



Bioaccessibility and health risk assessment of arsenic in soil and indoor dust in rural and urban areas of Hubei province, China

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ABSTRACT

Incidental oral ingestion is the main exposure pathway by which human intake contaminants in both soil and indoor dust, and this is especially true for children as they frequently exhibit hand-to-mouth behaviour. Research on comprehensive health risk caused by incidental ingestion of both soil and indoor dust is limited. The aims of this study were to investigate the arsenic concentration and to characterize the health risks due to arsenic (As) exposure via soil and indoor dust in rural and urban areas of Hubei province within central China. Soil and indoor dust samples were collected from schools and residential locations and bioaccessibility of arsenic in these samples was determined by a simplified bioaccessibility extraction test (SBET). The total arsenic content in indoor dust samples was 1.78–2.60 times that measured in soil samples. The mean As bioaccessibility ranged from 75.4% to 83.2% in indoor dust samples and from 13.8% to 20.2% in soil samples. A Pearson's analysis showed that As bioaccessibility was significantly correlated with Fe and Al in soil and indoor dust, respectively, and activity patterns of children were utilised in the assessment of health risk via incidental ingestion of soil and indoor dust. The results suggest no non-carcinogenic health risks ($HQ < 1$) or acceptable carcinogenic health risks ($1 \times 10^{-6} < CR < 1 \times 10^{-4}$) in all studied locations. Indoor activities comprised between 64.0% and 92.7% of the total health risk incurred during daily indoor and outdoor activities. The HQ and CR values for children in urban areas were 1.59–1.95 times those for children in rural areas. The HQ and CR values for children three to five years of age were 1.40–1.47 times those for children six to nine years of age. The health risk accounting for bioaccessibility was only 50.8–59.8% of that obtained without consideration of bioaccessibility.

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1. Introduction

Soil and indoor dust are important components in the living environment of the general population in both rural and urban areas, and among all environmental materials, they are the most significant in terms of human health effects (Ibanez et al., 2010). Indoor dust is a heterogeneous mixture of particles derived from numerous sources, including soil particles, atmospherically deposited particulates, cooking and heating residues, paint particles, fibres, skin particles, and building and furnishing materials from both outdoor and indoor environments (Paustenbach et al., 1997, 2000). Concentrations of many metals and metalloids in indoor

dust are commonly higher than those in soil in ordinary urban environments (Ibanez et al., 2010). Arsenic (As) is a carcinogenic metalloid found in soil and dust and may pose a risk to human health. Exposure to As may result in serious harmful effects to humans, such as skin lesions, cardiovascular diseases and metabolic disorders. As in soil and indoor dust could enter the human body through dermal contact, inhalation, and ingestion (Reeder et al., 2006). Ingestion is the most important exposure route and may account for 90% of all arsenic entering the human body (Ruby and Lowney, 2012). Compared with the incidental soil ingestion rate of adults, the rate among children is higher; children intake more soil and indoor dust each day as a consequence of their frequent playing on the ground and frequent hand-to-mouth and object-to-mouth behaviours during outdoor and indoor activities (Fillol et al., 2013). Additionally, the less developed enzymatic metabolism and lighter body weight of children makes children more sensitive to arsenic poisoning than adults (Carrizales et al., 2006; Ramirez-Andreotta et al., 2013; Zhang et al., 2013).

Total As concentrations in soil/indoor dust have generally been

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utilised to determine potential As exposure in previous human health risk assessments, under the assumption that the entire amount of As present in the soil/indoor dust is available for uptake into the gastrointestinal tract. However, whether As poses a human health risk depends on the potential of As to dissolve in gastrointestinal fluid, reach the central (blood) compartment and cause adverse health effects. Therefore, the potential health risks may be overestimated by assessments based on total As concentrations in soil/indoor dust. As bioaccessibility, defined as the fraction of an administered dose that is soluble in the gastrointestinal environment and available to be absorbed by the gastrointestinal tract (Ruby et al., 1999), is an essential measure for more accurate assessments of health risks associated with oral ingestion of soil and dust. Bioaccessibility is calculated by in vitro simulations of digestive fluid using different extractants (Wragg and Cave, 2002). Many in vitro methods are developed to mimic the biochemical conditions in the human gastrointestinal tract (Bruce et al., 2007; Schroder et al., 2004). The physiologically based extraction test (PBET), for instance, is essentially a two-stage sequential extraction using various enzymes to simulate both gastric and small intestine compartments. A simplified bioaccessibility extraction test (SBET) is a simplified form of the PBET and has been used extensively to estimate the bioaccessibility of metals and metalloids in soil exposure studies (Das et al., 2013; Juhasz et al., 2007). This method uses a one-step extraction to simulate gastric conditions, making it a time efficient and reproducible method that is therefore more applicable to large batches of samples (Kelley et al., 2002; Wragg and Cave, 2002).

