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Rapid Increase in the Lateral Transport of Trace Elements Induced by Soil Erosion in Major Karst Regions in China

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Abstract

Soil erosion, which has been recently shown to significantly perturb carbon cycling, occurs naturally but can be either enhanced or reduced by human activities. However, the impacts of soil erosion on terrestrial contaminant cycles remain unclear. Here, we select eight trace elements, i.e., arsenic, cadmium, chromium, copper, nickel, lead, zinc, and mercury, to examine the erosional impacts of the elements' fate and transport across China. By synthesizing the detailed distribution of soil erosion fluxes, soil element inventories, and diverse modeling methods, we reveal that while human activities have reduced the lateral transport of these elements in the Loess Plateau (Central North China, a 56% decline in the past two decades with a range of 46% to 110%) due to soil conservation projects, they have increased these transport fluxes in China's major karst regions (Southwest China, a 84% increase with a range of 55% to 150%) because of severe rocky desertification. These fluxes have completely overwhelmed the soil conservation efforts in the Loess Plateau. Fluxes of these elements into aquatic environments from Southwest China reached 46% of the total input in China in 2010. These fluxes were higher than the inputs from point sources in the region by a factor of 50 because of impacts of excessive agricultural cultivation and geographical and climatic factors. These findings indicate the enormous perturbation of terrestrial

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ASSOCIATED CONTENT

Supporting Information

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Distribution of soil erosion area; changes of surface soil concentrations of trace elements; releases of trace elements from direct anthropogenic sources into aquatic environment; relationships between sediment yield and major driven factors; annual precipitation, population density, crop yield; topographical slope change; fitting errors of path coefficients (PDF)

Volume of industrial wastewater and municipal sewage discharges; concentrations of trace elements; and databases of trace element transports into streams induced by soil erosion (XLSX)

Notes

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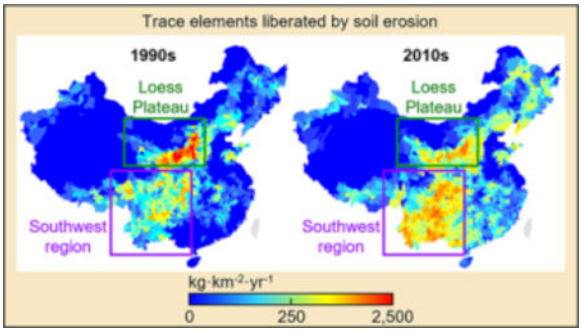
contaminant cycles caused by soil erosion in karst regions and demonstrate the need for long-term sustainable management of soil erosion and contaminant discharge to protect fragile terrestrial ecosystems.

Graphical Abstract



INTRODUCTION

The earth's terrestrial ecosystems are undergoing rapid changes as a result of the changing climate and anthropogenic perturbation.[1,2] Terrestrial ecosystems are the foundations of healthy soil and their surrounding aquatic environments and thus have substantial impacts on material cycles, such as the carbon cycle.[1,3] Recent studies have indicated that cropland expansion has driven an overall increase in global soil erosion, resulting in significant perturbations in global carbon cycling.[1,4,5] Terrestrial ecosystems have also received large quantities of contaminants during the industrial era.[6,7] Increasing experimental evidence has suggested that soil erosion could have a significant impact on trace element cycles in small basins, such as increases of particulate and dissolved element concentrations in the water column and enrichment of them in sediment.[8–10] A previous study has highlighted that soil erosion could significantly induce the lateral transport of soil contaminants and contribute to the aquatic environment and their biogeochemical contaminant cycles globally,[11] but the regional distribution and the detailed process of the transport are not well understood. Our previous study also suggested that soil erosion could induce substantial terrestrial mercury (Hg) transport in China,[12] but the impact is still unknown for other contaminants. Hence, quantitative descriptions of the lateral transport of contaminants, such as trace elements, in terrestrial ecosystems are urgently needed since these processes have the potential to accelerate accumulation of contaminants in aquatic systems and into the human food web.

Karst rocky desertification is a process that transforms highly sensitive and vulnerable karst areas covered by vegetation and soil into rocky landscapes. This process has occurred in the Mediterranean basin, the Dinaric Karst, and Southwest China (including Chongqing, Guangxi, Guizhou, Sichuan, and Yunnan Provinces).[13] The karst ecosystem in Southwest China (Figures 1 and 2, Figure S1 in Supporting Information, SI) is one of the largest exposed carbonate rock areas in the world with an area of more than 5.4×10^5 km^2.[14] The human exploitation of natural resources in this region has resulted in a large-scale increase in rocky desertification (1.3×10^5 km^2 in recent years) which increased by a factor of 4 from 1970 to 2005.[13,14] The concentrations of most of the soil trace elements in Southwest China

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are higher than those in other regions in China and the global average (Figure 3a,b, Figure S2 in SI), due to the higher geochemical background of these elements and extensive and continued mining activity in Southwest China.[15] Consequently, substantial amounts of trace metals and other contaminants may be liberated from soils in Southwest China and transported laterally, as is the case with carbon and Hg.[12,16] Hence, rocky desertification is labeled as the primary ecological disaster in Southwest China. Quantification of the changes in lateral contaminant transport induced by soil erosion in this region through time is needed, and an understanding of the potential driving factors is important and necessary for an effective remediation of this process.

