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Human health risk assessment of heavy metals in soil–vegetable system: A multi-medium analysis

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HIGHLIGHTS

- Flourishing private economy caused increasing heavy metal damages.
- Leafy and rootstock vegetables posed higher hazards.
- Cr has the biggest non-carcinogenesis effect while Cd generates the greatest cancer risk.
- Negative impacts on humans and the environment may cause additional costs not included in sales expenditures.

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ABSTRACT

Vegetable fields near villages in China are suffering increasing heavy metal damages from various pollution sources including agriculture, traffic, mining and Chinese typical local private family-sized industry. 268 vegetable samples which included rape, celery, cabbages, carrots, asparagus lettuces, cowpeas, tomatoes and cayenne pepper and their corresponding soils in three economically developed areas of Zhejiang Province, China were collected, and the concentrations of five heavy metals (Pb, Cd, Cr, Hg and As) in all the samples were determined. The health risk assessment methods developed by the United States Environmental Protection Agency (US EPA) were employed to explore the potential health hazards of heavy metals in soils growing vegetables. Results showed that heavy metal contaminations in investigated vegetables and corresponding soils were significant. Pollution levels varied with metals and vegetable types. The highest mean soil concentrations of heavy metals were 70.36 mg kg^{-1} Pb, 47.49 mg kg^{-1} Cr, 13.51 mg kg^{-1} As, 0.73 mg kg^{-1} for Cd and 0.67 mg kg^{-1} Hg, respectively, while the metal concentrations in vegetables and corresponding soils were poorly correlated. The health risk assessment results indicated that diet dominated the exposure pathways, so heavy metals in soil samples might cause potential harm through food-chain transfer. The total non-cancer and cancer risk results indicated that the investigated arable fields near industrial and waste mining sites were unsuitable for growing leaf and root vegetables in view of the risk of elevated intakes of heavy metals adversely affecting food safety for local residents. Chromium and Pb were the primary heavy metals posing non-cancer risks while Cd caused the greatest cancer risk. It was concluded that more effective controls should be focused on Cd and Cr to reduce pollution in this study area.

1. Introduction

There has long been concern about the issue of pollution by heavy metal because of their toxicity for plant, animal and human beings and their lack of biodegradability (Li et al., 2006; Jang et al., 2006; Zhuang et al., 2009). Soil is the primary reservoir of heavy metals in the atmosphere, hydrosphere and biota, and thus plays a fundamental role in the overall metal cycle in nature (Cao et al., 2010). Heavy metals in soil pose potential threats to the environment and can damage human health through various absorption pathways such as direct ingestion, dermal contact, diet through the soil–food chain, inhalation, and oral intake (Lu et al., 2011; Komárek et al., 2008; Park et al., 2004; Al-Saleh et al., 2004).

Vegetables play important roles in our daily diet as economic crops. However, various human activities such as mining, industrial processing like smelting, pesticides, automobile exhausts and fertilization, especially the huge annual applications of organic livestock manure, which is the traditional agricultural fertilizer, are causing elevated heavy metal concentrations in the environment in China (Zhuang et al., 2009; Cao et al., 2010; Zheng et al., 2007, 2010; Shi et al., 2011). Vegetables take up heavy metals by absorbing them from contaminated soils, as well as from deposits on parts of the vegetables exposed to the air from polluted environments (Wang et al., 2005). Chronic intakes of heavy metals have damaging effects on human beings and other animals (Zheng et al., 2007;

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Lai et al., 2010; John and Andrew, 2011). For example, Cr, Cu and Zn can cause non-carcinogenic hazardous such as neurologic involvement, headache and liver disease, when they exceed their safe threshold values (US EPA, 2000). There is also evidence that chronic exposure to low doses of cancer-causing heavy metals may cause many types of cancer. For example, Park et al. (2004) found increased lifetime risk of lung cancer death resulted from occupational exposure to dusts and mists containing hexavalent chromium. Dietary cadmium intake due to the consumption of environmentally contaminated rice and other foods was associated with an increased risk of postmenopausal breast cancer (Hiroaki et al., 2013). Acute and chronic arsenic exposure could also cause numerous human health problems. These included dermal, respiratory, cardiovascular, gastrointestinal, hematological, hepatic, renal, neurological, developmental, reproductive, immunological, genotoxic, mutagenetic, and carcinogenic effects (such as liver cancer) (Kapaj et al., 2006; Lin et al., 2013). Despite the economic benefits of industry, improved income and high crop yields due to fertilization, negative impacts on humans and the environment may cause additional costs not included in sales expenditures (Peter et al., 2012). Especially for the ubiquitous and non-biodegradable heavy metals, the negative effects persist for several decades and even longer.

The Household Responsibility System (HRS) was initiated during the late 1970s in China. It has brought a profound change to the rural economy. Farmland was allocated to each farmer household on the basis of family size. The farmers were then given the authority to manage their contracted land, including all decisions regarding production (Liu et al., 2009). In particular, vegetable fields were located very near to the villages and conveniently close to the farmers. Unfortunately, this means that the growing vegetables and soils are at high risk of contamination by local industrial pollution, since many small family-sized factories such as metal smelting and battery making businesses are located in villages due to the booming private economy.

