



Dietary cadmium intake from rice and vegetables and potential health risk: A case study in Xiangtan, southern China

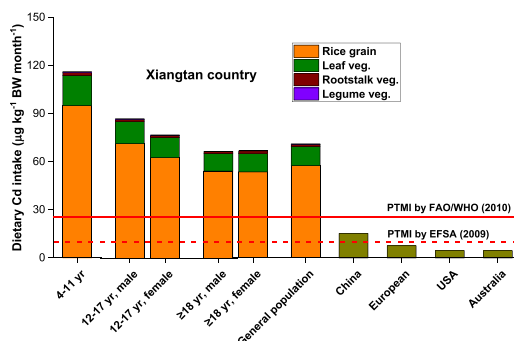
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HIGHLIGHTS

- A paired soil-crop survey was conducted in a county in southern China.
- 82% of the soil samples exceeded the Chinese soil Cd threshold.
- 88% and 29% of rice and vegetable samples exceeded the Chinese limits for Cd.
- Median Cd intake from rice and vegetables was 2.7–4.6 times the FAO/WHO guideline.

GRAPHICAL ABSTRACT



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ABSTRACT

Soil contamination in some areas of southern China has resulted in elevated dietary intake of cadmium (Cd), posing a potential risk to human health. A survey of paired soil-rice ($n = 200$) and soil-vegetable ($n = 142$) samples was conducted in Xiangtan county of Hunan province, southern China. The concentrations of Cd in all the samples were determined by inductively coupled plasma mass spectrometry. Dietary intakes of Cd from the consumption of locally produced rice and vegetables were estimated for different age groups. Among the 342 crop samples collected in the survey, 88% and 29% of rice grain and vegetable samples, respectively, exceeded the Chinese maximum permissible limit for Cd ($0.2 \text{ mg dry weight kg}^{-1}$, $0.2 \text{ mg fresh weight kg}^{-1}$ and $0.1 \text{ mg fresh weight kg}^{-1}$ for rice, leafy vegetables and for rootstalk and legume vegetables, respectively). The median dietary Cd intake varied from 66.5 to $116 \mu\text{g Cd kg}^{-1} \text{ body weight (BW) month}^{-1}$, with children (4–11 years) exhibiting the highest intake. These values are 2.7–4.6 times the tolerable dietary intake of $25 \mu\text{g kg}^{-1} \text{ BW month}^{-1}$ recommended by the Joint FAO/WHO Expert Committee on Food Additives. For the general population in Xiangtan county, rice contributed the majority (81%) of the Cd intake with vegetables contributing only 19%. The median hazard quotient calculated from dietary Cd intake was 2.4 times the permissible level, indicating a high risk to the local residents. This study highlights an urgent need to reduce the transfer of Cd from soil to the food chain in the investigated region.

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1. Introduction

Soil contamination with heavy metals is a major environmental concern worldwide (McLaughlin et al., 1999). Cultivation of crops on contaminated soils can potentially lead to elevated accumulation of heavy

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metals in the edible parts, threatening food safety and human health (Zhuang et al., 2009). According to a recent nationwide survey in China, approximately 19.4% of agricultural soil sites are contaminated (China MEP, 2014). Among various contaminants, cadmium (Cd) ranks the first in the percentage of soil samples (7.0%) exceeding Chinese soil quality limit (SEPA, 1995). Moreover, the concentrations of Cd in soils have increased considerably over the last three decades, at an average rate of $4 \mu\text{g Cd kg}^{-1} \text{ year}^{-1}$, much higher than those reported in Europe (e.g. $0.33 \mu\text{g Cd kg}^{-1} \text{ year}^{-1}$) (Luo et al., 2009; Smolders and Mertens, 2013; Zhao et al., 2015). Soil contamination by Cd was much more serious in southern China, due to mining of base metals and metal smelting and acidic nature of the soils (Zhao et al., 2015; Zhu et al., 2016).

