, such as the north part of the Hai River (Haihe) basin, also have shown significant increases in THg flux over the 20 years (72%, Figure 2c, d). The mass of water-induced soil removal in the northern part of the Hai River basin decreased 26% from 1990 to 2010, which could be attributed to the increase in vegetation coverage and a rapid urbanization process (SI Figure S6),⁵⁴ while at the same time, the soil THg concentration in this basin increased 170% in 20 years. Similar results were also found in the Pearl River Delta (South China, SI Figures S4 and S6).

Erosion-Induced Horizontal Terrestrial THg Release into the Aquatic Environment

In total, 310 Mg/yr (250 to 400 Mg/yr, with a 32 g/km²·yr average flux) of THg was released into aquatic environments around 2010 in China from erosion (Figure 3a), while 110 Mg of THg was released from direct anthropogenic sources (including industrial wastewater and municipal sewage) into aquatic environments in China in 2010.^{26,27} In contrast to THg emissions into the atmosphere,^{24,25} the contribution of THg release from natural processes (including background and legacy anthropogenic sources) into aquatic environments is significantly larger than direct anthropogenic release (Figure 3b). The rate of THg release into the aquatic environment around 2010 in China for soil erosion was approximately 3-fold higher than other remote and pristine environments (0.10 to 4.0 g/km²·yr), as estimated by a previous global assessment but significantly lower than the release rate in a human deforestation location of the Amazon Basin in Brazil (500 g/km²·yr).^{11,21} We estimated that 69% of the erosion-induced THg released into aquatic environments around 2010 in China was from total anthropogenic sources, of which 24% resulted from the direct anthropogenic emission, and previous anthropogenic emission accounted for 76% of the anthropogenic THg in surface soil. This is a conservative estimation because (1) the contribution of some point sources and direct anthropogenic sources (such as sewage sludge) may have been neglected,²⁷ and (2) the fraction of background THg release induced by human-derived land cover changes should be considered as anthropogenic perturbations; however, this is difficult to quantify. Nevertheless, we can conclude from these estimates that the flux was dominated by THg from anthropogenic sources in China.

The THg releases from direct anthropogenic processes into aquatic environments were high in Eastern China, especially in the North and East China regions, due to their high

population densities and elevated economic development level.^{26,27} Erosion-induced THg release was high in the Loess Plateau (central Northern China), Central and Southwest China regions (Figure 3a, b), where climate change is a factor (mostly precipitation) and soil type and human activities (such as Hg mining and cultivation) play important roles. THg released into the aquatic environment was also high in the Yangtze, Yellow and Pearl River basins (Figure 3a). The three river basins cover more than $300 \times 10^4 \text{ km}^2$ area, and their main streams account for 70% to 80% of the total freshwater and suspended sediment discharge from Mainland China into adjacent seas.³⁶ The Yangtze, Yellow, and Pearl River basins received 120, 35, and 50 Mg/yr THg around 2010, while direct anthropogenic THg releases were 25, 14, and 6.8 Mg in 2010, respectively (Figure 3b).

Considering the secondary river basins, undoubtedly, the Wu River basin had the highest THg release rate into the aquatic environment ($220 \text{ g/km}^2\cdot\text{yr}$) around 2010, followed by Xi River ($150 \text{ g/km}^2\cdot\text{yr}$) and the main upstream section of the Yangtze River ($100 \text{ g/km}^2\cdot\text{yr}$) basins, which are all located in the Southwest China region and flow through Guizhou or Guangxi provinces. At the county level, THg release rates of some counties, such as Liupanshui and Zunyi (located in Guizhou province), reached $\sim 1200 \text{ g/km}^2\cdot\text{yr}$ around 2010, where severe rocky desertification and mining activities have been documented.^{50,53,55}

In this study, we explored the potential driving factors, including regional climate factors (precipitation and temperature), elevation change and population density, of the THg release into aquatic environments induced by water erosion (Figure 4). We selected the best-fitting model suggested by the R Project software to test the strength of the relationships between each of the variables with the amount of THg released into aquatic environments, following the approach in a previous study.⁵⁶ As predicted, the relationship between precipitation and THg release was significant, and it verified the dominant role of precipitation in China (Figure 4a) in driving erosion, similar to what was found in previous studies.^{57,58} Increases in precipitation and THg release were not linear, indicating that the influence of precipitation on erosion-induced THg release into aquatic environments may have a threshold value, which is approximately 1400 mm/yr in this study. The relationship obeyed the Langbein–Schumm curve.⁵⁹ However, the mechanism of erosion-induced THg release is also linked closely with sedimentary function and form, which may be influenced by other factors such as tectonics, topography, soil type, land cover, etc.^{4,57,58,60,61}

