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Mercury in soil, vegetable and human hair in a typical mining area in China: Implication for human exposure

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ABSTRACT

Concentrations of total mercury (T-Hg) and methylmercury (MeHg) in soil, vegetables, and human hair were measured in a mercury mining area in central China. T-Hg and MeHg concentrations in soil ranged from 1.53 to 1054.97 mg/kg and 0.88 to 46.52 μg/kg, respectively. T-Hg concentrations was correlated with total organic carbon (TOC) content ($R^2 = 0.50$, $p < 0.01$) and pH values ($R^2 = 0.21$, $p < 0.05$). A significant linear relationship was observed between MeHg concentrations and the abundance of sulfate-reducing bacteria (SRB) ($R^2 = 0.39$, $p < 0.05$) in soil. Soil incubation experiments amended with specific microbial stimulants and inhibitors showed that Hg methylation was derived from SRB activity. T-Hg and MeHg concentrations in vegetables were 24.79–781.02 μg/kg and 0.01–0.18 μg/kg, respectively; levels in the edible parts were significantly higher than in the roots (T-Hg: $p < 0.05$; MeHg: $p < 0.01$). Hg species concentrations in rhizosphere soil were positively correlated to those in vegetables ($p < 0.01$), indicating that soil was an important source of Hg in vegetables. Risk assessment indicated that the consumption of vegetables could result in higher probable daily intake (PDI) of T-Hg than the provisional tolerable daily intake (PTDI) for both adults and children. In contrast, the PDI of MeHg was lower than the reference dose. T-Hg and MeHg concentrations in hair samples ranged from 1.57 to 12.61 mg/kg and 0.04 to 0.94 mg/kg, respectively, and MeHg concentration in hair positively related to PDI of MeHg *via* vegetable consumption ($R^2 = 0.39$, $p < 0.05$), suggesting that vegetable may pose health risk to local residents.

Introduction

Mercury (Hg) is a hazardous and persistent environmental pollutant released from both natural and anthropogenic sources. It exists as the inorganic form Hg^0, Hg(II), Hg(I) and organic mercury compounds, such as methylmercury (MeHg). Hg toxicity depends on its chemical form (Clarkson and Magos, 2008). MeHg is considered the most toxic form, because of its tendency to bioaccumulate and biomagnify. Inorganic Hg species transform into MeHg through a variety of processes, involving both abiotic and biotic pathways. MeHg is produced by abiotic methylation *via* transmethylation reactions between Hg and other compounds containing methyl groups (Ullrich et al., 2001). Biological methylation is dominated by

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microorganisms and considered the main process of MeHg production (Yu et al., 2012). Parks et al. identified two gene clusters, *hgcA* and *hgcB*, which correlate with the ability of bacteria to methylate Hg (Parks et al., 2013). Sulfate-reducing bacteria (SRB) and iron-reducing bacteria (FeRB) are methylators of inorganic Hg, and utilize sulfate and Fe(III), respectively, as the terminal electron acceptor (Compeau and Bartha, 1985; Fleming et al., 2006). SRB plays the dominant role in methylation of inorganic Hg in the environment.

Hg methylation is prevalent in soil, and this process poses environmental risk to humans because MeHg can bioaccumulate and bioamplify in the soil–plant system. Multiple studies have focused on microbial or abiotic methylation of Hg in wetlands and sediment (Lehnherr et al., 2012; Schartup et al., 2013), but little attention has been paid to the microbial methylator and its contribution to MeHg production in soil. Since some studies reported that crops such as rice and vegetables were important pathways of Hg exposure for people living in Hg-mining areas (Zhang et al., 2010), concerns have been raised regarding Hg methylation in soil and transfer to edible plants.

