



Contamination, bioaccessibility and human health risk of heavy metals in exposed-lawn soils from 28 urban parks in southern China's largest city, Guangzhou



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ABSTRACT

The total concentrations and oral bioaccessibility of heavy metals in surface-exposed lawn soils from 28 urban parks in Guangzhou were investigated, and the health risks posed to humans were evaluated. The descending order of total heavy metal concentrations was Fe > Mn > Pb > Zn > Cu > Cr > Ni > Cd, but Cd showed the highest percentage bioaccessibility (75.96%). Principal component analysis showed that Grouped Cd, Pb, Cr, Ni, Cu and Zn, and grouped Cr and Mn could be controlled two different types of human sources. Whereas, Ni and Fe were controlled by both anthropogenic and natural sources. The carcinogenic risk probabilities for Pb and Cr to children and adults were under the acceptable level ($<1 \times 10^{-4}$). Hazard Quotient value for each metal and Hazard Index values for all metals studied indicated no significant risk of non-carcinogenic effects to children and adults in Guangzhou urban park soils.

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1. Introduction

Rapid urbanization is a worldwide phenomenon (Seto et al., 2010; UN, 2013). Urban environments thus become supremely important pertaining to human health and wellbeing (Liu and Diamond, 2005; Tzoulas et al., 2007; Hänninen et al., 2014). Anthropogenic activities related to industry and economy are usually more concentrated in urban areas, and then cities have become the geographic focus of resource consumption and chemical emissions, which may cause many problems including environmental pollution (Alloway, 1995; Li et al., 2001; Luo et al., 2012b; Gu et al., 2014a). Heavy metals are typical contaminants in urban environment, and consequently useful indicators of environmental pollution (Manta et al., 2002; Sun et al., 2010; Gu et al., 2012a). Metals may originate from many different sources in urbanized areas, such as vehicle emissions, industrial discharges and other activities (Li et al., 2001; Harrison et al., 1981). As a crucial component of urban ecosystems, urban soils are subject to a

continuous accumulation of metals; their generally long residence time enables metals to remain in the urban soils for lengthened period, and then may become a source for further urban environmental pollution under such dynamic conditions, as runoff, re-suspension, leaching, and weathering (Shi et al., 2008; Guney et al., 2010; Yang et al., 2014).

Metals in urban soils can be easily transferred into humans through ingestion, inhalation, and dermal contact and will pose a health risk to urban residents, especially the most susceptible children and senior citizens who use parks more often (Miguel et al., 1997; Wei and Yang, 2010; Xia et al., 2011; Peña Fernández et al., 2014). Numerous studies have demonstrated that metals could accumulate in the fatty tissues, subsequently affect the functions of the organs, and disrupt the nervous system or endocrine system (Waisberg et al., 2003; Bocca et al., 2004; Wang, 2013). In contrast to agricultural soils which mainly influence human health indirectly through food chain, urban park soils can cause direct exposure for children through hand-to-mouth oral ingestion during outdoor recreational activities, and therefore it is significant to quantify the oral exposure risk levels of park soil contamination for urban park users (Ljung et al., 2007; Guney et al., 2010; Luo et al., 2012a).

The actual health risks of metals in ingested soil depend strongly

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on the fraction that is soluble in the gastrointestinal tract available for absorption, so that only a fraction of the total soil metals is human accessible (Oomen et al., 2002; US EPA, 2007; Hu et al., 2011). The physiologically based extraction test (PBET) and the simple bioaccessibility extraction test (SBET) have been successfully developed and verified to estimate the oral bioaccessibility of soil metals (Ruby et al., 1993; Oomen et al., 2002; US EPA, 2007; Sialelli et al., 2010; Luo et al., 2012a). SBET is a simplified form of PBET designed for fast, easy, and reproducible extraction test, and a standard operational protocol of SBET has been set up to investigate *in vitro* lead bioavailability (Wragg and Cave, 2003; US EPA, 2007; Hu et al., 2011). In order to assess the potential health risks associated with contaminant exposures from soils and soil-like materials, human health risk assessments have also been developed (US EPA, 1989, 2000, 2001). These tests have recently been used and have proven to be valid and useful (Granero and Domingo, 2002; Guney et al., 2010; Mingot et al., 2011).

Guangzhou (Canton), the capital city of Guangdong Province, is a major industrial and economic centre in South China (Liu et al., 2015). This largest city in South China has a total area of 7434.4 km², a history of more than 2200 years and a population over 10 million (GZG, 2015), and is one of the most important cities in Pearl River Delta, the fastest growing economic district in China. Several researchers have investigated the total heavy metal concentrations in urban soils of Guangzhou, and their results showed that contamination by some heavy metals has occurred in the urban soils (Guan et al., 2001; Lee et al., 2007; Lu et al., 2007; Cai et al., 2013). However, to the best of our knowledge, the oral bioaccessibility and health risk of heavy metals in urban park soils of Guangzhou remain unknown. Therefore, this study was: (1) to estimate the oral bioaccessibility of metals (Cd, Pb, Cr, Ni, Cu, Zn, Fe and Mn) in urban soils using SBET; and (2) to assess their carcinogenic and non-carcinogenic health risks.

