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Concentrations and human health implications of heavy metals in market foods from a Chinese coal-mining city

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ABSTRACT

Concentrations of heavy metals (As, Cd, Co, Cr, Cu, Hg, Pb and Sb) in vegetables, meat and fish purchased from traditional agri-product markets in Huainan, China, were measured. Concentrations of the eight metals in most of the measured samples were lower than their respective maximum allowable concentrations (MACs), except for Pb, Cd, Cr and Cu in some of the samples exceeded safe limits. Based on local food consumption, the intake of individual metals was estimated to be less than their respective recommended limits. However, the overall target hazard quotient (THQ) for the eight metals was 1.07 based on the digestion of leafy vegetables and 2.12 based on the consumption of all of the investigated foods. The results of this study suggest that the overall risk from exposure to multiple metals in foods should be of concern, even though low-to-moderate heavy metal pollution is present in foods from Huainan markets.

1. Introduction

Heavy metal concentrations in foodstuff and their associated population health risks have drawn increased attention globally. It is well known that many heavy metals, such as arsenic, lead and mercury, are hazardous to human health, even at trace concentrations (Abdul et al., 2015; Zhou et al., 2016). Other metals (e.g. Cu, Ni and Zn) are essential for maintaining human health, but excessive intake can cause several clinical and physiological problems (Valko et al., 2005; Brewer, 2010). Although the relative contributions for heavy metal intake have not been clearly established, dietary intake is considered to be the critical exposure pathway, accounting for >90% of exposure, with inhalation and dermal contact the other exposure routes (Kachenko and Singh, 2006; Loutfy et al., 2006; Islam et al., 2015a). As such, concentrations of heavy metals in vegetables, fish and meat are likely of significant public concern because the safety of these foodstuffs is essential.

Heavy metal contamination in vegetables, fish and meat occurs through many pathways. Anthropogenic activities, including mining, industrial processing and the use of large quantities of agrochemicals, such as metal-based pesticides and fertilizers, are the main sources of heavy metal pollution in the environment (Islam et al., 2015a; Liu et al., 2016). These metals cannot be degraded or destroyed, although they can change their chemical forms. Once the heavy metals are dispersed into water, soil and air, plants and animals can accumulate them. Recently, surveys and monitoring programs focused on heavy metal accumulation in foodstuffs have been carried out in many countries and a wide range of concentrations were found in fish, rice, vegetables and meat that were associated with differing health risks (Castro-González and Méndez-Armenta, 2008; Yang et al., 2011; Fang et al., 2014; Islam et al., 2015a,b; Cheng et al., 2016; Gu et al., 2016; Pan et al., 2016; Zhou et al., 2016). Compared with developed countries, China may have higher health risks from heavy metals in vegetables and fish because of locally severe and regionally significant environmental contamination associated with the rapid development of mining and manufacturing industries. Elevated concentrations of and higher health risks from metals were reported in the vegetables and fish from some polluted areas of China (Feng and Qiu, 2008; Luo et al., 2011; Liu et al., 2013). Lead and cadmium concentra-

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tions greater than the Chinese safety and quality requirements for pollution-free vegetables were found in all investigated vegetable samples from the Daye mining area in China (Sun et al., 2013). It is reasonable to hypothesize that there are higher potential health risks to consumers from exposure to metals in vegetables, fish and meat that originate from mining areas.

China has more than 120 mining cities, 55 of which are coal-mining cities (Lou and Gu, 2005). Elevated levels of heavy metals have been widely reported in the surrounding environment of coal-mining cities; such contamination is likely due to a lack of environmental protection and pollution control technology for mining activities (Liu et al., 2015; Pang et al., 2016). Studies have reported heavy metal pollution was present in the vegetables and fish from the coal mines and their surrounding areas (Zhang et al., 2008; Wang et al., 2012; Fang et al., 2015; Cheng et al., 2016). The non-carcinogenic risk from exposure to some metals in local vegetables exceeded the acceptable levels (Fang et al., 2015; Cheng et al., 2016). Most researchers have focused on field-grown vegetables or wild fish in limited areas; analyses of heavy metals in market-sold foods are scarce. Specific surveys of heavy metal concentrations in marketed foodstuffs in coal-mining cities are limited. In these areas, heavy metals can enter into the marketed foodstuffs by a variety of ways (Fig. 1). But there have been no reports about the health risk of consuming fish and meat purchased from traditional agri-product markets.

In this study, we used Huainan, a coal-mining city in eastern China, as a case study. Our main aim is to: (1) determine the concentrations of heavy metals in vegetables, fish and meat purchased from markets in Huainan; (2) calculate, based on questionnaires, the estimated daily intake (EDI) and target hazard quotient (THQ) of heavy metals via consumption of these foodstuffs; and (3) evaluate the potential health risks to local consumers in Huainan. The results will improve our understanding of the extent of heavy metal accumulation in daily consumable foods and serve as a basis for comparison to other regions in China and worldwide.