The change in elevation is an important factor influencing sediment transport into aquatic environments.⁵⁷ Figure 4b shows the inverse-U-shaped pattern between elevation change and erosion-induced THg release into aquatic environments in the study area. It suggests that THg release rates were high in the transition area (500 to 2000 m) between the rolling country in Eastern China and the plateau sections (i.e., the Tibetan Plateau and Mongolian Plateau) in western China. Low THg release rates in the plateau sections were because of the low precipitation and human activity intensity in these regions (SI Figure S4).

The influences of changes in land cover on erosion-induced THg release into the aquatic environment are also significant.²¹ Figure 4c shows the inverse-U-shaped pattern between the population density and the erosion-induced THg release. We assumed that the appearance of the inflection of the curve was mainly caused by (1) substantial decline of soil

erosion due to large-scale vegetation restoration projects such as those taking place in the Loess Plateau,⁵² and (2) the transformation of the natural land into an impervious surface in some locations (SI Figure S6). Both of these factors can significantly reduce the influence of precipitation on THg released from eroded soils. It should be noted that in some cases, urban stormwater THg fluxes might be high due to the higher accumulation of dry THg deposition on impervious surfaces, which could be washed off in precipitation events.⁶² The high vegetation cover rate in some regions can also weaken the influence of precipitation, such as in the South China region. In this region, the precipitation was significantly negatively correlated with the THg release ($R^2 = 0.3749$, $P < 0.0001^{**}$), where the vegetation coverage is the highest in China (SI Figure S2).⁵⁹ Many studies have shown similar results in that they found that vegetative cover was crucial for runoff generation and for the increase in soil moisture, which can alter the erosional activity.^{58,59,63} Although the Southwest China region has similar vegetation coverage as South China, the increase in karst rocky desertification induced by excessive agricultural cultivation has enhanced the impacts of precipitation in this region, similar to the case of human deforestation in the Amazon Basin in Brazil.²¹

Finally, the positive relationship between the surface air temperature and the erosion-induced THg release into the aquatic environment (Figure 4d) indicated that the regional surface air temperature may be a potential driving factor of pollutant release in China, which could be partly explained by the relatively low soil moisture content driven by higher evaporation rates in regions of higher surface air temperature, which could make the soil surface more sensitive to precipitation erosion.^{64–66} The most remarkable correlation was found in the Tibetan region ($R^2 = 0.3619$, $P < 0.0001^{**}$), which may be attributed to the significant change in ecosystem properties with temperatures associated with the elevation changes.⁶⁷ Further study is needed to improve the understanding of the interactions between the changing climate conditions and THg release into aquatic environments.

Erosion-Induced Hg(0) Emission in the Erosional Area

During the precipitation and erosion processes, part of the subsurface soil (>20 cm) in the erosion site could be turned over and become new surface soil.³⁹ Hg(0) emission from this new surface soil could be enhanced by precipitation (see Methods), which will induce a new source of Hg(0) emission into the atmosphere (6.0 Mg/yr around 2010). When the subsurface soil becomes the new surface soil as a result of erosion, the previous surface soil THg would also be buried, and the Hg(0) emission from this soil would no longer exist (1.1 Mg/yr). Hence, water erosion can induce a net increase in Hg(0) emission to the atmosphere in the erosion areas in China, which was estimated at 4.9 Mg/yr around 2010 (1.1% of the total emission from soil).²⁵ The contribution rate is relatively low compared with CO₂ emissions from this process (~4%) in China, due to the high degradation of SOC into CO₂ (20% to 63%).¹⁷ We did not consider Hg(0) emissions during the process of water-induced soil transport in this study. To our knowledge, there is insufficient information on the mechanism of Hg(II) reduction during the soil-transport process.