To understand the impacts of soil erosion on terrestrial element cycles, we first quantitatively described how the lateral transport of different trace elements through terrestrial ecosystems is induced by soil erosion and then determined the elemental contributions to the aquatic environments in different regions across China, which has not been investigated in detail elsewhere. The impacts of eight trace elements, i.e., arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), nickel (Ni), lead (Pb), zinc (Zn), and Hg, were quantitatively described as their mobilization is potentially harmful to humans. The consumption of drinking water and crops contaminated by As has led to widespread death and disease in some Asian regions, for example, Bangladesh, Mainland China, Taiwan, as well as some regions in the U.S.[17] Cd is an inorganic toxicant of great environmental concern and has been classified as a human carcinogen.[18] Pb and Hg are cumulative toxicants that damage multiple body functions, especially neurologic and cardiovascular systems and the kidneys.[19,20] The exposure to Pb and Hg is dominantly through food consumption.[21] Previous studies have suggested that low ingestion of Cr, Cu, Ni, and Zn may be necessary as essential trace elements by mammals, but at high exposures, they can significantly produce adverse health consequences, for example, kidney and liver damage and severe neurological defects.[22–25] Among these elements, As, Cd, Pb, and Hg have been labeled as four of the top ten chemicals of public health concern by the World Health Organization (WHO, web site: www.who.int). An accurate assessment of the erosion-induced impacts of soil trace elements at broad spatial scales should therefore be obtained based on extensive elemental measurements and detailed field surveys of soil erosion mechanisms. Anthropogenic point source releases (includes industrial wastewater and municipal sewage) might be important sources of these trace elements to the aquatic environment in China, such as Hg.[26,27] However, besides Hg, the relative importance of soil erosion compared with point source releases of these elements to the aquatic environment has remained unknown. Therefore, to obtain this information, we quantified the amounts of the seven elements besides Hg released from different point sources in China and compared them with those from releases from soil erosion. We further quantified the potential driving factors, including geographical, climatic, and anthropogenic perturbations, for the lateral transport of these elements in Southwest China. Overall, we have three related reasons for focusing on Southwest China: (1) This region is a good “natural laboratory” because of its severe soil erosion (Figures 1 and 2).[13,16] (2) Several major Asian rivers (e.g., the Yangtze, Mekong, Pearl, Salween, and Song Hong Rivers, Figure 1) flow through the region and bring substantial fluxes of terrestrial trace elements to the adjacent seas.[28,29] (3) Detailed data sets on soil erosion and trace element concentrations

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are available in China, providing a unique opportunity to examine impacts, which are in turn linked with global water and food security.

MATERIALS AND METHODS

Soil Trace Elements Liberated by Soil Erosion.

The lateral transport of trace elements induced by soil erosion is calculated based on two national surveys on soil erosion (carried out in 1995−1996 and 2010−2012) and two national soil databases of element concentrations (established in the 1990s and the early 2010s). The two national surveys of soil erosion were conducted using remote sensing images and field survey observation, with inputs of topographical, land use, vegetation cover, and meteorology information, to provide the spatial distributions of soil erosion for a total of 2359 counties in China. The two national soil databases of elemental composition were established based on samples and measures made by national monitoring institutions and major scientific research institutions in China. For the two soil pollution databases, soil samples were collected from approximately 4000 and 38000 locations, respectively, for all types of soils, reflecting different land use patterns (such as urban areas, cropland, and unused land but excluding landfill). The surface soil elemental concentration data (0−20 cm) from these databases were used because soil erosion occurs mainly in this layer.[16] Details of the databases have been provided in previous studies.[12,16]

Following other studies,[12,16] the erosional components of soil trace elements were derived as presented below

$$E_{ij}(x) = \sum_{kl}\left(C_{ik}(x) \times M_l \times A_{kl} \times K\right)$$

where $E_{ij}(x)$ is the probabilistic distribution of the flux of eroded element i (Gg·yr^{-1}) in each province j, and $C_{ik}(x)$ is the probabilistic distribution of the element i concentrations ($\mu g \cdot g^{-1}$) in county k. As described above, we selected As, Cd, Cr, Cu, Ni, Pb, and Zn from the database to quantify the impacts of soil erosion. Data for Hg were referenced from another study.[12] We used the ordinary kriging (spatial optimal linear prediction) method to interpolate the spatial distributions of the element concentrations for each county, and the calculations were accomplished using ArcGIS version 10.3. The kriging interpolation is a modeling method that provides a spatial estimate based on variogram function and structure analysis and has long been used in geology, ecology, and environmental sciences by interpolating between data collected from different locations.[30] The standard errors of the interpolation results (<20%) were considered in the uncertainty analysis. In eq 1, M_l is the erosion modulus, i.e., the mass of soil erosion (kg·km^{-2}·yr^{-1}) for erosion grade l, following the Standards for Classification of Soil Erosion (SL190−2007).[16] A_{kl} is the soil erosion area (km2) of grade l in county k. A previous study divided soil erosion areas into five grades based on the standards.[16] Finally, K is the unit conversion factor for the elements (10^{-9}).

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Erosion-Induced Trace Element Releases into Aquatic Environments.

We quantified the amounts of trace elements released into streams induced by soil erosion based on mass balance principals, as shown in the following equation[27]

$$S_{ij}(x) = \sum_{kl}\left(E_{ij}(x) \times \mathrm{SDR}_l\right) \quad 2$$

where $S_{ij}(x)$ is the probabilistic distribution of element i released into streams ($Gg \cdot yr^{-1}$) of province *j*; SDR_l is the sediment delivery ratio (%) of erosion grade *l*, which is defined as the mass of the sediment input into local streams to the mass of soil eroded along the catchment hillslope, which has been discussed in other studies.[16] Different elements mobilized by the erosion probably have different mobilization potentials under the influence of particle size as they may be preferentially associated with different size fractions. Following previous studies,[11,12,16] we estimated amounts of transports of these elements by multiplying their bulk concentrations in soils with the flux of soil erosion, due to the lack of detailed information on particle size. The differential influence of organic carbon on transport of these elements is also not quantified, and while this may vary over time, data suggest that the content of organic carbon in surface soil in China in the last three decades has remained relatively constant.[31]

Point-Source Releases of Trace Elements into Aquatic Environments.

