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# Transport mechanisms of soil-bound mercury in the erosion process during rainfall-runoff events<sup>☆</sup>

Yi Zheng<sup>a, b, \*</sup>, Xiaolin Luo<sup>b</sup>, Wei Zhang<sup>c</sup>, Xin Wu<sup>b</sup>, Juan Zhang<sup>a</sup>, Feng Han<sup>a</sup><sup>a</sup> School of Environmental Science and Engineering, South University of Science and Technology of China, Shenzhen 518055, Guangdong Province, China<sup>b</sup> College of Engineering, Peking University, Beijing 100871, China<sup>c</sup> School of Environment and Natural Resources, Renmin University of China, Beijing 100872, China

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## ABSTRACT

Soil contamination by mercury (Hg) is a global environmental issue. In watersheds with a significant soil Hg storage, soil erosion during rainfall-runoff events can result in nonpoint source (NPS) Hg pollution and therefore, can extend its environmental risk from soils to aquatic ecosystems. Nonetheless, transport mechanisms of soil-bound Hg in the erosion process have not been explored directly, and how different fractions of soil organic matter (SOM) impact transport is not fully understood. This study investigated transport mechanisms based on rainfall-runoff simulation experiments. The experiments simulated high-intensity and long-duration rainfall conditions, which can produce significant soil erosion and NPS pollution. The enrichment ratio (ER) of total mercury (THg) was the key variable in exploring the mechanisms. The main study findings include the following: First, the ER-sediment flux relationship for Hg depends on soil composition, and no uniform ER-sediment flux function exists for different soils. Second, depending on soil composition, significantly more Hg could be released from a less polluted soil in the early stage of large rainfall events. Third, the heavy fraction of SOM (i.e., the remnant organic matter coating on mineral particles) has a dominant influence on the enrichment behavior and transport mechanisms of Hg, while clay mineral content exhibits a significant, but indirect, influence. The study results imply that it is critical to quantify the SOM composition in addition to total organic carbon (TOC) for different soils in the watershed to adequately model the NPS pollution of Hg and spatially prioritize management actions in a heterogeneous watershed.

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## 1. Introduction

Soil is the largest reservoir of mercury (Hg) storage in global terrestrial ecosystems (Selin, 2010). A recent study (Smith-Downey et al., 2010) found that the soil Hg storage has increased by approximately 20% since 1840, largely due to the atmospheric Hg deposition primarily derived from anthropogenic sources such as coal burning, cement production and other industrial and mining activities. Soil contamination by Hg has been a global environmental issue, and extensive studies on Hg content and distribution in soils have been conducted (e.g., López-Blanco et al., 2015; Navrátil et al., 2014; Renneberg and Dudas, 2001; Zhou et al.,

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\* Corresponding author. School of Environmental Science and Engineering, South University of Science and Technology of China, Shenzhen 518055, Guangdong Province, China.

E-mail address: [yizheng@pku.edu.cn](mailto:yizheng@pku.edu.cn) (Y. Zheng).

2015). Yin et al. (2009) reported that the Hg content in soils close to secondary copper smelters in China ranged from 317 to 515 g/ha. Hg-contaminated soils can pose threats to surface water (Grigal 2002; Tersic et al., 2014). During rainfall-runoff events, soil-bound Hg can be transferred to neighboring water bodies by eroded sediments (Babiarz et al., 2012; Caron et al., 2008; Hope, 2005). In watersheds with a significant storage of soil Hg (e.g., Gong et al., 2014; Soares et al., 2015; Zhang et al., 2015), the soil erosion process can result in nonpoint source Hg pollution and extend the environmental risk from soils to aquatic ecosystems (Wiener et al., 2006). Abatement of Hg pollution has been a focus of watershed water quality management in developed countries (Chen et al., 2008; McCord and Heim, 2015), and quantification of the nonpoint source load of Hg is required for impaired water bodies (Chen et al., 2008).

It has been revealed in many studies (e.g., Burns et al., 2014; Caron et al., 2008; Mierle and Ingram, 1991; Obrist et al., 2011; Teisserenc et al., 2014; Wiener et al., 2006) that organic matter

(OM) plays a critical role in sorption of Hg and in the transfer of Hg from soils to aquatic environments. Nevertheless, most of the existing studies investigated the role of soil OM (SOM) by analyzing the correlation between Hg and total organic carbon (TOC) based on soil, water and sediment samples collected in field campaigns. Although the correlation was statistically significant in most cases, a substantial portion of Hg's variance could not be explained by TOC. The physical process of the Hg transfer still lacks direct exploration, and how SOM impacts the process has not yet been fully understood. This knowledge gap can cause significant uncertainty in calculating the nonpoint source load of Hg and in predicting its water quality response, a challenge yet to be tackled in water quality management.

Empirical modeling approaches have been adopted to quantify the nonpoint source load of Hg and its response. For example, Babiarz et al. (2012) established a multivariate regression model for the watershed yield of Hg into Lake Superior, considering land cover and soil types as the predictors. Although mathematically simple and generally reliable, such approaches are in general data-intensive and watershed-dependent. Physically based modeling approaches have also been used. For example, the Watershed Analysis Risk Management Framework (Goldstein, 2001), a watershed water quality model recommended by the United States Environmental Protection Agency (USEPA), simulates the physical processes of soil erosion and sediment (clay, silt, and sand) transport and explicitly models the partitioning between dissolved and adsorbed phases of sediment adsorbed chemicals such as Hg (Chen et al., 2008). Although approaches such as the one used in the Watershed Analysis Risk Management Framework have a theoretical foundation, they oversimplify reality. Additionally, the partitioning coefficient of Hg on sediments involves substantial uncertainty and is not measurable under field conditions. To achieve adequate modeling results, sufficient water quality observations are needed to infer the partitioning coefficient through a model calibration procedure.

