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In vitro bioaccessibility and health risk assessment of heavy metals in atmospheric particulate matters from three different functional areas of Shanghai, China

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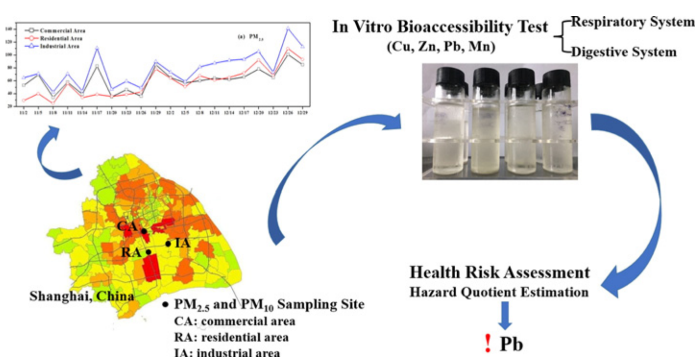
HIGHLIGHTS

- $PM_{2.5}$ and PM_{10} concentrations in three different areas of Shanghai were investigated.
- Both $PM_{2.5}$ and PM_{10} concentrations in the three areas exceeded the WHO guideline.
- Zn and Pb were the most abundant heavy metals in both $PM_{2.5}$ and PM_{10}.
- Bioaccessibility of metals in both respiratory and digestion systems were evaluated.
- Health risks to children and adults mainly resulted from Pb via ingestion exposure.

GRAPHICAL ABSTRACT



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ABSTRACT

The bioaccessibility and human health risks of heavy metals in $PM_{2.5}$ and PM_{10} samples from three functional areas of Shanghai, China including a commercial area (CA), a residential area (RA), and an industrial area (IA), were investigated. Gamble's solution and physiologically based extraction test were employed to simulate human respiratory and digestive system, respectively. Both $PM_{2.5}$ and PM_{10} concentration in the three areas exceeded the guideline of WHO, and followed the order of IA > CA ≈ RA. Zinc and Pb were the most abundant metals with a concentration range of 0.19–0.44 and 0.05–0.42 μg m^{-3}, respectively. In respiratory system, heavy metal bioaccessibility for $PM_{2.5}$ and PM_{10} varied within the range of 5.3%–71.4% and 4.8%–51.5%, respectively. Heavy metals in RA showed higher bioaccessibility than those in CA and IA in the respiratory system. In digestive system, heavy metal bioaccessibility for $PM_{2.5}$ and PM_{10} reached 24.6%–90.9% and 28.5%–88.9% in the gastric phase and was reduced to 8.7%–85.5% and 8.5%–81.8% in the intestinal phase, respectively. The bioaccessibility of heavy metals in CA was highest among three areas in the digestive system. Based on the bioaccessibility analysis, the hazard quotient values of heavy metals in PMs via inhalation exposure were far below 1, the safe level, for both adults and children. However, potential risks via ingestion exposure resulted from Pb existed for children of three areas and for adults of RA as their hazard quotient values could reach up to 11. The obtained results indicated that the air quality in Shanghai need to be improved and the health risks to humans via ingestion exposure to atmospheric Pb must be considered.

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1. Introduction

Atmospheric particulate matters (PMs) pose potential health risks to human beings, including upper respiratory irritation, asthmatic attack, lung cancer and heart disease, et al. (Gao et al., 2015; Kan et al., 2012; Wang and Mauzerall, 2006). Heavy metals present in PMs can play a significant role in inducing these health impairments. Human exposure to PMs contaminated with heavy metals is mainly through the respiratory system, but many studies also show that air particles could deposit on food or object surfaces, transporting into human body via ingestion exposure (Hu et al., 2012; Sun et al., 2014; Zheng et al., 2015). The potential health effects of heavy metals in PMs depend on their total contents, bioavailability, relative toxicity and a number of factors related to inhalation and ingestion exposure (Bi et al., 2015; Huang et al., 2016; Wiseman and Zereini, 2014). Animal-based in vivo evaluation of heavy metal bioavailability is complex, expensive, and time-consuming (Zia et al., 2011), therefore, a variety of in vitro methods have been developed to estimate the bioaccessibility of heavy metals by using the simulated human body fluids. Gamble's solution and the physiologically based extraction test (PBET) are widely acknowledged in vitro methods and have been frequently applied to represent the uptake of heavy metals in the human respiratory and digestive system, respectively (Bruce et al., 2007; da Silva et al., 2015; S.W. Li et al., 2016; Ruby et al., 1996). The bioaccessibility obtained from these two methods have been proved to be fairly good correlated with in vivo bioavailability (Deshommes et al., 2012; H. Li et al., 2015). However, many of such tests, especially the tests in the digestive system were applied in soil and dust samples, while the bioaccessibility of heavy metals in PM samples has received limited attention.

