



Soil mercury accumulation, spatial distribution and its source identification in an industrial area of the Yangtze Delta, China

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ABSTRACT

Understanding soil mercury (Hg) accumulation, spatial distribution, and its sources is crucial for effective regulation of Hg emissions. We chose a study area covering approximately 100 km² representing one of the rapid growing industrial towns of the Yangtze River Delta (YRD), China, to explore soil Hg accumulation. In surface soil, total Hg ranged from 310 to 3760 µg/kg, and 53% samples exceeded the most generous Chinese soil critical value (1500 µg/kg). Hg concentration in rice ranged from 10 to 40 µg/kg, and 43% samples exceeded the regulatory critical value (20 µg/kg). Total Hg concentrations in soil profiles gradually decreased, reaching background levels up to 60 cm profile depth. Meanwhile, proportions of mobile, semi-mobile and non-mobile Hg to total Hg at every soil depth were similar, leading us to deduce that soil Hg has accumulated in this area over a long period. Total and bioavailable Hg in topsoil exhibited the highest concentrations in the center of the study area, and radially decreased towards the periphery, which might be explained by the distribution of industry and the prevailing wind. To trace the Hg sources, we selected soil and atmospheric dust samples for isotope analysis. Hg isotopic composition of surface soil ($\delta^{202}\text{Hg} = -0.29 \pm 0.10\text{‰}$ and $\Delta^{199}\text{Hg} = 0.03 \pm 0.03\text{‰}$) was close to that of atmospheric dust ($\delta^{202}\text{Hg} = -0.54 \pm 0.10\text{‰}$ and $\Delta^{199}\text{Hg} = 0.03 \pm 0.05\text{‰}$), but considerably different from Hg isotopic composition in subsoil ($\delta^{202}\text{Hg} = -0.90 \pm 0.09\text{‰}$ and $\Delta^{199}\text{Hg} = -0.04 \pm 0.04\text{‰}$). Thus, we speculated that atmospheric deposition could change Hg isotopic composition in topsoil. Our findings suggest that when Hg atmospheric dust deposition changes Hg levels in surface soil, soil remediation, and crop safety might be compromised.

1. Introduction

Mercury (Hg) has been listed as one of the top ten chemicals of public health concern by the World Health Organization, posing a serious threat for ecosystem and human health, such as nervous, digestive and immune systems, lungs, kidneys, and skin and eyes, at local and global scales (World Health Organization WHO, 2016). Modern global Hg contamination is dominated by anthropogenic influences, such as fossil fuel combustion, smelting, electroplating, etc. (Sun et al., 2014; United Nations Environmental Programme UNEP, 2013), which is more serious in developing countries because of their accelerated industrialization in the last few decades (Feng et al., 2006; Liang et al., 2013; Sun et al., 2014; United Nations Environmental Programme UNEP, 2013).

Prior studies have extended the research area on Hg accumulation into soil environment (Feng et al., 2010, 2004, 2006; Ottesen et al.,

2013; Wiederhold et al., 2013). Due to cumulative effects and long-term interactions of heavy metals, accumulation of Hg in soil poses a threat to crops which can transfer to humans through food chain (Chang et al., 2014). Different from Hg accumulation in mining soils (Feng et al., 2010, 2004, 2006; Wiederhold et al., 2013), Hg transferring from agricultural soil to crops depends not only on its accumulation but also on soil properties such as soil pH, oxidation-reduction potential (Eh) (Jing et al., 2007; Randall et al., 2004), soil organic matters (SOM) (Qian et al., 2009), and residue time (Grimaldi et al., 2008). Therefore, it is of practical significance to identify and understand the accumulation, spatial distribution, sources of Hg in soil, and the correlations in soil and crops to effectively regulate emissions and reduce safety threat of Hg to food and populations.

Hg accumulation in agricultural soil in China is getting increased attention, such as in the Yangtze River Delta (YRD) (Huang et al., 2011; Xu et al., 2014) and the Pearl River Delta (Chang et al., 2014) in the

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southeast of China. As one of the fastest economic development regions in China, the YRD (covering Jiangsu, Shanghai, and Zhejiang provinces), Hg accumulation in soil has occurred because of industrial emissions of Hg-containing chemicals and uncontrolled agricultural inputs over the last three decades (Huang et al., 2011; Shao et al., 2006; Shen et al., 2010; Xu et al., 2014). For example, Xu et al. (2014) pointed out that Hg pollution covered approximately 38% area of the YRD, and the sources of Hg pollution may be attributed to atmosphere deposition from industrial emissions and historical use of Hg containing pesticides. Shen et al. (2010) revealed that Hg pollution in surface soil in Suzhou, Jiangsu province exceeded national/local background levels of Hg (150/289 $\mu\text{g}/\text{kg}$). However, the existing studies mainly focused on the current levels of Hg accumulation in soils in the YRD area and its spatial distribution, less on exploring sources of Hg emissions.

Furthermore, Huang et al. (2011) found that the highest total Hg and bioavailable Hg (HCl-Hg) concentrations in soil of Zhangjiagang, Jiangsu were shown in the southwest of the wastewater outlet within about 50 m away from the outlet, and total Hg and HCl-Hg concentrations decreased as the distance from the outlet increased. However, the degree of influence on Hg distribution by wastewater was limited compared to atmospheric pathways Feng et al., 2006). Thus, it is important to further consider atmospheric influence on the Hg spatial distribution pattern in soil.