2. Materials and methods

2.1. Study area and sample collection

Huainan, a prefecture-level city situated in the mid-northern part of Anhui Province, is located at 32°23.3′–33°0.5′ N, 116°21.0′–117°12.5′ E and encompasses a total area of 5571 km^2 and has a population of 3,800,000. It has a warm, semi-humid climate, with the wind direction predominantly from the east, and annual average temperature, precipitation, and wind speed is 15.2 °C, 923–926 mm, and from 1.30 to 2.90 m/s, respectively. Coal-mining activities in Huainan began a century ago. Nine mines are currently active, and an additional 10 plants will be built in the near future. These mines are located mainly in Datong, Xiejiaji, and Panji Districts, and Fengtai County. As one of 14 large coal bases planned to develop by China, Huainan is the main industrial city that depends on coal-based industries in eastern China. In Huainan, raw coal production in 2015 reached 82 million tons (Mt) (HBS, 2016), with coal gangue accounting for 10–15% of coal production (Zhou et al., 2012). Although the levels of all heavy metals (except Cr) in groundwater are safe, elevated levels of heavy metals, especially for Cd, Cr, Pb and Hg, have been frequently reported in soils and surface water from the mines and their surrounding areas (Hu and Gao, 2009; Sun and Li, 2013; Fang et al., 2015; You et al., 2016; Zhang et al., 2016a,b). Huainan is an important production base for vegetables, aquatic foods and meat. Such pollutions have even resulted in metal concentrations in some vegetables or grains that exceed the maximum allowable concentrations (MACs) in food set by China (Wang et al., 2012; Fang et al., 2015; Cheng et al., 2016).

Samples of 11 vegetables, six fish and four types of meat were randomly acquired from traditional agri-product markets in five districts (Tianjia'an, Panji, Xiejiaji, Bagongshan and Datong) of Huainan in September 2014. These traditional agri-product markets, with different scales, were leaders in fresh agri-product retailing in Huainan. The vegetable species in this study included: *Brassica oleracea var. capitata* (cabbage), *Celery coriandrum sativum linn.* (celery), *Brassica oleracea L. var. botrytis L.* (cauliflower), *Ipomoea aquatica Forsk* (water convolvulus), *Spinacia oleracea* L. (spinach), *Capsicum annuum* L. (green pepper), *Cucumis sativus L. var. sativus* (cucumber), *Lycopersicon esculentum Mill.* (tomato), *Solanum melongena L.* (eggplant), *Glycine max (Linn.) Merr.* (soybean) and *Vigna unguiculata (Linn.) Walp* (cowpea). Approximately 0.5 kg of each vegetable was purchased from different vendors at each market and was randomly collected in different plastic bags to ensure that sampling was unbiased. Freshwater fish included: *Aristichthys nobilis* (bighead carp), *Cyprinus carpio* (carp), *Oreochromis spp* (tilapia), *Carassius auratus* (crucian), *Ctenopharyngodon idellus* (grass carp) and *Hypophthalmichthys molitrix* (silver carp). Meat products included: *Gallus domesticus* (chicken), *Bos taurus domestica* (beef), *Sus scrofa domestica* (pork) and *Anatinae* (duck).

2.2. Chemical analysis

All of the samples were washed with distilled water and then cut into small pieces. For fish, only the muscle tissues were collected. Each food sample was dried with filter paper to obtain a fresh weight (fw) and oven-dried at 70 °C to obtain a dry weight (dw). The dried samples were crumbled and pulverized with a porcelain mortar and pestle and then sieved through a 150-mesh (about 100 μm) nylon sieve.

Based on the results of previous investigation on metals in soils and plants from Huainan Coal Mining Area (Sun and Li, 2013; Fang et al., 2015; Cheng et al., 2016; You et al., 2016; Zhang et al., 2016a,b), eight metals, i.e., As, Cd, Co, Cr, Cu, Hg, Pb and Sb in the market foods were selected and determined. A microwave digestion method was based on methods described by Chen et al. (2011). For each sample, 0.500 g of dry homogenized sample was digested under pressure in Teflon vessels with 5 mL HNO_3 and 2 mL H_2O_2 in a microwave digestion system. Digestion conditions for the microwave system were applied as: 2 min for 250 W, 2 min for 0 W, 6 min for 250 W, 5 min for 400 W, 8 min for 550 W, vent: 8 min. After cooling, the solution was filtered and diluted up to 10 mL with 1 mol/L HNO_3. The solution was analyzed, using an inductively coupled plasma mass spectrometer (Thermo, Germany), for Cd, Co, Cr, Cu, Zn, Pb and Sb, and an atomic fluorescence spectrometer (XGY-1011A, China) was used for quantifying As and Hg. Quality control was assured through the analysis of duplicate samples, a reagent blank, procedural blanks, and standard reference materials. Blank concentrations were below their respective detection limits and measurements of standards were generally within ±5%. Repeat measurements produced values within ±10%. The relative standard deviations (RSDs) were generally better than 10%. The analytical procedure was verified by repeated analysis of certified reference material (plants: GSB–2, GSB–4, GSB–5 and GSB–11; animals: GSB–9 and GSB–15). The recoveries of the reference samples for all tested elements were satisfactory and ranged between 97.9 and 117%. The detection limits were: 0.2 μg/kg for As, 1 μg/kg for Co and Cu, 2 μg/kg for Cr and Pb, 0.05 μg/kg for Sb, 0.02 μg/kg for Cd and 0.002 μg/kg for Hg.

2.3. Questionnaire on vegetable consumption

To assess the ingestion rates of the studied foods, a questionnaire-based survey of consumption was performed during the sampling period. Self-administered questionnaires were