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Lead in Chinese villager house dust: Geographical variation and influencing factors

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ABSTRACT

House dust has been recognized as an important contributor to Pb exposure of children. Here we conducted a comprehensive study to investigate geographical variation of Pb in Chinese villager house dust. The influences of outdoor soil Pb concentrations, dates of construction, house decoration materials, heating types, and site specific pollution on Pb concentrations in house dust were evaluated. The concentrations of Pb in 477 house dust samples collected from twenty eight areas throughout China varied from 12 to 2510 mg/kg, with a median concentration of 42 mg/kg. The median Pb concentrations in different geographical areas ranged from 16 (Zhangjiakou, Hebei) to 195 mg/kg (Loudi, Hunan). No correlations were found between the house dust Pb concentrations and the age of houses, as well as house decoration materials. Whereas outdoor soil, coal combustion, and site specific pollution may be potential Pb sources. Principal component analysis (PCA) confirmed that elemental compositions of the house dust were controlled by both anthropogenic and geogenic sources. Using scanning electron microscopy (SEM), the Pb bearing particles in the house dust were also studied.

1. Introduction

In the last few decades, huge amounts of metal/metalloid pollutants have been released from the growing industrial plume (Bollhöfer and Rosman, 2001; Bi et al., 2013; Luo et al., 2012; Peña–Fernández et al., 2014) as well as natural sources (Argyraki and Kelepertzis, 2014). These hazardous pollutants pose a high risk to human health and our surrounding environments. Thus, great concerns have been raised over their adverse health effects on a global scale (Nordberg et al., 2015).

House dust is an important carrier for trace metal/metalloids which can enter human body through dermal contact, inhalation and direct ingestion via the hand-to-mouth activity. House dust has long been recognized as a significant contributor to blood lead levels (BLLs) in children (Duggan, 1983; Duggan and Inskip, 1985; Paustenbach et al., 1997; Lanphear et al., 1998; Dixon et al., 2009; Gaitens et al., 2009; Laidlaw and Taylor, 2011; Gulson et al., 2014; Li et al., 2015). For example, Li et al. (2015) compared the relationship between BLLs of children living in an industrial town in China and different environmental media (house dust, soil, PM_{10}, vegetables, rice, and drinking water) using Pb isotope ratios, and found that incidental ingestion of house dust was the predominant contributor to children's blood Pb. According to mixed model analyses, Gulson et al. (2014) observed that BLLs (geometric mean 21 μg/L) of 108 children ≤5 years in Sydney were strongly associated with interior house dust and soil.

In China, the BLLs of children (44.5–163.5 μg/L) (He et al., 2009; Li et al., 2014a,b) were notably higher than those in developed countries, such as U.S. (1–5 years, 13–19 μg/L) (CDC, 2013). Furthermore, the mean BLLs of Chinese children living in suburban and rural areas (115.8 μg/L) were higher than those found in urban areas (90.3 μg/L) (He et al., 2009). This is understandable because as a number of industrial enterprises gradually shifted to the suburban areas, the suburban children have more access to soil/dust. Additionally, in suburban areas, both the environmental governance and health conditions are normally poorer compared with urban areas (He et al., 2009). This serious situation necessitates an urgent need to fully investigate the occurrence of Pb in house dust and their contribution to daily Pb exposure of Chinese children. In fact, such work has been extensively conducted in many other

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countries, such as U.S. (Clark et al., 1991; Lanphear et al., 1998; Farfel et al., 2005; Dixon et al., 2009; Gaitens et al., 2009), U.K. (Thornton et al., 1990), Japan (Yoshinaga et al., 2014), Canada (Rasmussen et al., 2001, 2011, 2013), and Australia (Gulson et al., 2014).

Lead contamination in house dust was closely associated with Pb-based paints used for house decoration in Euro–American countries (Meyer et al., 1999; Davis and Gulson, 2005; Dixon et al., 2009; Rasmussen et al., 2001, 2011). However, Pb in house dust may be derived from various sources other than Pb-based paints, especially for Asian residences where residential Pb-based paints had not been used (Yoshinaga et al., 2014; Li et al., 2014a,b). In limited dust samples, Li et al. (2014a,b) had found that the levels of Pb in Chinese rural residences (92 and 126 mg/kg for mean and median concentrations, respectively) were notably higher than the background values. According to stable Pb isotope ratios, the authors suggested that coal combustion was a major source of Pb in Chinese house dust (Li et al., 2014a,b). Our previous study in Chinese rural residences had also observed elevated Pb (mean 208 mg/kg) in the house dust (Han et al., 2012) compared with the background values. Here we extend our work geographically and discuss the possible reasons behind the elevated dust Pb. For this purpose, house dust samples were collected from 477 rural residences throughout China, and Pb and other elements were subsequently determined. In addition, the morphology of Pb-bearing particles in the house dust was analyzed with a scanning electron microscopy (SEM) equipped with an energy dispersive X-ray spectrometer (EDX) microanalyzer.