We applied a model that was developed in our previous studies to estimate the direct release of the seven elements (excluding Hg) from anthropogenic sources, including industrial wastewater and municipal sewage, into aquatic environments in China from 2001 to 2015 (Figure S3 in SI).[26,27] The element releases from various industrial sectors (e.g., mining activities, coal-fired power plants, and nonferrous metal production) were estimated based on the volume of wastewater discharge multiplied by the trace element concentrations.[26] The concentrations refer to end-of-pipe concentration data following wastewater treatment.[26] The element releases from municipal sewage were divided into treated and untreated sewage discharges.[27] We parametrized the model through Monte Carlo simulations. Details on the model and data on Hg were included in previous studies.[26,27]

Statistical Analyses and Structural Equation Modeling.

No statistical methods were used to predetermine the sample size in this study. All statistical analyses presented in this study were conducted in R version 3.3.3 (R Project for Statistical Analysis). Results were considered significant at $P < 0.05$* and <0.01**.

We applied a structural equation model (SEM, lavaan Project) framework to quantitatively describe different direct and indirect hypotheses of driving factors associated with erosion-induced sediment yields to streams in Southwest China for the 1990s and the early 2010s, respectively.[2,32] SEM is a popular framework for formulating, fitting, and testing data and has been extensively used in measurement and hypothesis testing.[33] The development of SEM has been seen in social science fields, such as psychology and sociology,[34] while recently, SEM has been gradually applied in other fields, such as ecology and biology.[2,35]

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Many direct and indirect pathways contribute to the sediment yield to streams from catchments. On the basis of the literature,[13,36–39] we selected population density, elevation, slope, annual precipitation, and crop yield density in each county in the two periods for the models (sample size, $n = 481$) based on testing of their significance (Figure S4 in SI). We did not consider industrial development in the model as Southwest China is a traditional agricultural region and the relationship between sediment yield and the number of factories in this region is nonsignificant ($R^2 = 0.03$, $P = 0.26$).

We formulated a path model based on the relationships between sediment yield and the potential driving factors. A resampling approach based on clustered bootstrapping was performed to infer the total effects.[2] A maximum likelihood method was applied to estimate the missing data in the two models. Following a previous study,[2] we used an overall test of model fit adjusted for clustering effects to examine the fitting of the two models. An omnibus test was used to test the hypotheses in this study, and the results indicated that the hypotheses for the two models were significant and could not be rejected (1990s: $\chi^2 = 1532$, $P = 0.392$, d.f. = 9 and early 2010s: $\chi^2 = 927$, $P = 0.064$, d.f. = 9). Additional model fit statistics, including the root-mean-square error of approximation (RMSEA = 0.000 and 0.071), standardized root-mean-square residual (SRMR = 0.001 and 0.004), and comparative fit index (CFI = 1.000 and 0.997), also indicated that the present framework for the two periods was reasonable.[2] Standardized path coefficients (ranging from −1 to 1) were computed and applied to interpret the different direct and indirect influences on the independent variables. The overall fitting errors of the path coefficients of the two models range from 0.014 to 0.054 and 0.009 to 0.081, respectively.

Databases Used in the Present Study.

The two soil erosion surveys in China (Figure S1 in SI) were carried out by the Ministry of Water Resources of China.[16] The two national soil databases of trace elements in the 1990s and the early 2010s (Figure S2 in SI) were collected from the China National Environmental Monitoring Centre (http://www.cnemc.cn/) and the China Geological Survey Bureau (http://www.cgs.gov.cn/). Data for the industrial wastewater and municipal sewage discharges in each province in China were collected from the *China Environmental Statistical Yearbook* (http://data.cnki.net/). Data for the concentrations of the seven elements (excluding Hg) in wastewater were derived from published literature and are included in the Supporting Information. Data sets of detailed population data (Figure S5 in SI) were collected from two tabulations of population censuses in 1995 and 2010 in China (http://data.cnki.net/). Crop yield data for each county (Figure S5 in SI) for the two periods were collected from the China Statistical Yearbook (*County-Level*) (http://data.cnki.net/). Data sets of digital elevation models (Figure 1) in different regions (Landsat ASTER at a resolution of 30 m) were provided by the Geospatial Data Cloud site, Computer Network Information Center, Chinese Academy of Sciences (http://www.gscloud.cn). We applied the seamless mosaic tool in the Environment for Visualizing Images (ENVI version 5.3) software to join different digital elevation model data sets together. We applied the spatial analyst tool in ArcGIS version 10.3 to calculate the slope change (Figure S6 in SI) based on the digital elevation model. Annual precipitation data (Figure S5 in SI) were provided by the National Meteorological Administration of China (http://www.cma.gov.cn/).

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Uncertainty Analysis.

We applied a Monte Carlo method (performed for 10000 runs) to simulate the probabilistic distributions of all the results.[26] The median values and the 60% confidence intervals (ranging from 20% to 80%) of the statistical distributions were modeled to quantify the element fluxes and to characterize the uncertainty.[29] The concentrations of these soil elements followed log-normal distributions. Details of parameter settings are provided above. For the two SEMs, we provided all the fitting errors of the path coefficients of the two models in Figure S7 in SI.