The study area was a mining region located in central China. Hg was found there around 770 BC, and mining operations were taken over by the government in the 1980s. Intense Hg pollution in nearly all environmental compartments occurred from the mining and retorting activities. This Hg environmental contamination can pose serious threats to soil ecosystems and food safety (Zhang et al., 2009; Qiu et al., 2012). Therefore, it is necessary to quantify Hg methylation through abiotic or microbial processes. The aim of this study was to investigate Hg contamination and identify the main soil methylator of Hg in the study area. We also measured the concentrations of total mercury (T-Hg) and MeHg in vegetables and the corresponding rhizosphere soil collected, and estimated the health risk posed by T-Hg and MeHg in vegetables. To evaluate human exposure, T-Hg and MeHg levels in human hair samples were quantified.

1. Materials and methods

1.1. Study area

The study area was an Hg-mining area in a valley in the southern part of Shaanxi Province in central China. The mining area is approximately 5 km^2; a map of the area is shown in Fig. 1. The study area has a typical sub-tropical humid climate with an annual average rainfall of 859.4 mm. The annual mean temperature is 14.8°C, with temperature typical high of 38°C in July and low of 1°C in January. The soil is yellow-brown earth and the primary ore minerals are cinnabar (HgS) and stibnite (Sb_2S_3). Mining and smelting of Hg ores still occurs in the study area, and the locations of the air outlets from the mine, smelting workshop, ore-concentration workshop, and drain outlet are marked in Fig. 1. The smelting workshop and ore-concentration workshop are at the north end of the valley. A river flows eastward at the southern end of the valley in proximity to the mining area. The wastewater generated during the mining, concentrating, and smelting processes is treated. Some of the treated water is reused in the Hg production processes, and the rest is discharged directly into the river.

Approximately 200 inhabitants, including 100 workers and 96 residents, live in the study area. The area residents seldom eat fish, and rice and vegetables are the staple foods. Since residents have some awareness of the health risks associated with Hg mining activities, they consumed commercial rice from an uncontaminated area. However, the vegetables they consumed are grown on their own farmland.

1.2. Sample collection

Surface soil samples (n = 45) were collected from the study area in August of 2014. Four of the samples were collected within the Hg mining and smelting area, and the rest were

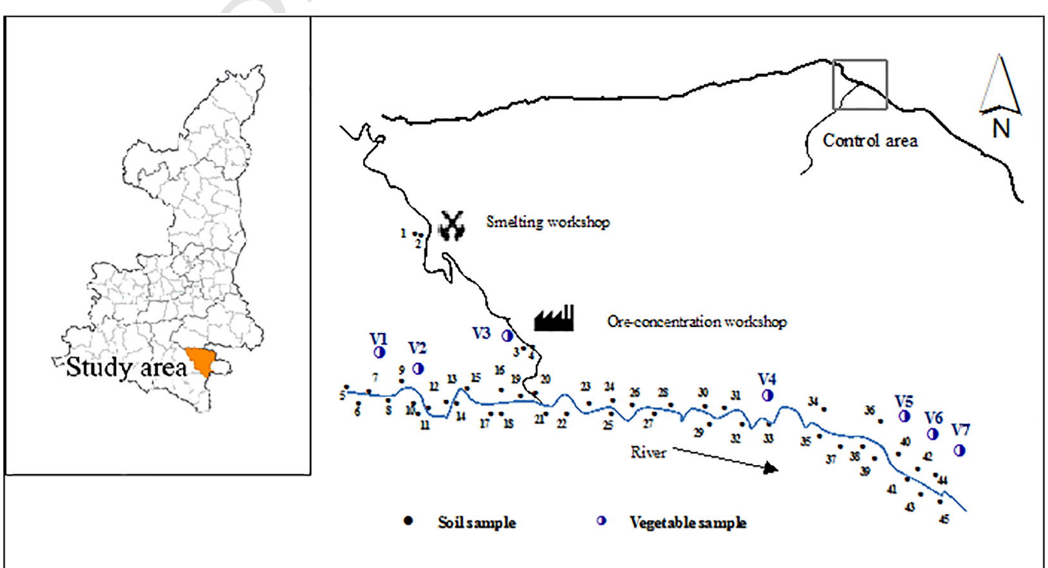

Fig. 1 – Soil and vegetable sampling sites in the study area. 1–45: soil sample; V1–V7: vegetable sample.

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