Implications

In this study, we provide information that fills an important knowledge gap concerning the large-scale quantitative description of lateral transport and atmospheric emissions of THg

induced by soil erosion by water. This improved knowledge is critical for the understanding of the Hg biogeochemical cycle and the control of Hg pollution. Our analyses indicate that vast quantities of THg may move laterally over the land surface in China as a result of water erosion (Figure 5). The erosion conveyor excavates substantial soil THg at eroded locations and buries it in the redepositional areas or transports it downslope horizontally over land surfaces into streams. It also enhances the Hg(0) emission to the atmosphere. These processes are influenced by precipitation, tectonics, topography, soil type, THg background concentration, anthropogenic activities, land cover, and, potentially, global warming. More research with the adoption of multiple statistical analysis methods are needed to identify and further clarify potential drivers for Hg released into aquatic environments. On the basis of the mass balance principles used here,³⁴ we estimated that there was 410 Mg (230 to 640) of THg accumulated in surface soils (excluding landfills) in China in 2010 (Figure 5), which might be attributed to other anthropogenic THg inputs (excluding atmospheric deposition) such as municipal sewage sludge and intentional use of THg (contributed 33 and 35 Mg, respectively).^{27,68}

As the first attempt to quantitatively describe the processes of THg transport in terrestrial ecosystems in China, we highlight some potential biases in the present study, including the mismatch of time periods between soil THg concentrations and soil erosion inventories, and the use of THg concentration data from two different national surveys, which might increase the uncertainties of the results. Also, soil particle size and soil organic matter might influence the transport of Hg,³⁷ and were not considered in the present study. A previous study indicated that the differences of the contents of soil organic carbon in surface soil in most regions in China between 1980s and 2000s were within $\pm 2\%$.⁶⁹ In addition, we involved the vegetation in the soil mass balance model, but processes such as the deposition of litterfall and transformation between different forms of Hg in vegetation are not included in the model, since they are not well studied in China. Increasing evidence suggested that vegetation plays an important role in connecting atmospheric and edaphic Hg cycles.^{51,70} Further studies should be carried out on the THg fluxes and their uncertainties reported in the present study, when more measurement data and better estimation methodologies are available in the future.

We further compared our estimated THg releases to aquatic environments with the amount of THg discharged into Chinese adjacent seas. In total, 420 Mg/yr of THg was released from direct anthropogenic sources and natural processes into aquatic environments in the early 2010s in China, while 160 Mg/yr of riverine THg was discharged into Chinese adjacent seas in 2010.³¹ Although beyond the range of the present study, we hypothesize that substantial riverine THg is buried in reservoirs and lakes in China. Construction of hydroelectric dams is rapidly increasing in China, and as most riverine THg is particulate-bound, it will be trapped by the reservoirs. For example, the Three Gorges Dam, the world's largest dam construction, induced retention of $\sim 90\%$ of the sediment of the Yangtze River in the Three Gorges Reservoir in recent years.⁷¹ Future studies are needed for both the quantification of THg retention in reservoirs and for the future impacts of their remobilization on the fate of the legacy THg in reservoirs, especially for regions with extensive reservoir construction hotspots such as the Yangtze and Yellow River basins, as well as for other global hotspots such as the Mississippi, Ganges, and Amazon River basins.⁷²

So far, the map of the Hg budget in environmental media in China has been almost completed (Figure 5). We identified four key conclusions based on accurately evaluating erosion-induced Hg transport in China: (1) soil erosion induced substantial terrestrial THg transport in China, which is critical for the Hg biogeochemical cycle; (2) the enhancement of Hg pollution in soil can significantly negate the effects of soil protection policies; (3) rocky desertification could result in tremendous negative impacts on the THg transport; and (4) the changing climate may exacerbate the horizontal transport of THg. Our analysis provides new understandings of Hg transport within the terrestrial ecosystem. These understandings are crucial for advancing the science of Hg pollution study and for the management of its environmental and human impact.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

Acknowledgments

The authors very much appreciate the editor's and reviewers' insightful comments and suggestions on the paper. This work was funded by the National Natural Science Foundation of China (41571484, 41630748, 41701589, 41571130010, 41130535, and 41471403) and, for Robert Mason, by the US National Institute of Environmental Health Sciences (Dart-mouth Superfund Research Program; P42 ES007373). L.C. thanks the China Postdoctoral Science Foundation Grant (2017M611492).