RESULTS

Trace Elements Stored in Soil and Liberated by Soil Erosion across China.

Our analysis revealed that vast quantities of the eight trace elements were stored in the surface soil layer (<20 cm) in China. In total, the amount was up to 3.6×10^5 Gg (with a range of 2.5×10^5 to 5.3×10^5 Gg using a 60% confidence interval based on a Monte Carlo simulation) in the early 2010s, 11% (0.85% to 22%) higher than that in the 1990s. This is an evident anthropogenic signal.[40] The amounts of Cd and Hg stored in the surface soil layer rapidly increased in the 1990−2010 period by 78% (52% to 120%) and 50% (32% to 94%), respectively. These changes are associated with the rapid increase in anthropogenic atmospheric emissions of all these eight elements in recent years.[40,41] According to a previous study, in total, 61 $Gg \cdot yr^{-1}$ of these eight elements were emitted from anthropogenic sources to the atmosphere in China in 2012 and had rapidly increased from 19 $Gg \cdot yr^{-1}$ in 1980.[40] We assume that the atmospheric deposition of these eight elements are also increasing in concert, but there is a lack of observation data. Historical records of Cd and Hg accumulations in sediments in some watersheds and estuaries partially verify the magnitude of these increases.[42–45] We also verified that substantial fluxes of trace elements could be transported laterally from soils (Figure 4). In total, 1200 $Gg \cdot yr^{-1}$ (870 to 1800 $Gg \cdot yr^{-1}$) of the eight elements were liberated from soils and transported laterally in China in the early 2010s (Figure 3c).

Releases of Trace Elements from Soil Erosion and Point Sources into Streams across China.

Water pollution induced by trace elements is often associated with anthropogenic point source releases. Here, we found that the contributions of trace elements released from soil erosion to aquatic environment pollution are larger than those from point sources in China. On the one hand, 42 Gg (27−62 Gg) of these eight elements were released from point sources, including industrial wastewater (accounted for 67%) and municipal sewage (accounted for 33%), into aquatic environments in China in 2015, slightly lower than the 45 Gg (25−71 Gg) released in 2001 (Figure S3, SI). Considering individual elements, the amounts of Cr, Ni, Pb, and Hg releases from point sources decreased from 2001 to 2015,[27] while those of As and Cd increased and those of Cu and Zn had no significant change. On the other hand, erosion liberates substantial amounts of soil trace elements and transports these elements horizontally. These elements can be subsequently redeposited or transported into streams. In total, soil erosion released 660 $Gg \cdot yr^{-1}$ (440−950 $Gg \cdot yr^{-1}$) of the eight elements into streams in all of China in the early 2010s (Figure 3d), while 540 $Gg \cdot yr^{-1}$

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(320−910 Gg·yr^{-1}) of the elements were redeposited onto land surfaces. The fluxes of these elements were substantially higher than the direct anthropogenic inputs into streams by a factor of 15 (mass-weighted average, Figure 3e).[26,27,46] The total flux of the releases of the eight elements into streams induced by soil erosion in China in the early 2010s was 4.2% (3.5% to 8.3%) higher than that in the 1990s, and the most rapid increase, 20-fold higher than that experienced by the whole country, was found in Southwest China (Figure 3f).

Rapid Increase in the Lateral Transport of Trace Elements in Southwest China.

Our calculations revealed that the karst region in Southwest China, rather than the Loess Plateau in the Yellow River Basin,[47] is the primary hotspot for trace element transport into streams induced by soil erosion in China today (Figure 4) and that such transport rapidly increased in the past two decades. The total flux (220 kg·km^{-2} yr^{-1} with a range of 130−350 kg·km^{-2}·yr^{-1}) of the eight elements in Southwest China was 2 times greater than that in the Loess Plateau in the early 2010s and 71% lower than that in the Loess Plateau in the 1990s. The sharp decline of the flux in the Loess Plateau in the past two decades (56% with a range of 46% to 110%) can be attributed to the implementation of the “returning farmland to forests and grassland” soil conservation initiative that began in Northern China in the 1990s.[47] The rapid increase in this flux in Southwest China (a 86% increase with a range of 55% to 150%) is in accordance with the historical records of some of these elements in downstream areas.[42–44] For example, according to previous experimental results, influxes of As, Cd, and Hg to the sediment in the north margin of the South China Sea (Figure 1) increased approximately 1- to 2-fold in the past four decades.[43,44] This is similar to the observation from the Woburn Experimental Farm in the U.K., which showed that soil erosion significantly increased the enrichment of Cr, Cu, Ni, and Pb in the sediment.[9] Here, we explain that the increase was mainly caused by severe rocky desertification induced by excessive agricultural cultivation in karst landscapes.[13]

Southwest China, which covers 14% of the territory of China, contributed 46% of the total flux of the eight elements into streams induced by soil erosion. We found that in the early 2010s, erosion in Southwest China transported 150 (92− 230), 69 (43−110), 36 (22−57), 23 (14−37), and 10 (6.2−Gg·yr^{-1} of the eight elements into the upstream reaches of the Yangtze, Pearl Rivers, Mekong, Song Hong, and Salween River basins, respectively. We calculated that in Southwest China, increases in soil erosion contributed 65% to 80% of the increases in the lateral transport of As, Cr, Cu, Ni, Pb, and Zn into streams in the past two decades (Figure 3f,g). The rise in the concentrations of Cd and Hg caused by anthropogenic atmospheric emission and deposition in local catchments contributed 66% and 60% to their total fluxes.

Impacts of Climate, Topography, and Human Activity on Soil Erosion across Southwest China.