Zhejiang province, one of the most important economic development provinces in China, has been leading the national private economy and the flourishing private enterprises bring about severe and numerous negative environmental effects. As is typical of regions in Southeast China, Zhejiang Province is densely covered with drainage ditches that form a network waterway and consequently the arable fields nearby are readily polluted by domestic and industry wastes. Many investigations have been conducted in which heavy metal pollution was evaluated using traditional methods (Granero and Domingo, 2002; Jarup, 2003; Chary et al., 2008).

In our research, particular emphasis is placed on the use of descriptive statistics in determining the effects of heavy metals pollution. This is the first study that has assessed the potential health risks of heavy metal exposure to multiple medium in such critical vegetated areas. From this data we can use various options to reduce human health hazards.

Primary objectives were: (a) to explore the current extent of local heavy metal pollution in vegetated soils and plants, (b) to determine the potential health risks of heavy metals as cumulative carcinogenic and non-carcinogenic risks via the multiple routes of ingestion, inhalation, dermal exposure and diet from the soil–vegetable system, and (c) to provide a reference for policy decision making on the prevention and treatment of heavy metal pollution.

2. Materials and methods

2.1. Study area

Zhejiang province is located in the Yangtze River delta region of Southeast China, covering a total area of 104,141 km^2 and having a total population of 5442.69 million inhabitants. With a high population density and developed industries and agriculture, Zhejiang province has 3000–4000 years of a history of food production. In this study, we selected three counties of Hangzhou, Changxing, and Shangyu in Zhejiang province as the study area. The first area is a typical suburban belt located in the northeast of Hangzhou county, the famous provincial capital, which was planted with 24.1 km^2 vegetables (30°16′16″–30°20′6″ N, 120°11′25″–120°14′58″ E). In the suburban areas in China, the heavy metals in soil are commonly affected by multiple factors including traffic, agriculture, and industry. As one of the "Top 100 counties" in China, Changxing is famous for its battery industry which could cause potential heavy metal pollution in the local environment. We sampled from the vegetated zone covering a latitude of 30°58′27″–31°02′3″ N and a longitude of 119°50′56″–119°57′16″ E. In Shangyu, another developed "Top 100 counties" in China, we sampled in a vegetated area covering from 29°59′42″–30°04′25″ N to 120°45′25″–120°49′38″ E. This sampling area is near a lead and zinc mine tailing which has been abandoned for almost a century. Due to long-term exposure to weathering, the pollutants are distributed around the mine within a heavy metal polluted area of about 800 hectares.

2.2. Sample collection

There were 268 vegetable samples (1 kg edible part of each) including 127 leafy vegetables (57 rapes, 43 celeries, 27 cabbages), 68 rootstock vegetables (26 carrots and 42 asparagus lettuces (*Lactuca sativa*)), 25 legume vegetables (25 cowpeas), and 48 solanaceous vegetables (26 tomatoes and 22 cayenne peppers) collected in 2011 from the study area. Simultaneously, 268 soil samples were collected at the vegetable sampling sites. When sampling, the study area was split into many sampling units, selected by planting mode and pollution background. Within the same sampling unit, five soil samples were collected using an "S" sampling procedure and then bulked to provide an individual composite sample. The vegetable samples were collected along the same gradients. Soil samples were taken in the immediate vicinity of the roots of the crop samples from 0 to 15 cm depth. Only the edible part of each vegetable was collected for analyses. All the soil or vegetable samples were quartered separately to provide sub-samples.

2.3. Pre-treatment and analysis of soil and vegetable samples

Soil samples were air-dried in the laboratory and sieved <2 mm. A part of the soils were ground in a porcelain mortar <100 mesh. They were stored in polyethylene bags at 4 °C prior to analysis. The edible portions of the vegetables were rinsed in distilled water and subsequently rinsed again with high-purity deionized water. After being milled by a ceramic-coated grinder, the vegetable samples were frozen at 18 °C until chemical analysis.

Soil pH (H_2O) and electrical conductivity (EC) were determined in distilled water (1:2.5 w/v). Soil mechanical composition (sand, silt, clay) was determined by a hydrometer method (Agricultural Chemistry Committee of China, 1983). Organic carbon (OC) contents were measured using the Walkey–Black wet oxidation method (Agricultural Chemistry Committee of China, 1983). Total Pb, Cd, Cr in the soils and vegetables were digested by $HF–HNO_3–HClO_4$ and analyzed by an inductively coupled plasma-mass spectrometer (ICP-MS, Agilent 7500a, USA). Total Hg and As were digested by $HNO_3–HCl$ in a water bath and determined by a double channel Atomic Fluorescence Spectrometer with a hollow cathode lamp of Hg and As and high purity argon gas as a carrier (AFS-9100). The amounts of soil and vegetable samples for analysis were 0.5 g and 5 g, respectively. The limits of detection (LOD) for Pb, Cd, Cr, Hg and As were 11.9, 2.3, and 113.7, 2.0 and 20.0 ng L^{-1} respectively, the limits of quantity (LOQ) for these five metals were 35.7, 6.9, 341.1, 6.0 and 60.0 ng L^{-1} respectively.

2.4. Risk assessment methods

The human health risk models including carcinogenic and non-carcinogenic ones raised by US EPA, have proved successful and adopted worldwide. Currently, there is no agreed limit for acceptable