2. Materials and methods

2.1. Sample collection

The sampling was carried out by college student volunteers. The participants were randomly chosen and a total of thirty five students joined this study. The students were asked to collect house dust samples around their own hometown. During the sampling, a questionnaire related to dust composition (e.g., heating types, dates of construction, building and decoration materials) was completed in parallel. A total of 477 house dust samples were collected from twenty eight areas in China between August 2009 and October 2010. The studied areas and sampling sites are showed in Fig. 1. Dust samples were collected from the inside floor (bed room and living room) using a brush and plastic spatula. All dust samples were stored in sealed polyethylene bags, labeled and then transported to the laboratory. Lead results for some samples collected in 2009 were published in Han et al. (2012).

For comparison with the indoor dust, the concentrations of soil Pb from an unpublished database accomplished by the China Geochemical Baselines Project (CGB) (Wang and the CGB Sampling Team, 2015) were obtained in this study. A total of 215 soil samples were selected from the same twenty eight areas (the same towns or cities) where the dust samples were collected. The sampling campaign for the soil samples had been fully described by previous study (Wang and the CGB Sampling Team, 2015). Briefly, the samples were collected at a depth of approximately 0–25 cm and each sample was composited from 3 pits in a layout of an equilateral triangle within 50 m interval. Samples were not collected within 100 m of motorways/highways or roads, or within 2000 m of towns or cities.

2.2. Sample analytical procedure

The dust samples were air-dried at room temperature, and passed through a 60 mesh sieve (250 μm) to remove rocks, plants, hair and other impurities. The passed fraction was used for chemical analysis because it was likely to adhere to children's hands (Duggan and Inskip, 1985; Yamamoto et al., 2006). About 250 mg of the homogenized dust sample was digested with a concentrated HNO_3–$HClO_4$–HF–HCl mixture. The concentrations of Pb and other elements of the digested solution were determined by an inductively coupled plasma-atomic emission spectrometer (ICP-AES) (Varian Vista-pro). Sb and As concentrations were determined by a hydride generation-atomic fluorescence spectrometer system (HG-AFS) (Beijing jitian, AFS-810). Quality assurance (QA) and quality control (QC) included reagent blanks, analytical duplicates, and analysis of the standard reference materials (SRMs) (MRGeo08, OGGeo08, and OREAS90 from Australia). The results of our SRM analyses agreed well with the certified values as shown in Table S1. The measurement of soil samples can be seen in a previous study (Wang and the CGB Sampling Team, 2015). Air-dried samples were ground to less than 200 mesh (<70 μm) in an agate and digested with a concentrated HNO_3–$HClO_4$–HF–HCl mixture. The concentrations of Pb of the digested solution were determined by an inductively coupled plasma-mass spectrometer (ICP-MS). Analytical accuracy and precision for the laboratory quality was strictly controlled by laboratory replicate samples and SRMs.

Several dust samples with elevated Pb concentrations (above 300 mg/kg) were randomly selected for morphology observation. Subsamples of the selected house dust were fastened on testing platform with carbon coated adhesive tape, and then analyzed with a field emission scanning electron microscopy (SEM) (Hitachi SU8010). The photomicrograph of the dust particles was obtained at an operating voltage of 1.5kv. Based on energy dispersive X-ray microanalysis (EDX) system, the Pb-bearing particles was identified using a surface scanning map at an operating voltage of 15.0kv.

2.3. Statistical analysis

The data were statistically analyzed using SPSS v19.0 and Excel 2007. The differences in Pb concentrations among different settings were analyzed using non-parametric statistical tests. Principal component analysis (PCA) was conducted to the elemental data after log-transformed of the raw values. The factor extraction (eigenvalues >1) method was used after varimax rotation.

3. Results and discussion

3.1. Element properties

The profiles of the studied elements related to dust properties are presented in Table 1, along with data from Japanese (Yoshinaga et al., 2014) and Canadian (Rasmussen et al., 2001) dusts and Chinese background soils (CEMS, 1990) for comparison. In comparison with the background values of Chinese soils, the concentrations of Al, Ba, Fe, K Mg, Mn, and Na in Chinese villager house dusts were comparable, whereas the Ca and Sr concentrations were much higher and the Sc, Ti, and V concentrations were notably lower. When compared with results of Japanese and Canadian dusts, most of the considered elements in Chinese villager house dusts exhibited higher concentrations (Table 1), which might point to the different constituents between them.

The whole data set of Pb in the villager house dust is not log-normally distributed, thus the median values were used to summary the distribution of Pb in this study, but the arithmetic mean and geometrical mean values were also given to facilitate comparison with other studies (Tables 1 and 2). The overall range of Pb concentrations varied from 12 to 2510 mg/kg, with a median concentration of 42 mg/kg, which were similar to those of Japanese dust but notably lower than those of Canadian dust (Table 1).