To further understand the potential factors driving soil trace element transport in Southwest China and to quantitatively describe direct and indirect effects of changes in erosion-induced sediment yield in the region in the 1990s and early 2010s, a SEM framework was applied.[2] The best-fitting model revealed that although rocky desertification induced by agricultural cultivation was taking place in the 1990s, the sediment yield in the region was influenced by

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geographical factors more than agricultural cultivation (Figure 5). Among the four direct impact factors, elevation change most affected the sediment yield (contributing 42% to the total change in sediment yield) in this period, followed by agricultural cultivation (31%). The SEM highlights the negative relationship between sediment yield and regional precipitation in Southwest China. This relationship suggested that increases in precipitation can alter erosional activity in this region, a finding that may potentially be attributed to increasing vegetative cover,[37,48] which is crucial for the increase in soil moisture.[49] The negative impacts of climate change on precipitation in Chinese karst ecosystems have already been identified,[50] and these impacts could reduce soil moisture content and make the soil surface more sensitive to subsequent precipitation events.[50–52]

The SEM suggests that in the early 2010s, agricultural cultivation was the dominant driver (50%) rather than geographical factors (25% for the impact of slope changes and 9.0% for elevation change) or climatic factors (16% for precipitation). Although an increase in landscape slope may be unsuitable for crop growth, the SEM (Figure 5) suggested that in the early 2010s, agricultural areas may have already expanded into more hillside areas. The negative consequences of excessive agricultural cultivation in unsuitable locations on soil erosion and subsequent lateral trace element transport must therefore be further characterized in Southwest China. A recent paper found that the leaf area index (a proxy for vegetation cover) has been increasing in Guangxi, Guizhou, and Yunnan Provinces in recent years due to “the rocky desertification treatment project” in China that ran from 2008 to 2015.[14,53] It would be premature to conclude that the lateral transport of trace elements induced by soil erosion in Southwest China has reached a turning point, as suggested by that paper, since agricultural cultivation in this region is still increasing thus countering the impact of remediation projects, and the influence of the changing climate is a large uncertainty but is likely enhancing erosion in the region.[50]

DISCUSSION

Our analysis closes a knowledge gap in terms of metal(loid) cycling in terrestrial environments as to whether lateral contaminant transport induced by soil erosion can cause large and significant perturbations and fluxes to inland aquatic environments. Rivers are dynamic conduits connecting the terrestrial and oceanic contaminant cycles.[28] Biogeochemical studies of contaminants in watersheds and landscapes commonly focus on direct anthropogenic inputs (point sources, including industrial wastewater and municipal sewage discharges) into freshwater ecosystems and the quantification of riverine fluxes.[3,28,54] Our analysis provides new evidence that not only Hg, as has been illustrated previously,[12] but also other trace elements released to streams induced by soil erosion could cause negative perturbations to the aquatic environment. Hence, this study is critical in presenting a better understanding of the global biogeochemical cycles of trace elements and highlights the potential importance of the release of legacy anthropogenic contaminants presently stored in some of the earth’s critical zones, e.g., the terrestrial ecosystem.[3,55] We have demonstrated here that humans have reduced the lateral transport of contaminants to the aquatic environment through large-scale environmental restoration projects, such as those in the Loess Plateau, but have also increased these fluxes through anthropogenic perturbations, such as the excessive agricultural cultivation present in Southwest China.

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We acknowledge that there are potential biases in our analysis, such as the mismatch of time periods between soil contaminant concentration measurements and soil erosion inventories from different national surveys, which might increase the uncertainties of the results.[12] Additionally, soil properties, such as particle size, might have different influences on the fate and transport of the different contaminants, and this was not considered in the present study. Finally, although the high contaminant content of soil in Southwest China (Figure 3g) measured in the last two decades could be attributed to the high geochemical background values for these elements and/or to the continued mining activity, as mentioned above, we propose that erosion is the most important factor. However, it is clearly difficult to evaluate which factor is more important, and further study on the various potential sources is desirable. The overall correlation coefficients of the two models are not high (Figure 5), which may result partly from uncertainties in the census survey data,[16,56] interactions between different driving factors,[57] and the contributions of other factors such as mining activity and regional climate change.[47] Despite the uncertainty, the impact of excessive agricultural cultivation on soil erosional transport of trace elements can be characterized.