The health risk assessment model proposed by USEPA has been used for investigating the potential health risks related with exposure to heavy metals in particles in many different regions, such as Kalimantan, in Indonesia (Betha et al., 2013), Shanxi, Shanghai, and Beijing in China (Chen et al., 2011; Cao et al., 2014; Gao et al., 2015). The bioaccessibility of heavy metals in respiratory and digestive systems significantly affected the health risk evaluation of PMs, since these soluble portions are more likely to be capable of inducing toxicity (Bi et al., 2015; Wiseman and Zereini, 2014). However, most of these studies used the total concentration of heavy metals, rather than their bioaccessible fractions to model the health risks of PMs. As a result, the health risk of heavy metals in PMs would be somewhat overestimated in this case. Meanwhile, the bioaccessibility of heavy metals in PMs from different areas might be discrepant due to their different chemical forms and particle sizes (Juhasz et al., 2007; Walraven et al., 2015). However, most sampling areas in the studies mentioned above were randomly arranged to describe the atmospheric environment of whole cities, thus PM sources associated with the functions of sampling areas were not taken into account for bioaccessibility analysis of PMs. In our opinion, bioaccessibility analysis of heavy metals in PMs based on different functional areas was more reasonable and directional, rather than the assessment at the scale of a whole district or city. Therefore, it is crucial to evaluate the bioaccessibility of heavy metals in PM from different functional areas via inhalation and ingestion in order to regionally assess the health risk of PM in a specific city.

In this study, $PM_{2.5}$ and PM_{10} samples were collected from three different functional areas in Shanghai, China including a commercial area, a residential area, and an industrial area during November to December 2015, and evaluated for the bioaccessibility via inhalation and ingestion and health risk of heavy metals existed in these samples. Specifically, this study aimed to: (1) estimate the in vitro bioaccessibility of heavy metals in $PM_{2.5}$ and PM_{10} in the respiratory and digestive system by applying Gamble's solution and physiologically based extraction test (PBET) methods, respectively; and (2) assess the health risks of heavy metals in $PM_{2.5}$ and PM_{10} on adults and children via inhalation and ingestion exposure based on bioaccessibility analysis.

2. Materials and methods

2.1. Sample collection

The samples of $PM_{2.5}$ and PM_{10} were collected from three different functional areas at a height of about 18 m in Shanghai. The commercial area (CA) sampling site is located on the roof of a shopping mall, Xuhui District (121°26′0″ E, 31°11′54″ N). The residential area (RA) sampling site is in a residential building located in the junction of Minhang District and Fengxian District, beside the entrance of Shanghai-Jiaxing-Huzhou Expressway and Shanghai-Jinshan Expressway (121°26′38″ E, 30°59′27″ N). The industrial area (IA) sampling site is in a thermal power plant located in Minhang District (121°28′9″ E, 31°3′11″ N). All samples were collected every third day for a 24-h period from November to December of 2015 with Whatman quartz microfiber filters (φ 90, QMA, Whatman International Ltd., Maidstone England) using a medium-flow air sampler (Laoying 2030, Qingdao Laoshan Applied Technology Research Institute, Qingdao, China). The daily air temperature and wind speed during sampling period were shown in Fig. S1. The sampling flow rate was 100 L min^{-1}. A total of 190 valid samples including $PM_{2.5}$ and PM_{10} were collected, as some of the samples were invalid or missing due to mechanical failures. The quartz microfiber filters were heated at 500 °C for 4 h to remove the organic matter and then cooled down in a desiccator for 24 h prior to use (Hu et al., 2012; Zhai et al., 2014). After sampling, the filters were treated with the same procedure as before (500 °C for 4 h, desiccated for 24 h). All samples were weighed by an analytical balance to determine the particulate matter mass by subtracting the filter mass (AL204-IC, Mettler Toledo, 0.0001 g). The samples with quartz microfiber filters were then placed into a clean glass vial with a Teflon-lined cap and stored at 4 °C prior to analysis.

2.2. Sample treatment and heavy metals analysis

One quarter of the filter was cut into fragments by a Teflon scissor and then dissolved in the mixture of 6 mL 65% HNO_3 and 3 mL 30% H_2O_2 using a microwave digestion system (Topwave, Analytik Jena Co. Ltd., German) (Hu et al., 2012). The solution was first heated to 210 °C within 10 min and then the temperature was held at 210 °C for 30 min. When the digestion was finished, the solution was cooled to the room temperature naturally and filtrated with 2% v/v HNO_3 washing, then made up to 50 mL with 2% v/v HNO_3. Next, the concentrations of heavy metals (Zn, Pb, Cu, Mn, Ni, Cr, Cd, etc.) in the digested solution were measured using an inductively coupled plasma-optical emission spectrometer (ICP-OES, PS3500 DD, Hitachi Co. Ltd., Japan). The detection limits for all heavy metals were 0.8–10.0 μg L^{-1}. Finally, the volume concentration of each heavy metal in the atmosphere was calculated according to the concentrations in digested solution and the sample sizes. For quality control, two blanks including filter and chemicals alone and one standard reference material (No. SRM2783; National Institute of Standard and Technology, Gaithersburg, MD, USA) were processed and analyzed in parallel with samples to determine the digestion of each blank and to provide reference material recovery. The recovery values for all heavy metals were between 87.0% and 108%, within the error range. In addition, several samples were cut equally into two parts, one was added certain amounts of heavy metals by spiking standard solution, and analyzed as same as the untreated part to calculate the recovery. The recoveries of standard addition were within 85.0% and 103%. Samples were prepared in duplicate. HNO_3 and H_2O_2 were guaranteed reagents, and the water used in the procedure was deionized.

2.3. Bioaccessibility test

Gamble's solution and PBET were employed to simulate the human respiratory and digestive system in this study, respectively. Gamble's