References

1. Driscoll CT, Mason RP, Chan HM, Jacob DJ, Pirrone N. Mercury as a global pollutant: sources, pathways, and effects. *Environ Sci Technol*. 2013; 47(10):4967–4983. [PubMed: 23590191]
2. Mason RP, Sheu GR. Role of the ocean in the global mercury cycle. *Glob Biogeochem Cycle*. 2002; 16(4):40-1–40-14.
3. Krabbenhoft DP, Sunderland EM. Global change and mercury. *Science*. 2013; 341(6153):1457–1458. [PubMed: 24072910]
4. Obrist D, Kirk JL, Zhang L, Sunderland EM, Jiskra M, Selin NE. A review of global environmental mercury processes in response to human and natural perturbations: Changes of emissions, climate, and land use. *Ambio*. 2018; 47(2):116–140. [PubMed: 29388126]
5. Lamborg CH, Hammerschmidt CR, Bowman KL, Swarr GJ, Munson KM, Ohnemus DC, Lam PJ, Heimbürger L-E, Rijkenberg MJ, Saito MA. A global ocean inventory of anthropogenic mercury based on water column measurements. *Nature*. 2014; 512(7512):65–68. [PubMed: 25100482]
6. Streets DG, Horowitz HM, Jacob DJ, Lu Z, Levin L, Ter Schure AF, Sunderland EM. Total Mercury Released to the Environment by Human Activities. *Environ Sci Technol*. 2017; 51(11):5969–5977. [PubMed: 28448134]
7. Nriagu JO, Pacyna JM. Quantitative assessment of worldwide contamination of air, water and soils by trace metals. *Nature*. 1988; 333(6169):134–139. [PubMed: 3285219]
8. Amos HM, Jacob DJ, Kocman D, Horowitz HM, Zhang Y, Dutkiewicz S, Horvat M, Corbitt ES, Krabbenhoft DP, Sunderland EM. Global biogeochemical implications of mercury discharges from rivers and sediment burial. *Environ Sci Technol*. 2014; 48(16):9514–9522. [PubMed: 25066365]
9. Horowitz HM, Jacob DJ, Amos HM, Streets DG, Sunderland EM. Historical mercury releases from commercial products: Global environmental implications. *Environ Sci Technol*. 2014; 48(17):10242–10250. [PubMed: 25127072]
10. Sundseth K, Pacyna JM, Pacyna EG, Pirrone N, Thorne RJ. Global Sources and Pathways of Mercury in the Context of Human Health. *Int J Environ Res Public Health*. 2017; 14(1):105–119.