Our findings on the erosion-induced lateral transport of contaminants in Southwest China can partly verify the legacy hypothesis presented in previous studies,[3,55] which suggests that the catchment contaminant balance may be in a legacy depletion stage and this may last for a long time. Although the amounts of some of the contaminants released into the aquatic environment in Southwest China have been decreasing in recent years, rapid increases of lateral contaminant fluxes into streams, due to enhanced anthropogenic perturbations in the past two decades, have completely overwhelmed governmental efforts to lower anthropogenic contaminant releases, primarily from point sources. Hence, we suggest that long-term perspectives and sustainable management plans, such as those for phosphorus,[55,58] are needed for other soil contaminants in the construction of a framework to protect fragile terrestrial ecosystems and drive forward the science on pollution from legacy sources.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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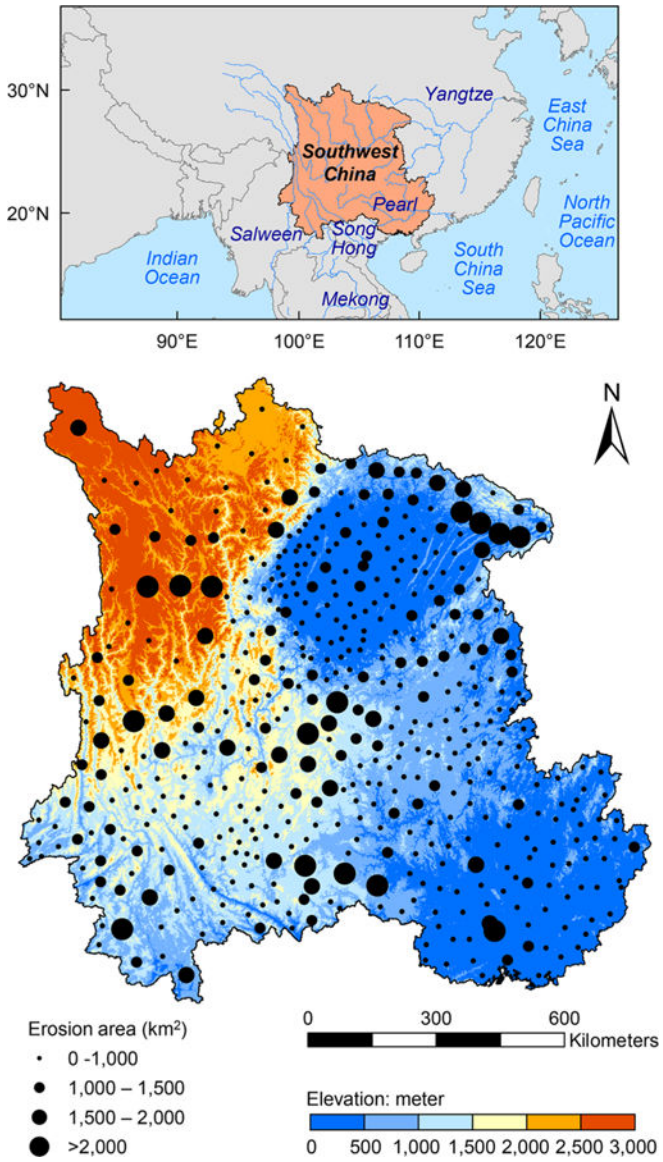

Figure 1.

Location and elevation of Southwest China, and soil erosion in this region in the early 2010s.

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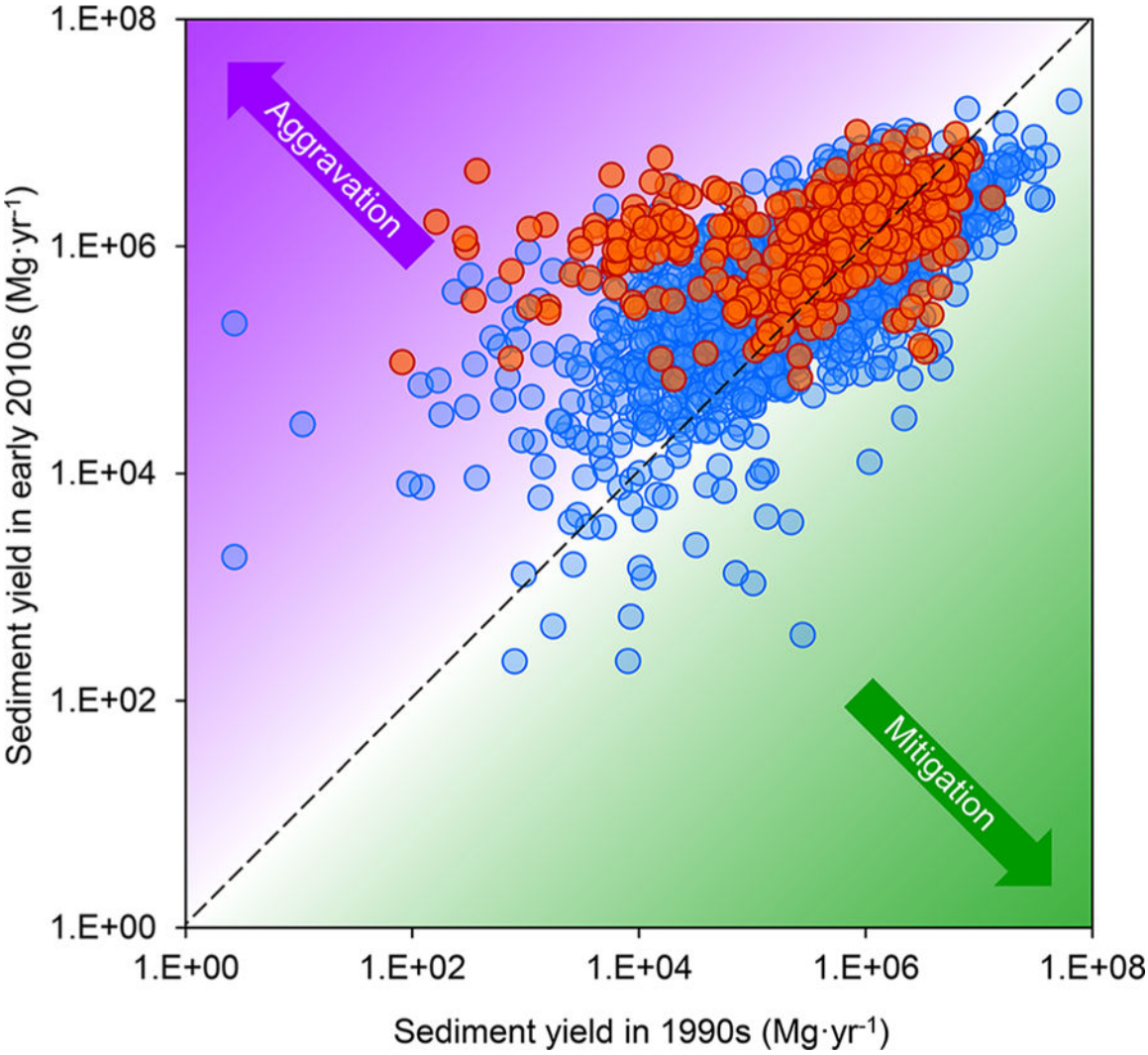

Figure 2.

