Science of the Total Environment 521–522 (2015) 144–151

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Concentration and transportation of heavy metals in vegetables and risk assessment of human exposure to bioaccessible heavy metals in soil near a waste-incinerator site, South China

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HIGHLIGHTS

- Similar pattern of heavy metal in settled air particle and aerial parts of vegetables
- Leaf lettuce accumulated the highest heavy metals concentrations
- The bioaccessibility of heavy metals in soil ranged from 2% to 71.78%.
- Cd and Pb in soil resulted in the highest non-cancer risk.
- Cd resulted in unacceptable cancer risk for children and risk.

ARTICLE INFO

Article history:
Received 6 December 2014
Received in revised form 8 March 2015
Accepted 20 March 2015
Available online 30 March 2015

Editor: Charlotte Poschenrieder

Keywords:
Waste incinerator
Heavy metals
Bioaccessibility
Risk assessment

ABSTRACT

There is limited study focusing on the bioaccumulation of heavy metals in vegetables and human exposure to bioaccessible heavy metals in soil. In the present study, heavy metal concentrations (Cr, Ni, Cu, Pb and Cd) were measured in five types of vegetables, soil, root, and settled air particle samples from two sites (at a domestic waste incinerator and at 20 km away from the incinerator) in Guangzhou, South China. Heavy metal concentrations in soil were greater than those in aerial parts of vegetables and roots, which indicated that vegetables bioaccumulated low amount of heavy metals from soil. The similar pattern of heavy metal (Cr, Cd) was found in the settled air particle samples and aerial parts of vegetables from two sites, which may suggest that foliar uptake may be an important pathway of heavy metal from the environment to vegetables. The highest levels of heavy metals were found in leaf lettuce (125.52 μg/g, dry weight) and bitter lettuce (71.2 μg/g) for sites A and B, respectively, followed by bitter lettuce and leaf lettuce for sites A and B, respectively. Swamp morning glory accumulated the lowest amount of heavy metals (81.02 μg/g for site A and 53.2 μg/g for site B) at both sites. The bioaccessibility of heavy metals in soil ranged from Cr (2%) to Cu (71.78%). Risk assessment showed that Cd and Pb in soil samples resulted in the highest non-cancer risk and Cd would result in unacceptable cancer risk for children and risk. The non-dietary intake of soil was the most important exposure pathway, when the bioaccessibility of heavy metals was taken into account.

1. Introduction

With the rapid increase of urbanization, great concerns have been raised regarding the waste incinerators due to their potential impacts on the environment by the toxic chemicals emitted from incomplete combustion (Wang et al., 2008). Waste disposal has become an important factor restricting economic and social sustainable development. After 1988, waste burning was rapidly developed in our country (Chen, 2004). Municipal wastes such as leather waste shavings were likely to contain toxic heavy metals (e.g. cadmium and chromium) The heavy metals were converted to their oxide or chloride forms during combustion process, and the heavy metal compounds were able to volatilize and be carried out of the incinerator device with the hot flue gases due to the high combustion temperature (Louhab and Akssas, 2006; Lu-shi et al., 2004). The burning activities have resulted in high levels of pollution in the ambient environment and have further

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http://dx.doi.org/10.1016/j.scitotenv.2015.03.081
0048-9697/

threatened the local ecosystem and the health of surrounding inhabitants. Emission is one of the important pathways for these chemicals entering the environment. It is known that serious systemic health problems will be caused as a result of excessive accumulation of dietary and non-dietary heavy metals in the human body (Oliver, 1997; Chen, 2004). The prolonged consumption of unsafe concentrations of heavy metals through foodstuffs may lead to the chronic accumulation of heavy metals in the kidney and liver of humans causing disruption of numerous biochemical processes, leading to cardiovascular, nervous, kidney and bone diseases (Jarup, 2003; Ali and Al-Qahtani, 2012). Some heavy metals such as Cu, Zn, and Ni act as micronutrients for the growth of human beings when present in trace quantities, whereas others such as Cd, As, Pb, and Cr act as carcinogens (Feig et al., 1994; Trichopoulos, 1997)

Nowadays, many studies for calculating non-cancer risks from exposure through the ingestion pathway are based on bioaccessible part of pollutants instead of total concentration (Hu et al., 2012; Luo et al., 2012; Read et al., 2015). The total heavy metal concentrations only provide very limited information about their chemical behavior and potential fate (Mench et al. 2006), whereas the bioaccessible fractions give results that are much closer to the actual risks involved. The bioaccessible fraction that can act as a toxicant in humans is the fraction of the ingested dose that crosses the gastrointestinal epithelium and becomes available to be distributed to internal target tissues and organs (Ruby, 1993). The physiologically based extraction test (PBET) is commonly used in the scientific literature to simulate stomach and intestine liquids to evaluate the bioaccessibilities of pollutants in the human gastrointestinal system (Tao et al., 2014).