Cadmium is a chronic potent nephrotoxin, as well as a class one carcinogen, and is associated with a range of severe diseases (EFSA, 2012). Cd is readily taken up by plants (McLaughlin and Singh, 1999; Smolders and Mertens, 2013) and is found in most foodstuffs. The Cd intake by general population is from multiple sources, including food, smoking, and drinking water, with the dietary intake accounting for about 90% of the Cd exposure in the non-smoking general population (EFSA, 2009). Foods vary in Cd concentration, but plant-derived foods, such as grain and vegetables, are usually the largest sources for the chronic Cd intake (FAO/WHO, 2015; Olsson et al., 2002; Song et al., 2017; Yang et al., 2017). Epidemiological studies have shown an association between prolonged chronic low Cd exposure and some diseases, including cardiovascular disease, decreased bone density, adverse kidney effects and cancers (Karalliedde, 2012; Riederer et al., 2013). Itai-Itai disease, a bone disease causing fractures and severe pains, is an extreme case of chronic Cd poisoning in humans (Inaba et al., 2005). This disease resulted from the consumption of rice contaminated with Cd, originally from contaminated soil (Nogawa et al., 2004). Residents consuming locally produced foods with elevated Cd concentrations are the most vulnerable to chronic Cd exposure. Dietary Cd intake depends on both the concentrations of Cd in the dietary items and the amounts of consumption. It is also important to consider different age groups because of the variation in the amount of food consumed relative to the body weight. The information on dietary Cd intake is needed to identify high-exposure subgroups and to assess the health risk associated with Cd exposure.

Widespread soil contamination with Cd has been previously reported in Hunan province, southern China (Zhao et al., 2015; Zhu et al., 2016). Locally-produced rice and vegetables are the main staple foods for the local residents in Hunan province. In the present study, we conducted a survey of paired soil-crop samples in Xiangtan county of Hunan province. Based on survey data, dietary Cd intake was calculated for the general and each age-sex subgroup, namely, children aged 4–11 years, young people aged 12–17 years (males or females), and adults over 18 years (males or females) of Xiangtan population and the hazard quotient associated with the dietary Cd intake was also assessed.

2. Materials and methods

2.1. Study area

This study was conducted in the countryside area ($27^{\circ}20' - 28^{\circ}05' \text{ N}$, $112^{\circ}25' - 113^{\circ}03' \text{ E}$) of Xiangtan county, Hunan province, southern China (Fig. 1). The total land area in Xiangtan county is 214,000 ha with 68,500 ha being used for farmland. The region has the subtropical monsoon climate with an annual mean temperature of $16.7 - 18.3^{\circ}\text{C}$ and an annual precipitation of 1300 mm. The county has a population of ca. one million. The majority of the farmland (80%) is used for growing rice (paddy fields), with the remainder being upland for growing vegetables. Typically, two rice crops are grown each year, with irrigation from the Hsiang River or adjacent reservoirs. Although Xiangtan county

is a major crop production region, it is also rich in mineral resources including manganese, vanadium and pyrite mines.

2.2. Soil and plant sampling

Paired soil-rice ($n = 200$) and soil-vegetable ($n = 142$) samples were collected from Xiangtan county when rice and vegetables were ready for harvest during October 2016. The sampling sites were evenly distributed in the paddy and upland fields across the county (Fig. 1). Each soil and plant sample consisted of five sub-samples evenly distributed within a sampling site ($100 - 200 \text{ m}^2$ for rice and $10 - 20 \text{ m}^2$ for vegetable). At each sampling site, ca. 500 g of soil sample was bulked from top 20 cm soil layer, and ca. 500 g of rice grain or ca. 500 g edible part of each vegetable collected from the field.