Comparison of sediment yields induced by soil erosion in China in the 1990s and the early 2010s. Red dots represent counties in Southwest China, and blue dots represent counties in other regions in China. Aggravation and mitigation in the figure: aggravation or mitigation of erosion-induced sediment yields from 1990s to early 2010s, respectively.

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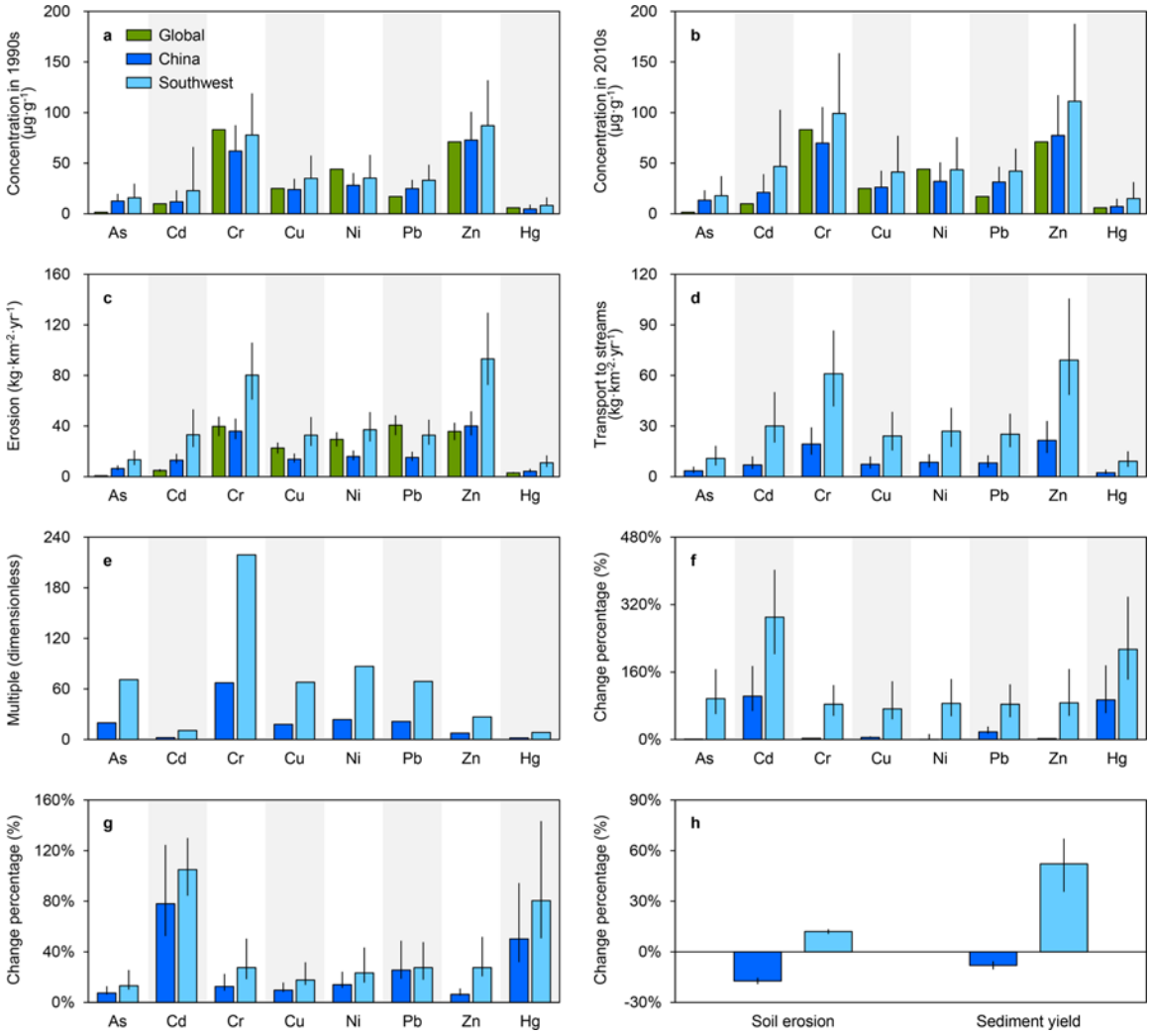

Figure 3.

Comparison of trace elements liberated by soil erosion and the subsequent lateral transport into streams for all of China and in Southwest China between the 1990s and the early 2010s. (a) Concentrations of trace elements in 1990s. (b) Concentrations of trace elements in the early 2010s. (c) Comparison of the fluxes of trace elements liberated by soil erosion in the early 2010s. (d) Comparison of the fluxes of the lateral transport into streams in the early 2010s. (e) Multiple of the fluxes of the lateral transport higher than their direct anthropogenic releases (point sources) into streams in the early 2010s. (f) Changes in the fluxes of the lateral transport into streams over the past two decades. (g) Changes in trace element concentrations in the past two decades. (h) Changes in soil removal and sediment yields in the past two decades. In parts a and b, the units for Cd and Hg are $10^{-2} \times \mu g \cdot g^{-1}$. In parts c and d, the units for Cd and Hg are $10^{-2} \times kg \cdot km^{-2} \cdot yr^{-1}$. The global data and data of Hg are from previous studies.[11,12,26,27]