In modeling nonpoint source pollution, the enrichment ratio (ER) is a useful metric for estimating chemical loading from soils to the sediment phase during rainfall (Bicknell et al., 2001; Neitsch et al., 2005). ER is defined as the ratio of the chemical content in suspended sediment to that in the original surface soil layer. ER usually exceeds one and diminishes as the rainfall proceeds. The theory behind ER (Menzel, 1980) is that surface runoff preferentially picks up and transports clay size particles because they weigh less than others, and chemicals can be primarily attached to clay size particles given their large surface-area-to-volume ratios. Empirical ER-sediment flux relationships, usually in the form of logarithmic or exponential functions, have been established for different chemicals (Menzel, 1980; Polyakov and Lal, 2004; Schiettecatte et al., 2008), but they do not vary for different soils. The nonpoint source load of a chemical can be easily derived, if its ER is estimated and the sediment flux is calculated. Recent studies on Polycyclic Aromatic Hydrocarbons (PAHs) (Zheng et al., 2012; Luo et al., 2013) revealed that the traditional theory interprets the enrichment phenomenon from an inappropriate perspective (i.e., particle size distribution instead of soil/sediment composition), which may not be valid for soils with significant amounts of anthropogenic carbonaceous materials such as black carbon particles. A different approach to calculating ER was also proposed (Luo et al., 2015). To the authors' best knowledge, there have been no studies specifically addressing the enrichment behavior of Hg. USEPA (1997) once applied the ER approach to assess the nonpoint source pollution of Hg in the U.S., but the ER values considered in the assessment were actually derived from the ER-sediment relationship for soil OM, not for soil Hg.

This study investigated transport mechanisms of soil-bound Hg

in the erosion process based on rainfall-runoff simulation experiments. The experiments simulated high-intensity and long-duration rainfall events, under which conditions significant soil erosion and nonpoint source pollution can occur. The ERs of total mercury, clay minerals (whose content is reflected by specific surface area) and TOC were the key variables in exploring the mechanisms. The impacts of different soil compounds, including clay minerals and different portions of SOM, on the enrichment behavior of soil-bound Hg were differentiated and compared. The major study objective was to identify the roles of different soil compounds in determining the enrichment behavior of Hg during the erosion process. The study results may be important to modeling and managing the nonpoint source pollution of Hg, and, hopefully, may elicit more research in this field.

## 2. Materials and methods

### 2.1. Soil for experiments

Soil samples were collected from two sites (see Table 1) in the downtown area of Beijing, the capital of China. These two soils were also investigated in our previous studies (Luo et al., 2013, 2015), where additional information about the soils can be found. At each sampling site, soil samples were taken from multiple spots in the site and were then mixed. Only the top 0–10 cm soil layer was sampled. The collected soil, stored in stainless-steel containers, was immediately transferred to a laboratory where the soil was first pretreated by removing any large pieces of debris (e.g., gravel, plant residues, etc.). After being passed through a 4 mm stainless steel sieve, the soil was air-dried at room temperature and then mixed completely. The soil was stored in stainless containers and kept sealed prior to analysis.

### 2.2. Experimental design

We conducted rainfall-runoff simulation multiple times for each of the two soils. This study analyzed two simulation experiments, labeled as Experiment I (using Soil I) and Experiment II (using Soil II), which had almost identical rainfall-runoff processes. In both experiments, the rainfall intensity was kept at 90 mm/h for more than 2 h. It is worth pointing out that the simulation experiments were designed to investigate transport mechanisms, rather than to reproduce specific rainfall-runoff conditions and estimate the corresponding pollutant loads. Using the unusual rainfall-runoff conditions was to ensure a sufficient amount of eroded sediment for laboratory analysis within a reasonable simulation period. Fig. 1 provides a graphic summary of the experimental design. The experimental system (Fig. 1a) was mainly composed of a transparent soil box and a rainfall simulator. The box (100 cm×15 cm×30 cm) was made of polymethyl methacrylate and fixed on a wooden wedge (slope = 5°). A V-shaped weir was installed at the lower end of the box, which directs water through a trough. The box was first packed with clean and dry sands (d = 0.8 mm) to form a bottom layer (approximately 28 cm in thickness) of relatively good permeability. The well-mixed soil was then packed above it to form a 2.2–2.5 cm thick soil layer, following Schiettecatte et al. (2008). The packed soil was well-aggregated and approximately 1.3 g/cm<sup>3</sup>. The rainfall simulator can provide a constant, controllable delivery rate (i.e., rainfall intensity) of water to the soil packed in the box. More details about the experimental system can be found in Zheng et al. (2012).

The rainfall durations in Experiment I and Experiment II were 2.29 h and 2.39 h, respectively. The artificial rainfall samples were sourced from tap water, and the THg was not detectable. Runoff samples were collected at the outlet of the trough, using acid-

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