Soil and indoor dust have been extensively investigated separately on exposure and health risk assessments via ingestion route, for instance, soil contaminated by human industrial activities such as mining, smelting and coking (Cao et al., 2014; Lin et al., 2014; Martinez-Sanchez et al., 2013), and urban soil/dust contaminated due to the extensive effects of traffic and living activities (Hu et al., 2011; Li et al., 2014b; Xia et al., 2011). Indoor dust and normal soil (such as soil in playgrounds and courtyards in residential and school locations) are important exposures for the general population, especially for children, who spend a great deal of time playing on the ground and have high levels of contact with both indoor dust and soil in gardens or playgrounds at schools and nearby homes. Therefore soil and indoor dust are both important components to As ingestion exposure, and health risk assessment based on soil or indoor dust separately cannot represent the real health risk caused by incidental ingestion properly. Unfortunately, few studies paid attention to the comprehensive health risk caused by ingestion of both soil and indoor dust.

This study is focused on the soil and indoor dust in residential locations and schools where children mainly live and play in rural and urban areas in Hubei Province, central China. The main objectives of this study are to: (1) quantify both the total and the bioaccessible As (determined by SBET method) in the soil and indoor dust of residential and school locations in rural and urban areas; (2) explore the relationships between total As content, bioaccessible As content, As bioaccessibility and physicochemical properties of soil/indoor dust to identify factors which may influence the bioaccessibility of As in soil and indoor dust; and (3) estimate comprehensive potential health risks due to As exposures via incidental ingestion of soil and indoor dust by children during daily indoor and outdoor activities.

2. Materials and methods

2.1. Study sites and sampling

The study sites are situated in Yuyang Guan and Wuhan

(typical rural and urban areas, respectively) in Hubei province of central China. Yuyang Guan is a typical rural town in north western Hubei province located between 30°03' N and 30°15' N and between 110°08' E and 111°08' E and experiences an average annual precipitation and temperature of 1215 mm and 16.9 °C, respectively. Wuhan is the capital city of Hubei province located between 29°58' N and 31°22' N and between 113°41' E and 115°05' E and experiences an average annual precipitation and temperature of 1269 mm and 16.6 °C, respectively.

This study investigated children from three to nine years old. A questionnaire-based survey was conducted in the studied areas to determine body weight, daily activity patterns, and lifestyle of the local children. A total of 164 questionnaires in the rural area and 180 questionnaires in the urban area were collected.

Soil and indoor dust samples were collected from 25 houses and three schools in the rural area and 26 houses and three schools in the urban area. For soil and indoor dust samples, outdoor (gardens, playgrounds, etc.) and indoor (bedroom, classroom, etc.) areas that may have been routinely contacted by children were chosen as sampling sites. In each soil sampling site, four to five locations (about 2 m distance) were selected and soil samples were collected by a columnar sampler and then mixed together to composite one sample (about 1.0 kg). In each indoor dust sampling site, settled dust in 4–5 m² floor area was collected by a polyethylene brush and then mixed together to composite one sample (about 5.0 g). A total of 79 soil samples and 43 indoor dust samples were collected. Collected soil and indoor dust samples were air dried in shade at room temperature, sieved (< 250 µm), and then refrigerated at 4 °C until analysis. The total and bioaccessible As contents in soil and indoor dust samples were analysed using the < 250 µm particle size fraction, as soil and indoor dust fractions < 250 µm closely represent the soil particles that would be expected to adhere to children's hands and thus be more available for accidental ingestion as a consequence of hand-to-mouth contact behaviour (Acosta et al., 2009; Ruby and Lowney, 2012).

2.2. Physicochemical properties of soil and indoor dust

The pH of each soil sample was measured in a 1:2.5 suspension of soil in pure water. Soil organic matter (SOM) concentrations were measured based on the weight loss upon combustion to 400 °C (Ben-dor and Banin, 1989). Soil samples were heated at 105 °C for 24 h, were cooled and weighed and were then ignited in a muffle furnace at 400 °C for 16 h before being cooled and weighed again. The weight of the organic matter was calculated by subtraction, and the SOM content of soil samples was calculated as Eq. (1):

$$\text{SOM}(\%) = \frac{\text{Weight}_{105} - \text{Weight}_{400}}{\text{Weight}_{105}} \times 100\% \quad (1)$$

where Weight₁₀₅ is the weight of the soil sample after heating at 105 °C, and Weight₄₀₀ is the weight of the soil sample after ignition at 400 °C.

Soil and indoor dust samples were digested with HNO₃–HF–HClO₄ following the method described in detail by Lin et al. (2008). Total Al, Fe, and Mn in the extracts was measured with inductively coupled plasma-atomic emission spectrometry (ICP-AES, IRIS Intrepid II, Thermo Fisher Co. Franklin, MA, U.S.), while total As was measured with liquid chromatography atomic fluorescence spectrometry (LC-AFS, LC-AFS9780, Haiguang Co., Beijing, China). The concentration (C, kg /kg) of As, Al, Fe and Mn in soil and indoor dust was calculated as Eq. (2):

$$C = \frac{c \times V}{m} \quad (2)$$

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