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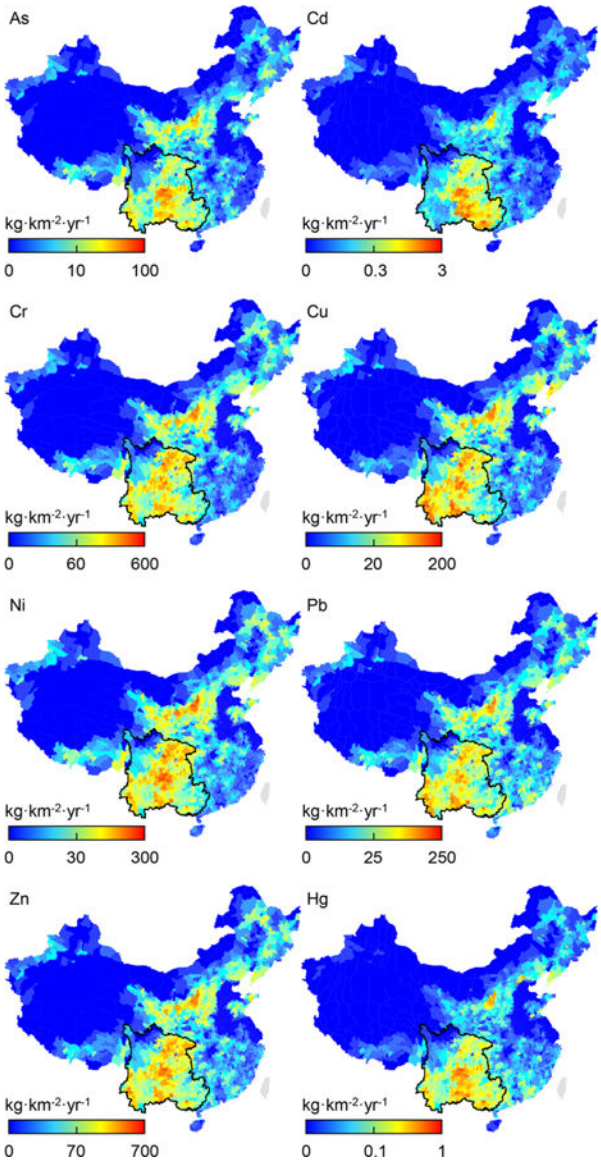

Figure 4.

Distribution of trace elements liberated by soil erosion in China in the early 2010s. Taiwan, Hong Kong, and Macao are not included in the calculations

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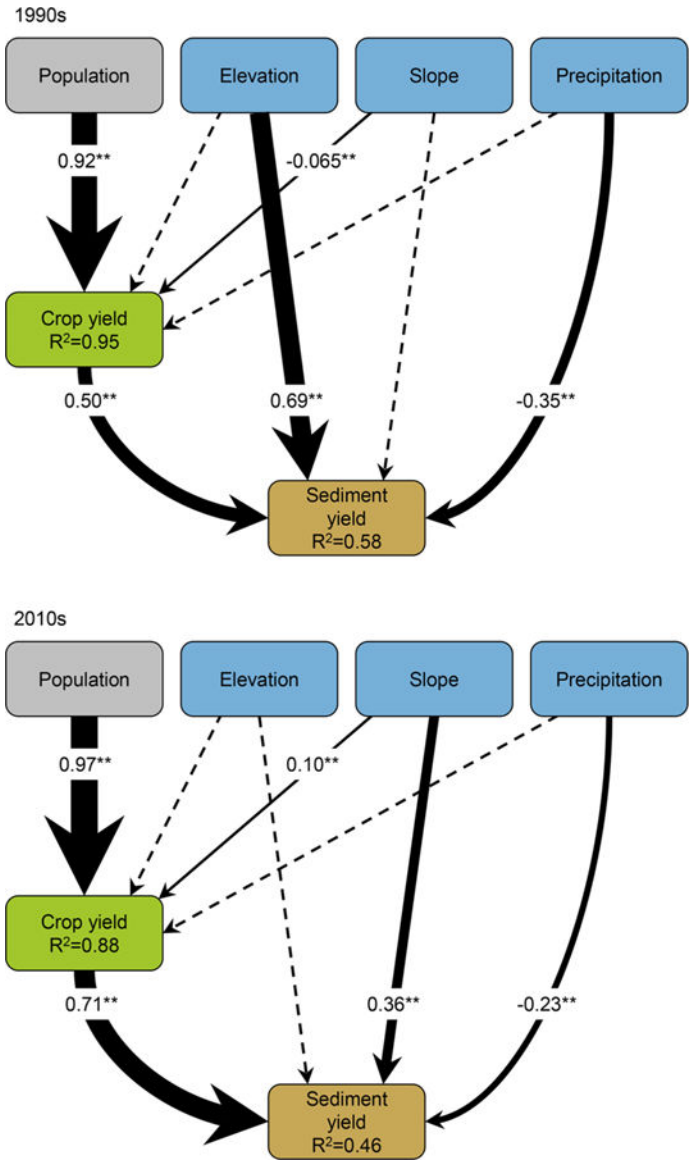

Figure 5.

Best-supported SEM for the major direct and indirect pathways to sediment yield induced by soil erosion in Southwest China in the 1990s and early 2010s. The solid arrows indicate significance at * $P < 0.05$ or ** $P < 0.01$, and dashed arrows indicate nonsignificant coefficient estimates. The numbers in the two figures are standardized path coefficients.