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Contamination and risk assessment (based on bioaccessibility via ingestion and inhalation) of metal(loid)s in outdoor and indoor particles from urban centers of Guangzhou, China

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HIGHLIGHTS

- The average content of Zn was the highest in road dust.
- The average content of Pb was the highest in household AC filter dust.
- Cr, Ni, Hg and Pb Contamination were significantly elevated in residential houses.
- Mobilization of metal(loid)s was significantly correlated from stomach to intestine.
- As was observed as the most risky element via ingestion and inhalation of particles.

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ABSTRACT

Road dust, household air-conditioning (AC) filter dust and $PM_{2.5}$ were collected to investigate the contamination of metal(loid)s (Cr, Mn, Ni, Cu, Zn, As, Cd, Sn, Sb, Hg and Pb) in outdoor and indoor urban environments of Guangzhou. Zinc was found to be the most abundant element in road dust and household $PM_{2.5}$, while the concentration of Pb was the highest in AC filter dust. Enrichment factor (EF) was used to assess the influence of human activity on the contamination of these metal(loid)s. Ingestion and inhalation were the two exposure pathways applied for risk assessment. Physiologically based extraction test (PBET) was used to estimate the oral bioaccessibilities of metal(loid)s in road dust and AC filter dust. Respiratory bioaccessible fraction of metal(loid)s via household $PM_{2.5}$ was extracted with lung simulating solution. Household AC filter dust was more hazardous to human health than road dust, especially to children. Arsenic was found to be the most risky element based on the risk assessment.

1. Introduction

As the sink and source of pollution, urban deposits (road dusts, gully sediments and tunnel ceiling dust) and urban soils are good indicators of heavy metal accumulation in the urban surface environment. In addition to outdoor dust, metal(loid)s contained in indoor dust and airborne particles should be taken into consideration as well when evaluating urban metal(loid) contamination. The total contents of metal(loid)s in the street dust and household floor dust have been studied extensively around the world (Al-Khashman, 2004; Chattopadhyay et al., 2003; Hassan, 2012; Li et al., 2001; Shi et al., 2011). Recently, research has extended to the investigation of re-suspended dust on building surfaces (roofs and window sills) (Kong et al., 2011) and indoor air-conditioner (AC) filter dust (Huang et al., 2012; Kang et al., 2010, 2011; Wang et al., 2013). AC filter dust is referred to as the particles that settle on the filter of air conditioner through air current effect. The AC filter dust is hypothesized to be less than 100 μm in particle size, which can be re-suspended from settled dust. The re-suspended particles will adhere onto the surface of food, skin, toys and furniture and be ingested by humans, especially by children (Butte and Heinzow, 2001). However, the fine portion of particles may pass through the AC filters rather than settle on it. Hence, the particles settling on the AC filter represent the coarse portion of re-suspended dust particles more than the fine portion.

Dust plays an important role on human health due to the complex chemistry and the possibility of re-emission. In particular, the indoor particles can be much more hazardous to human health, as people usually spend more than 70% of their time indoors (Maertens et al., 2004).

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Long-term exposure to particles contaminated with metal(loid)s may pose human health risks, such as renal dysfunction (Staessen et al., 1994), osteoporosis, incidence of bone fractures (Staessen et al., 1999) and lung cancer (Verougstraete et al., 2003).

Health risk assessment of exposure to total contents of metal(loid)s in soil and urban dust has been attempted in many different cities, such as Luanda, Angola (Ferreira-Baptista and De Miguel, 2005), Beijing, China (Khan et al., 2008), Hulodao, China (Zheng et al., 2010) and Shanghai, China (Shi et al., 2011). In general, these studies identified oral ingestion as the most critical exposure route to coarse dust particles for humans, compared with inhalation and dermal contact. The coarse particles can be directly swallowed or finally reach the gastro-intestinal tract after a short stay in tracheal and bronchial regions through inhalation (Butte and Heinzow, 2001). However, few of the studies mentioned above employed bioaccessibility to risk assessment.

Physiologically based extraction test (PBET), an in-vitro gastrointestinal method, has been extensively used to estimate the oral bioaccessibility of metal(loid)s in the stomach and intestinal tract (Turner and Hefzi, 2010; Turner and Ip, 2007). The oral bioaccessible fraction of metal(loid)s, referred to as the amount that is released during gastro-intestinal digestion thereby available for absorption, could be employed to estimate the daily intakes and allow risks to be characterized (Turner and Ip, 2007). Different from coarse particles, more than 80% of the particles smaller than 2.5 μm can reach the pulmonary alveoli after inhalation, where they can be deposited and stay for months to years (Falta et al., 2008). Accordingly, the lung serum stimulant should be selected to extract the potential respiratory bioaccessible fraction of metal(loid)s contained in $PM_{2.5}$ (Hodgson et al., 2002; Voutsa and Samara, 2002).

The Pearl River Delta (PRD) region is one of the largest metropolitan regions in south China, where the mega cities and a number of recently established urban centers are located with concentrated stationary and mobile pollution sources (power plants, factories and traffic emissions) (Shao et al., 2006; Zhang et al., 2008). Guangzhou (22″26′–23″56′N, 112″57′–114″03′E) is one of the most densely populated city in the PRD region, with a population of over 10 million and an area of 7545 km^2 (Zheng et al., 2011). The contaminations of Cu, Zn, Cd and Pb in the urban deposits of Guangzhou are evident (Duzgoren-Aydin et al., 2006). However, there is a lack of information of other trace elements contained in urban dust from Guangzhou, such as As, Cr, Sn and Sb, as these elements may pose potential health risks to residents. In houses, air conditioners are widely used during the summer, and therefore AC filter dust would provide abundant information about the indoor atmosphere and environment. Furthermore, the indoor airborne particles, such as $PM_{2.5}$ levels, in urban sites of Guangzhou have been found to frequently exceed the US National Ambient Air Quality Standard (NAAQS) (35 μg/m^3) (Huang et al., 2012; Li et al., 2005). Therefore, the contamination of metal(loid)s in such a high level of indoor $PM_{2.5}$ from Guangzhou has become a public health concern (Wang et al., 2006a, 2006b).