2. Materials and methods

2.1. Study area

Guangzhou (112°57'–114°3' E, 22°26'–23°56' N), the capital and a political, economic, and cultural center of Guangdong Province, is situated in the south China and in the north of the Pearl River Delta (Fig. 1). It is bordering on the South China Sea, adjacent to Hong Kong and Macao (GZG, 2015). As located on the north margin of tropics, the average temperature in Guangzhou is 22.8 °C and the annual rainfall at the urban area is over 1600 mm (GZG, 2015). The Pearl River, the second largest river of China, crosses the central urban area (Fig. 1).

2.2. Field sampling

Urban park soils of exposed lawns, the major playing and recreational areas for children and adults, were collected in May 2014 from 28 public parks of Guangzhou city (Fig. 1 and Fig. S1–S3). Depending on the size of the exposed-lawns, 15–50 sub-samples of the topsoil (0–5 cm) were collected and mixed thoroughly to get a representative sample. All the mixed samples collected were dried in an oven at 40 °C until constant weight, ground gently with agate pestle and mortar, sieved with 150 µm mesh nylon sieve for homogenization, and then placed in black self-sealing polyethylene bags until analyses for organic matter (OM), CaCO₃ and metals.

2.3. Analytical methods

Samples were pretreated for particle size analysis in accordance with the Chinese National Standard (GB/T12763.8–2007). The

sample granulometry was analyzed through a Malvern Mastersizer 2000 laser diffractometer with a measurement range between 0.02 and 2000 µm. The percentages of the following three size ranges were determined; <4 µm (clay), 4–63 µm (silt), and >63 µm (sand). The OM and inorganic carbonate (CaCO₃) were determined by the loss-on-ignition technique. Specifically, the sample was heated at 550 °C for 4 h to estimate OM and for a further 2 h at 1000 °C to estimate CaCO₃ content (Rabenhorst, 1988). The pH of park soils was measured using a soil to ultrapure water suspension of 1:2.5 (W/V) (Nanos and Rodríguez Martín, 2012). Total metal microwave digestion followed the EPA Method 3050B with an Ethos Plus Microwave Labstation (Milestone Inc.).

Simple bioavailability extraction test (SBET) procedure following Ruby et al. (1993) was conducted to elucidate the metal bioaccessibility, and the detailed protocols adopted in this study have been described previously (Oomen et al., 2002). Metal concentrations were determined by atomic absorption spectrophotometry (AAS, Z2000, Hitachi).

2.4. Human health risk model

In order to quantify both carcinogenic and non-carcinogenic risk from ingesting contaminated soils by children and adults, the average chemical daily intakes of metals and risk characterization have been implemented by computing carcinogenic risk (CR) for Pb and Cr, as well as hazard quotient (HQ) for Pb and Cr (VI) and other metals by using respective cancer slope factor and reference dose values from US EPA (US EPA, 2007, 2010; Hu et al., 2011; US DOE, 2011). For exposure assessment model, chemical daily intake (CDI) of metals from incidental ingestion of soil was calculated as the following formula:

$$CDI = C_{soil} \times \frac{EF \times ED \times IR}{BW \times TA} \times 10^{-6}$$

where CDI is chemical daily intake (mg kg⁻¹ day⁻¹); C_{soil} is metal concentration in urban park soils; EF is the exposure frequency of 350 days year⁻¹ (US DOE, 2011); ED is exposure duration: in this study 6 years for children and 30 years for adults (US DOE, 2011); IR is the ingestion rate at 200 mg soil per day for children and 100 mg per day for adults (US DOE, 2011); BW is the average body weight: 16.2 kg for children and 61.8 kg for adults (Luo et al., 2012a); TA is averaging time (for noncarcinogens, TA = ED × 365; for carcinogens, TA = 70 × 365 = 25,550 days).

The potential carcinogenic and non-carcinogenic risks for individual metals were estimated with the US EPA (2007) protocol using the following equations:

$$\text{Carcinogenic Risk (CR)} = CDI \times RBA \times CSF$$

$$\text{Hazard Quotient (HQ)} = (CDI \times RBA) / RfD$$

Cancer slope factor (CSF) and oral reference doses (RfD) were achieved guidelines from the US government related to environmental protection (US EPA, 2010; US DOE, 2011). Accordingly, the values of CSF were 8.5E-03 for Pb and 5.00E-01 for Cr (US DOE, 2011); the values of RfD were 5.00E-04 for Cd, 3.50E-03 for Pb, 3.00E-03 for Cr, 2.00E-02 for Ni, 4.00E-02 for Cu, 3.00E-01 for Zn, 7.00E-01 for Fe and 2.4E-02 for Mn (US EPA, 2010, 2013). Relative bioavailability adjustment (RBA) was the ratio of metals' concentrations extracted using SBET method to their total concentrations in soils. The estimated value of Carcinogenic Risk (CR) is the probability for an individual to develop cancer over a lifetime exposure to carcinogenic hazards. The acceptable or tolerable risk for regulatory purposes falls between 1×10^{-6} and 1×10^{-4} . The

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