Source identification of soil Hg is crucial for regional soil remediation and management of emissions. With the improvement of analytical methods, Hg stable isotopic analysis is becoming an effective method to identify sources of Hg in the environment (Wiederhold et al., 2013; Yin et al., 2013; Yu et al., 2016). Hg possesses seven stable isotopes which can be systematically fractionated during specific transformation reactions, and Hg isotope compositions vary significantly between different source materials (Sherman et al., 2012; Sun et al., 2014; Wiederhold et al., 2013; Yin et al., 2013). With the development of multi-collector inductively coupled plasma mass spectrometry (MC-ICP-MS) (Blum and Bergquist, 2007), high-precision analyses of many “non-traditional” stable isotope systems including Hg have become feasible (Lauretta et al., 2001; Perrot et al., 2010; Sun et al., 2014; Wiederhold et al., 2013). Thus, it is appropriate to adopt this method to identify the source of agricultural soil Hg.

In order to cover the shortage on the research of Hg accumulation in the YRD area, we chose a typical industrial town in this area to determine two objectives: 1) assess levels of total Hg in soil and rice and bioavailable Hg in soil, and 2) explore sources of Hg based on spatial pattern of soil Hg and Hg isotopic ratios. Sources of Hg pollution were traced in the vicinity of a typical industrial town in the YRD based on its spatial distribution of Hg concentration in soil, whereas the effect of atmospheric disposition was discerned through the isotopic analysis. The contribution of this paper adds our knowledge on identifying Hg pollution sources in soil, which will provide a basis for evaluating regional soil Hg risks and food safety.

2. Materials and methods

2.1. Study area

The study area covers approximately 100 km^2 in Jiangsu province between longitude 31° 24' N to 31° 30' N and latitude 120° 26' E to 120° 35' E (Fig. 1). This area has a sub-tropical monsoon climate (prevailing southeast wind in summer and northwest wind in winter), with annual mean rainfall of 1040 mm and an average (lowest – highest) temperature of 17.6 (– 8.3 to 39.4) °C. The soil was developed from lacustrine deposits in the Holocene Epoch and have a neutral and weak acidic reaction and a clay loam texture according to the Soil Chronicle of Zhangjiagang County in Jiangsu province (Soil Survey Office of Zhangjiagang County SSOZJG, 1984). The distributions of industry, residence, and farmland around 2010 are shown in Fig. 1.

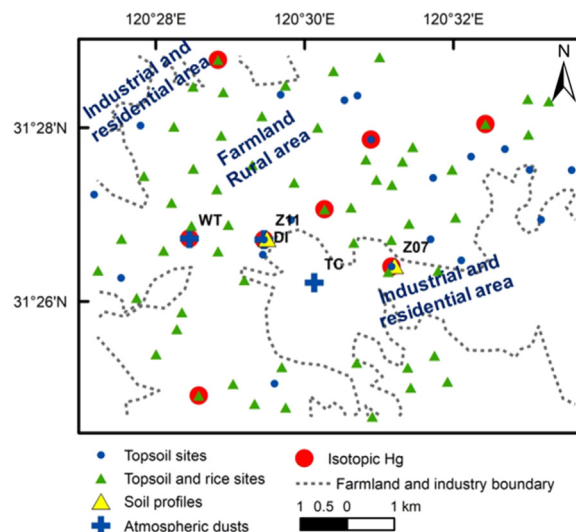


Fig. 1. Sampling map in the study area.

2.2. Sample collection and preparation

We considered the spatial homogeneity of samples, layout of factories and residential area, and town center, etc. when setting collecting sites in this area. The land use types of soil mainly involved rice field and industrial land. A total of 76 topsoil (0 – 20 cm) samples were collected throughout the study area, and the coordinates of each sample was recorded by GPS. Each soil sample was a composite of five subsamples within 100 m^2 collected in a sealed polyethylene bag. A total of 56 rice grain samples were collected directly from the rice field when the soil samples in the same sites were collected at the same time (Fig. 1). Two typical soil profiles were sampled in the farmland close to typical industrial areas (Z07 and Z11), for determining vertical distributions of Hg. A stainless steel core sampler (3 cm inner diameter) was dug 100 cm into the soil at five locations with 100 m^2 , and composited separately at the depths of 0–10 cm, 10–20 cm, 20–40 cm, 40–60 cm and 60–100 cm.

The soil was air-dried at room temperature, homogenized, and grinded to pass through a 2 mm mesh nylon sieve for measuring soil pH and bioavailable Hg. For SOM and total Hg determination, the soil was further grinded to pass through a 0.15 mm mesh. The rice grain was thoroughly washed and rinsed with deionized water, and then sucked by a filter paper. The grain was dried in an air-circulating oven at 60 °C until a constant weight, and then was manually de-husked and grinded by using a stainless steel grinding machine to pass a 0.25 mm sieve.

To trace the influence of atmospheric deposition on Hg in soil, three atmospheric dust samplers were installed at locations: (i) close to industries (DI), (ii) in the town center (TC) and (iii) in the rural area (WT). The dust sampler consisted of a collection barrel made of stainless steel, 30 cm high and 15 cm inner diameter, bucket lined with a plastic bag and filled up with a layer of glass balls (diameter 1.2 cm) under the plastic lining. The barrel was positioned on a metallic stand 1.5 m above ground level. The dust was allowed to accumulate for 30 ± 2 day and quantitatively washed in a glass beaker using distilled deionized water. The suspension volume was reduced to 10–20 mL on a hot plate below 60 °C. The suspension was transferred to Teflon baker, and dried in an oven until constant weight for further analysis.

As general characteristics, soil pH was measured in 1:2.5 soil to water ratio mixture (Lu, 2000). SOM was analyzed by wet oxidation using excess dichromate, and the unconsumed dichromate is then back-titrated with an iron sulfate solution (Walkley-Black method) (Nelson and Sommers, 1982). Mercury isotope analyses were carried for only selected soil and dust samples. Five surface soil from farmlands, three soil depths from each of the two profiles (0 – 10 cm, 10 – 20 cm, and 60

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