Among 142 vegetable samples collected, there were 99 leafy vegetables [including pakchoi (*Brassica rapa* L. *chinensis* Group.) ($n = 39$), water spinach (*Ipomoea aquatica* Forssk.) ($n = 16$), celery (*Apium graveolens* L.) ($n = 7$), choy sum (*Brassica rapa* subsp. *parachinensis*) ($n = 7$), leek (*Allium tuberosum* Rottl.) ($n = 5$), lettuce (*Lactuca sativa* L.) ($n = 5$), spinach (*Spinacia oleracea* L.) ($n = 5$), amaranth (*Amaranthus mangostanus* L.) ($n = 3$), cole (*Brassica napus* L.) ($n = 3$), mustard (*Brassica juncea* L.) ($n = 3$), watercress (*Nasturtium officinale* L.) ($n = 2$), kale (*Brassica oleracea* L.) ($n = 2$), and Swiss chard (*Beta vulgaris* L.) ($n = 2$)], 21 rootstalk vegetables [including radish (*Raphanus sativus* L.) ($n = 7$), ipomoea (*Ipomoea batatas* L.) ($n = 7$), carrot (*Daucus carota* L.) ($n = 6$), and taro (*Colocasia esculenta* L.) ($n = 1$)], and 22 legumes [including cowpea (*Vigna unguiculata* L.) ($n = 20$), and kidney bean (*Phaseolus vulgaris* Linn.) ($n = 2$)]. They were the most common vegetables grown and consumed by local residents in the study area. The fresh edible parts of vegetables were collected, stored in clean plastic bags, and transported to the laboratory for sample treatment as soon as possible. The edible portions of the vegetables were thoroughly washed with tap water, and subsequently rinsed with ultrapure deionized water ($18 \text{ M}\Omega \cdot \text{cm}$). Vegetable samples were blotted dry with filter paper and fresh biomass was recorded before being chopped into small pieces and oven-dried at 60°C for 48 h. The vegetable samples were powdered with a stainless steel grinder and passed through a 0.25 mm sieve before chemical analysis. Rice grain samples were oven-dried at 60°C for 48 h before being dehusked. Soil samples were air-dried and sieved $<2 \text{ mm}$ for pH measurement. Portions of the soil samples were ground in a porcelain mortar $<0.25 \text{ mm}$ for the measurement of soil organic matter (SOM), and $<0.15 \text{ mm}$ for total Cd analysis (Lu, 2010).

2.3. Chemical analysis

Soil samples were digested with aqua regia in a heating block (McGrath and Cunliffe, 1985). Grain samples were digested with 5 mL of high-purity concentrated HNO_3 in a microwave digestion system (CEM, Mars, USA) (Ma et al., 2014). Vegetable samples were digested with $\text{HNO}_3/\text{HClO}_4$ (87/13, v/v) in a heating block (Zhao et al., 1994). The extractable Cd in soil was extracted with 0.1 M CaCl_2 in a ratio of 1:5 (w/v) on a rotatory shaker for 4 h (180 r min^{-1} , $25 \pm 1^{\circ}\text{C}$) (Simmons et al., 2008). Suspensions were centrifuged ($4000 \times g$, 15 min) and the supernatant was filtered with a $0.2 \mu\text{m}$ membrane (mixed cellulose ester) immediately. Soil pH was measured in distilled water at a soil-solution ratio of 1:2.5 (w/v) using a glass pH electrode. Content of SOM was determined using the $\text{K}_2\text{CrO}_4\text{-H}_2\text{SO}_4$ method (Lu, 2010).

Cadmium in the aqua-regia digests (for soil pseudo-total Cd), in the HNO_3 digests (for rice Cd) and in the $\text{HNO}_3/\text{HClO}_4$ digests (for vegetable Cd) or CaCl_2 extracts (for available fraction of soil Cd) was determined by inductively coupled plasma mass spectrometry (ICP-MS, Perkin Elmer NexION 300X, USA). Indium supplied from Perkin Elmer (USA) was added to the samples as the internal standard. Quality control for Cd analysis was performed by including reagent blanks, duplicates

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