A number of studies have reported levels of heavy metal contamination in soil, sediment, and biota samples from open burning sites (Cao and Hu, 2000; Nan et al., 2002; Singh et al., 2004; Mahmood and Malik, 2014). Some studies indicated that air particle contributed to heavy metals accumulation by plants (Sapkota and Cioppa, 2012; Schreck et al., 2013). In the present study, we (1) measured the heavy metal concentrations in settled air particles, rhizosphere soils, roots, and vegetables, (2) investigated the contribution of settled air particles and soil to heavy metal uptake in vegetables, and (3) performed risk assessment of human exposure to heavy metals via non-dietary intake of soil considering the bioaccessibility. This provides a basis for guiding further activities aimed at preventing excessive exposure of humans through monitoring and control of amelioration of uptake to plants.

2. Materials and methods

2.1. Sampling

The samples, including soil, vegetables, and settled air particulates, were collected from two sites in Guangzhou in July 2012. There is a subtropical monsoon climate in Guangzhou. Site A is close to (<0.5 km; located in the downward direction of the prevailing wind flow) a domestic waste incinerator. The capacity of incinerator is about 346,660 t/y. The Selective Non-Catalytic Reduction (SNCR) Denitration technology and Fly Ash Solidification technology are employed to treat the flue gas in the waste incinerator. Site B is a less contaminated area (about 20 km away from the incinerator; located in the upward direction of the prevailing wind flow, and not directly exposed to industrial or municipal emissions). Vegetables grown in these two sites are supplied to local markets.

At site A, five representative vegetable species, including leaf mustard (*Brassica juncea*), leaf lettuce (*Lactuca sativa* L.), morning glory (*Ipomoea aquatica* Forsk), lettuce (*Mylopharyngodon*), and bitter lettuce (*Cichorium endivia*) were collected ($n = 45$). At site B, three vegetable species, including leaf mustard (*B. juncea*), morning glory (*I. aquatica* Forsk), and bitter lettuce (*C. endivia*) were collected ($n = 27$). The exposure time and vegetative phase of these vegetables was about 3–4 months. The sample collection was described in detail in our previous work (Zeng et al., 2014). Each rhizosphere soil or vegetable sample was a composite of three subsamples. For rhizosphere soil collection, when the vegetables were removed from the soil, they were gently shaken to remove soil loosely adhering to the roots. The remaining adherent soils were separated from the roots as rhizosphere soil. Three settled air particle samples were collected from the cement surface of a small house roof at each sampling site by a vacuum cleaner. Settled air particles on the cement surface located in the vegetable garden may represent settled air particles on the vegetable leaves. All samples were wrapped with aluminum foil, put into polythene zip-lock bags, and transported to the laboratory. Afterward, the vegetable samples were thoroughly washed with running tap water to remove airborne dust and soil particles. The roots and aerial parts of vegetables were separated. Rhizosphere soil samples were air dried at room temperature and ground in a grinder sufficiently to pass through a 2-mm sieve. Fine roots and aerial parts of vegetables were freeze-dried and ground with agate mortar. The settled air samples were sieved by a 100-μm mesh. All samples were kept frozen at −18 °C until analysis.

2.2. Determination of heavy metals

For all samples, 0.5 g of dried sample was digested with 15 mL of HNO_3 and HCl with 1:1 ratio at 90 °C until a transparent solution was obtained. The solution was filtered through Whatman No. 42 filter paper and the filtrate was diluted to 50 mL with distilled water.

The concentrations of heavy metal in all samples were determined with an atomic absorption spectrophotometer (Perkin-Elmer model 2130, USA) fitted with a specific lamp of a particular metal using appropriate drift blanks. Quality control measures were taken to assess the contamination and reliability of data. Blank and drift standards were run after three determinations to calibrate the instrument. The coefficient of variation of replicate analysis was determined for different determinations and for precision of analysis. Variations were found to be less than 10%. Precision and accuracy of analysis were also ensured through repeated analysis of samples, and standard reference material of the vegetables (GBW10015: spinach) and soils (GBW07430 (GSS-16): soil from Pearl River Delta) for all the heavy metals. The recovery of heavy metals in standard reference material of vegetables and soils ranged from 82% (Cd) to 114% (Pb).

Table 1
Heavy metal concentrations (μg/g dry weight) of soil, aerial parts of vegetables and roots from site A.

Samples		Cr	Cd	Pb	Cu	Ni
Soil	Mean	285.07	62.09	14.95	13.53	10.09
	Median	283	63.45	15	13.8	9.1
	Range	215–358	45.55–91	11.87–21.78	7.09–21.36	8–15.04
Aerial parts of vegetables	Mean	55.68	11.69	27.65	7.65	24.72
	Median	39.5	7	29.55	7.42	22.8
	Range	24.50–60	3.25–15.75	9.3–45.8	3.92–11.67	7.8–38.55
Roots	Mean	41.4	8.52	19.66	33.41	9.77
	Median	52.71	9	16.75	35.87	9.84
	Range	19.71–68.04	7–19.5	2.75–33.25	5.87–62.12	4.84–15.84