11. Kocman D, Wilson SJ, Amos HM, Telmer KH, Steenhuisen F, Sunderland EM, Mason RP, Outridge P, Horvat M. Toward an Assessment of the Global Inventory of Present-Day Mercury Releases to Freshwater Environments. *Int J Environ Res Public Health*. 2017; 14(2):138–154.
12. Jonsson S, Andersson A, Nilsson MB, Skjellberg U, Lundberg E, Schaefer JK, Åkerblom S, Björn E. Terrestrial discharges mediate trophic shifts and enhance methylmercury accumulation in estuarine biota. *Science Advances*. 2017; 3(1):e1601239. [PubMed: 28138547]
13. Schartup AT, Balcom PH, Soerensen AL, Gosnell KJ, Calder RS, Mason RP, Sunderland EM. Freshwater discharges drive high levels of methylmercury in Arctic marine biota. *Proc Natl Acad Sci U S A*. 2015; 112(38):11789–11794. [PubMed: 26351688]
14. Pirrone N, Cinnirella S, Feng X, Finkelman RB, Friedli HR, Leaner J, Mason R, Mukherjee AB, Stracher GB, Streets DG. Global mercury emissions to the atmosphere from anthropogenic and natural sources. *Atmos Chem Phys*. 2010; 10(13):5951–5964.
15. Amos HM, Jacob DJ, Streets DG, Sunderland EM. Legacy impacts of all-time anthropogenic emissions on the global mercury cycle. *Glob Biogeochem Cycle*. 2013; 27(2):410–421.
16. Van Oost K, Quine T, Govers G, De Gryze S, Six J, Harden J, Ritchie J, McCarty G, Heckrath G, Kosmas C. The impact of agricultural soil erosion on the global carbon cycle. *Science*. 2007; 318(5850):626–629. [PubMed: 17962559]
17. Yue Y, Ni J, Ciais P, Piao S, Wang T, Huang M, Borthwick AG, Li T, Wang Y, Chappell A. Lateral transport of soil carbon and land–atmosphere CO₂ flux induced by water erosion in China. *Proc Natl Acad Sci U S A*. 2016; 113(24):6617–6622. [PubMed: 27247397]
18. Lal R. Soil erosion and the global carbon budget. *Environ Int*. 2003; 29(4):437–450. [PubMed: 12705941]
19. Wang Q, Kim D, Dionysiou DD, Sorial GA, Timberlake D. Sources and remediation for mercury contamination in aquatic systems—a literature review. *Environ Pollut*. 2004; 131(2):323–336. [PubMed: 15234099]
20. Carroll R, Warwick J. Uncertainty analysis of the Carson River mercury transport model. *Ecol Modell*. 2001; 137(2):211–224.
21. Roulet M, Lucotte M, Farella N, Serique G, Coelho H, Passos CS, da Silva EdJ, de Andrade PS, Mergler D, Guimarães J-R. Effects of recent human colonization on the presence of mercury in Amazonian ecosystems. *Water, Air, Soil Pollut*. 1999; 112(3–4):297–313.
22. Dai Z, Feng X, Zhang C, Shang L, Qiu G. Assessment of mercury erosion by surface water in Wanshan mercury mining area. *Environ Res*. 2013; 125:2–11. [PubMed: 23937899]
23. Wang J, Zhang C, Jing Y. Multi-criteria analysis of combined cooling, heating and power systems in different climate zones in China. *Appl Energy*. 2010; 87(4):1247–1259.
24. Wu Q, Wang S, Li G, Liang S, Lin C-J, Wang Y, Cai S, Liu K, Hao J. Temporal Trend and Spatial Distribution of Speciated Atmospheric Mercury Emissions in China During 1978–2014. *Environ Sci Technol*. 2016; 50(24):13428–13435. [PubMed: 27993067]
25. Wang X, Lin C-J, Yuan W, Sommar J, Zhu W, Feng X. Emission-dominated gas exchange of elemental mercury vapor over natural surfaces in China. *Atmos Chem Phys*. 2016; 16(17):11125–11143.
26. Liu M, Zhang W, Wang X, Chen L, Wang H, Luo Y, Zhang H, Shen H, Tong Y, Ou L. Mercury Release to Aquatic Environments from Anthropogenic Sources in China from 2001 to 2012. *Environ Sci Technol*. 2016; 50(15):8169–8177. [PubMed: 27379546]
27. Liu M, Du P, Yu C, He Y, Zhang H, Sun X, Lin H, Luo Y, Xie H, Guo J, Tong Y, Zhang Q, Chen L, Zhang W, Li X, Wang X. Increases of Total Mercury and Methylmercury Releases from Municipal Sewage into Environment in China and Implications. *Environ Sci Technol*. 2018; 52(1):124–134. [PubMed: 29214801]
28. CNEMC Chinese Soil Element Background Value 1990 China National Environmental Monitoring Centre (CNEMC); Beijing, China: 1990
29. Chen H, Teng Y, Lu S, Wang Y, Wang J. Contamination features and health risk of soil heavy metals in China. *Sci Total Environ*. 2015; 512–513:143–153.
30. Cheng H, Li M, Zhao C, Li K, Peng M, Qin A, Cheng X. Overview of trace metals in the urban soil of 31 metropolises in China. *J Geochem Explor*. 2014; 139:31–52.