With the background mentioned above, the present study aims to give a holistic assessment of contamination and risk of metal(loid)s contained in urban dust and airborne particles from the Guangzhou urban area. More specifically, the objectives include: (1) investigating contamination and distribution of metal(loid)s (Cr, Mn, Ni, Cu, Zn, As, Cd, Sn, Sb, Hg and Pb) in road dust, household AC filter dust and $PM_{2.5}$ collected from the Guangzhou urban area; (2) evaluating the bioaccessibility of these metal(loid)s via ingestion and inhalation; and (3) applying the bioaccessibilities to estimate chronic daily intakes (DIs) of metal(loid)s via ingestion and inhalation, and characterize their risks.

2. Materials and methodology

2.1. Sampling and sample preparation

A total of thirty road dust samples, seven household $PM_{2.5}$ samples and ten household AC filter dust samples were collected in the Guangzhou urban area from July to the end of August, 2010 (Fig. S1). The road dust samples were collected from different locations, including scenic parks ($n = 3$), educational sites ($n = 6$), residential sites ($n = 9$), heavy traffic sites ($n = 6$), commercial sites ($n = 3$) and peri-urban district ($n = 3$). Ten out of thirty road dust samples were collected specifically near the sampling houses for AC filter dust. The household AC filter dust was collected using a 3M™ membrane. In brief, the 3M™ membrane, which was fixed on the inside of the AC filter during sampling, was used to collect the household re-suspended dust particles passing through the AC filter along with the indoor air current (Fig. S2). In order to collect enough dust samples, the sampling period lasted for 6–8 weeks for each house. The AC operation time was at least 10 h per day during the sampling period. The 3M™ membranes were desiccated and weighed before and after sampling respectively as the $PM_{2.5}$ membranes. Fig. S3 shows the illustrative diagram of split air-conditioner applied in the present study, in which the indoor system and outdoor system are separated. In order to compare the coarse particles with the fine ones in household environment, household $PM_{2.5}$ samples were additionally collected from seven out of all the sampling houses. The information of each sampled house is listed in Table S1. The sampling program, weighing methods for $PM_{2.5}$ membrane and AC filter dust, and sampling quality controls had been described in detail in our previous paper (Huang et al., 2012), in which household AC filter dust was mentioned as household total suspended particulate matter (TSP) with the diameters less than 100 μm. Hence, the road dust samples were sieved to 100 μm ahead of analysis so as to compare with the AC filter dust samples.

2.2. Extraction and determination

About 0.2 g road dust, a strip of 1″ × 8″ from the 8″ × 10″ 3M™ filter collecting dust from air conditioners (US EPA, 1999) and a half of $PM_{2.5}$ membrane were respectively microwave digested with 65% HNO_3 (US EPA, 1994) for the extraction of total contents of metal(loid)s. The digestion solutions were then centrifuged, filtrated with 5C Whatman filter paper and 0.45 μm syringe filter, and diluted with Milli-Q water. The diluted solution was determined using ICP-MS (Perkin Elmer Elan 9000) for all of metal(loid)s, except for Hg, which was determined using atomic spectrometry (FIMS 100 Perkin Elmer).

Physiologically based extraction test (PBET) was employed to evaluate the oral bioaccessibility of metal(loid)s, simulating the chemical conditions of the gastro-intestinal tract. The procedure adopted in this study was based on previous studies with slight modification (Ruby et al., 1996; Turner et al., 2009). In brief, gastric solution was prepared as in Table S2 immediately before use. About 0.25 g of road dust and a strip of 1″ × 8″ from the 8″ × 10″ AC filter were respectively added to 50 mL polyethylene tubes with 40 mL gastric solution and end to end shaken at 37 °C. After 1 h, 8 mL of the mixtures (stomach phase) was abstracted, centrifuged (37 °C, 2000 rpm, for 10 min) and filtrated with a 5C Whatman filter paper and a 0.45 μm syringe filter successively. The remaining contents in the reaction tubes were added with 70.0 mg of bile salts and 20.0 mg of pancreatin and adjusted to pH 7.0 with a few aliquots of saturated $NaHCO_3$, then digestion was continued. Next was the extraction of two other 8 mL mixtures after 2 h and 4 h (intestinal phase 1 and intestinal phase 2), and the rest of procedure was carried out as mentioned above. The digested solutions were stored at 4 °C before analysis with ICP-MS (Perkin Elmer Elan 9000) for the metal(loid)s. Hg was determined using atomic spectrometry (FIMS 100 Perkin Elmer).

The lung simulating serum was also prepared immediately before use (Table S2) (Hodgson et al., 2002; Voutsa and Samara, 2002). The other half of $PM_{2.5}$ membrane was extracted with 10 mL serum by shaking at 37 °C. After filtrations with 5C Whatman filter paper and 0.45 μm syringe filter, the extracted solutions were stored at 4 °C before analysis. Except for Hg, which was determined using atomic spectrometry (FIMS 100 Perkin Elmer), other metal(loid)s were determined with ICP-MS