31. Liu M, Chen L, Wang X, Zhang W, Tong Y, Ou L, Xie H, Shen H, Ye X, Deng C. Mercury Export from Mainland China to Adjacent Seas and Its Influence on the Marine Mercury Balance. *Environ Sci Technol*. 2016; 50(12):6224–6232. [PubMed: 27243109]
32. MWR Criterion of Classification of Soil Erosion Ministry of Water Resources of China (MWR); Beijing, China: 2008
33. Chen L, Meng J, Liang S, Zhang H, Zhang W, Liu M, Tong Y, Wang H, Wang W, Wang X. Trade-induced atmospheric mercury deposition over China and implications for demand-side controls. *Environ Sci Technol*. 2018; 52(4):2036–2045. [PubMed: 29328652]
34. Allesch A, Brunner PH. Material Flow Analysis as a Tool to improve Waste Management Systems: The Case of Austria. *Environ Sci Technol*. 2017; 51(1):540–551. [PubMed: 27936630]
35. Jing, K., Wang, W., Zheng, F. *Soil Erosion and Environment in China* Science Press; Beijing, China: 2005
36. MWR China Water Resources Bulletin Ministry of Water Resources of China (MWR); Beijing, China: 2010
37. Zheng Y, Luo X, Zhang W, Wu X, Zhang J, Han F. Transport mechanisms of soil-bound mercury in the erosion process during rainfall-runoff events. *Environ Pollut*. 2016; 215:10–17. [PubMed: 27176760]
38. Chen L, Zhang W, Zhang Y, Tong Y, Liu M, Wang H, Xie H, Wang X. Historical and future trends in global source-receptor relationships of mercury. *Sci Total Environ*. 2018; 610–611:24–31.
39. Van Oost K, Govers G, Quine TA, Heckrath G, Olesen JE, De Gryze S, Merckx R. Landscape-scale modeling of carbon cycling under the impact of soil redistribution: The role of tillage erosion. *Glob Biogeochem Cycle*. 2005; 19(4)doi: 10.1029/2005GB002471
40. Lindberg S, Zhang H, Gustin M, Vette A, Marsik F, Owens J, Casimir A, Ebinghaus R, Edwards G, Fitzgerald C. Increases in mercury emissions from desert soils in response to rainfall and irrigation. *J Geophys Res-Atmos*. 1999; 104(D17):21879–21888.
41. Song X, Van Heyst B. Volatilization of mercury from soils in response to simulated precipitation. *Atmos Environ*. 2005; 39(39):7494–7505.
42. Gustin MS, Stamenkovic J. Effect of watering and soil moisture on mercury emissions from soils. *Biogeochemistry*. 2005; 76(2):215–232.
43. Powers SM, Bruulsema TW, Burt TP, Chan NI, Elser JJ, Haygarth PM, Howden NJ, Jarvie HP, Lyu Y, Peterson HM, Sharpley AN, Shen J, Worrall F, Zhang F. Long-term accumulation and transport of anthropogenic phosphorus in three river basins. *Nat Geosci*. 2016; 9(5):353–356.
44. Sunderland EM, Mason RP. Human impacts on open ocean mercury concentrations. *Glob Biogeochem Cycle*. 2007; 21(4)doi: 10.1029/2006GB002876
45. Lin J, Pan D, Davis SJ, Zhang Q, He K, Wang C, Streets DG, Wuebbles DJ, Guan D. China's international trade and air pollution in the United States. *Proc Natl Acad Sci U S A*. 2014; 111(5):1736–1741. [PubMed: 24449863]
46. Lin J, Liu Z, Zhang Q, Liu H, Mao J, Zhuang G. Modeling uncertainties for tropospheric nitrogen dioxide columns affecting satellite-based inverse modeling of nitrogen oxides emissions. *Atmos Chem Phys*. 2012; 12(24):12255–12275.
47. Zhang Y, Jacob DJ, Horowitz HM, Chen L, Amos HM, Krabbenhoft DP, Slemr F, St Louis VL, Sunderland EM. Observed decrease in atmospheric mercury explained by global decline in anthropogenic emissions. *Proc Natl Acad Sci U S A*. 2016; 113(3):526–531. [PubMed: 26729866]
48. Liu Q, Liu Y, Zhang M. Mercury and cadmium contamination in traffic soil of Beijing, China. *Bull Environ Contam Toxicol*. 2012; 88(2):154–157. [PubMed: 22037632]
49. Futagami K, Nakajima T. Population aging and economic growth. *J Macroecon*. 2001; 23(1):31–44.
50. Feng X, Qiu G. Mercury pollution in Guizhou, Southwestern China—an overview. *Sci Total Environ*. 2008; 400(1):227–237. [PubMed: 18617222]
51. Obrist D, Agnan Y, Jiskra M, Olson CL, Colegrove DP, Hueber J, Moore CW, Sonke JE, Helmig D. Tundra uptake of atmospheric elemental mercury drives Arctic mercury pollution. *Nature*. 2017; 547(7662):201–204. [PubMed: 28703199]
52. Wang S, Fu B, Piao S, Lü Y, Ciais P, Feng X, Wang Y. Reduced sediment transport in the Yellow River due to anthropogenic changes. *Nat Geosci*. 2016; 9(1):38–41.

53. Jiang Z, Lian Y, Qin X. Rocky desertification in Southwest China: impacts, causes, and restoration. *Earth-Sci Rev.* 2014; 132:1–12.
54. Li X, Wu B, Zhang L. Dynamic monitoring of soil erosion for upper stream of Miyun Reservoir in the last 30 years. *J Mt Sci.* 2013; 10(5):801–811.
55. Li Y, Shao J, Yang H, Bai X. The relations between land use and karst rocky desertification in a typical karst area, China. *Environ Geol.* 2009; 57(3):621–627.
56. Tao S, Fang J, Zhao X, Zhao S, Shen H, Hu H, Tang Z, Wang Z, Guo Q. Rapid loss of lakes on the Mongolian Plateau. *Proc Natl Acad Sci U S A.* 2015; 112(7):2281–2286. [PubMed: 25646423]
57. Reiners PW, Ehlers TA, Mitchell SG, Montgomery DR. Coupled spatial variations in precipitation and long-term erosion rates across the Washington Cascades. *Nature.* 2003; 426(6967):645–647. [PubMed: 14668859]
58. Kosmas C, Danalatos N, Cammeraat LH, Chabart M, Diamantopoulos J, Farand R, Gutierrez L, Jacob A, Marques H, Martinez-Fernandez J. The effect of land use on runoff and soil erosion rates under Mediterranean conditions. *Catena.* 1997; 29(1):45–59.
59. Langbein WB, Schumm SA. Yield of sediment in relation to mean annual precipitation. *Trans, Am Geophys Union.* 1958; 39(6):1076–1084.
60. Wilson L. Variations in mean annual sediment yield as a function of mean annual precipitation. *Am J Sci.* 1973; 273(4):335–349.
61. Zhang XC, Nearing MA. Impact of climate change on soil erosion, runoff, and wheat productivity in central Oklahoma. *Catena.* 2005; 61(2):185–195.
62. Eckley CS, Branfireun B. Mercury mobilization in urban stormwater runoff. *Sci Total Environ.* 2008; 403(1–3):164–177. [PubMed: 18582918]
63. Kim Y, Wang G. Soil moisture-vegetation-precipitation feedback over North America: Its sensitivity to soil moisture climatology. *J Geophys Res–Atmos.* 2012; 117(D18)doi: 10.1029/2012JD017584
64. Koster RD, Dirmeyer PA, Guo Z, Bonan G, Chan E, Cox P, Gordon C, Kanae S, Kowalczyk E, Lawrence D. Regions of strong coupling between soil moisture and precipitation. *Science.* 2004; 305(5687):1138–1140. [PubMed: 15326351]
65. Savabi MR, Stockle CO. Modeling the possible impact of increased CO₂ and temperature on soil water balance, crop yield and soil erosion. *Environ Modell Softw.* 2001; 16(7):631–640.
66. Seneviratne SI, Corti T, Davin EL, Hirschi M, Jaeger EB, Lehner I, Orlowsky B, Teuling AJ. Investigating soil moisture–climate interactions in a changing climate: A review. *Earth-Sci Rev.* 2010; 99(3):125–161.
67. Mayor JR, Sanders NJ, Classen AT, Bardgett RD, Clément J-C, Fajardo A, Lavorel S, Sundqvist MK, Bahn M, Chisholm C. Elevation alters ecosystem properties across temperate treelines globally. *Nature.* 2017; 542(7639):91–95. [PubMed: 28117440]
68. Lin Y, Wang S, Wu Q, Larssen T. Material flow for the intentional use of mercury in China. *Environ Sci Technol.* 2016; 50(5):2337–2344. [PubMed: 26812394]
69. Xie Z, Zhu J, Liu G, Cadisch G, Hasegawa T, Chen C, Sun H, Tang H, Zeng Q. Soil organic carbon stocks in China and changes from 1980s to 2000s. *Glob Change Biol.* 2007; 13(9):1989–2007.
70. Jiskra M, Sonke JE, Obrist D, Bieser J, Ebinghaus R, Myhre CL, Pfaffhuber KA, Wängberg I, Kyllönen K, Worthy D. A vegetation control on seasonal variations in global atmospheric mercury concentrations. *Nat Geosci.* 2018; 11:244–250.
71. Yang S, Milliman J, Xu K, Deng B, Zhang X, Luo X. Downstream sedimentary and geomorphic impacts of the Three Gorges Dam on the Yangtze River. *Earth-Sci Rev.* 2014; 138:469–486.
72. Maavara T, Parsons CT, Ridenour C, Stojanovic S, Dürr HH, Powley HR, Van Cappellen P. Global phosphorus retention by river damming. *Proc Natl Acad Sci U S A.* 2015; 112(51):15603–15608. [PubMed: 26644553]

Figure 1.

Surface soil (0 to 20 cm) THg concentration of each province in China for ~20 years. Part a is the comparison of surface soil THg concentrations between around 1990 and around 2010. Parts b and c are the relationships of population densities and surface soil THg concentrations around 1990 and around 2010, respectively. The sizes of the dots in part a represent different areas of the provinces. Shaded areas in parts b and c are the 95% confidence intervals.

Figure 2.

Soil THg removal induced by water erosion in China. Parts a and b are the distributions of soil THg removal rates induced by water erosion at county levels in China around 1990 and around 2010, respectively. Part c is the distribution of changes of THg removal rates in ~20 years in China. Part d shows the changes in the masses of water-induced soil erosion, average soil THg concentrations and Hg removal rates in ~20 years in four typical regions in China. In part a, 1 to 4 are the upstream of the Yangtze River (Changjiang), the upstream of the Yellow River (Huanghe), Xi River (Xijiang), and north part of Hai River (Haihe) basins, respectively. Four Chinese adjacent seas are not included in the maps. In parts a, b, and c, the yellow and red contour lines represent the areas of the four basins in part d.

Figure 3.

THg transport and emission induced by water erosion in China. Part a shows the amount of erosion-induced THg release into the aquatic environment in secondary river basins around 2010 in China. Part b shows comparisons of the amounts of THg released into aquatic environments from erosion-induced processes (including background and legacy anthropogenic sources) and direct anthropogenic sources in the main secondary river basins around 2010 in China. Part c is the distribution of Hg(0) emission from soil induced by water erosion in the erosional areas at the county level around 2010 in China. In part b, the THg release data from direct anthropogenic sources are from our previous study.²⁶ Detailed information on the secondary river basins in China is provided in SI Figure S4.

Figure 4. Relationship of erosion-induced THg release into the aquatic environment with precipitation (a), elevation change (b), population density (c), and surface air temperature (d) around 2010 in China. The sizes of the dots represent the amount of precipitation. Shaded areas are the 95% confidence intervals.

Figure 5.

Hg budget in environmental media in Mainland China in 2010. Gray arrows are fluxes referenced from previous studies, blue arrows are from this study, and dotted arrows are approximate estimations in this study. Note: (a) net exchange of Hg(0) between the atmosphere and natural soil of Mainland China from Wang et al. (2016);²⁵ but without the emission induced by water erosion; (b) direct anthropogenic THg emission in China from Wu et al. (2016);²⁴ (c) net exchange of Hg(0) between the atmosphere and total water body (including rivers, lakes, and reservoirs) from Wang et al. (2016);²⁵ (d) direct anthropogenic release into the aquatic environment in China from Liu et al. (2016);²⁶ (e) Hg(0) emission from the erosional area induced by water erosion; (f) total amount of soil THg eroded by water erosion in China; (g) net exchange of atmospheric THg of Mainland China with other regions (including Chinese adjacent seas); and (h) net exchange of Hg(0) between atmosphere and vegetation in Mainland China cited from Wang et al. (2016).²⁵ All the fluxes for natural processes include background and legacy anthropogenic sources.