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Research article

Heavy metal pollution and health risk assessment of agricultural soils in a typical peri-urban area in southeast China

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ABSTRACT

Heavy metal pollution in peri-urban areas in China is serious and complex. We thus developed an integrated evaluation method to assess heavy metal pollution and potential health risk to residents in a typical peri-urban area with diverse anthropogenic emission sources and cropping systems. Ecological risk was evaluated using Nemerow's synthetical pollution index (P_n) and Potential ecological risk index (RI). Then polluted areas and responsible emission sources were identified by GIS mapping. Health risk caused by food intake and soil exposure was calculated by accounting for the influence of anthropogenic emissions and cropping systems. Agricultural soils in the study area were polluted by cadmium (Cd), mercury (Hg), lead (Pb), and arsenic (As). High concentrations mainly occurred near the mining area and along the roadsides. The accumulation of heavy metals in crops followed the order of tea leaves > rice grain > vegetables. The hazard index of potential human health risk caused by chronic soil exposure and food intake was 15.3, indicating obvious adverse health effects. 87.5% of health risk was attributed to food consumption, and significantly varied among different cropping systems with the decreasing order of rice (10.44) >vegetable (2.86) > tea (0.05). The integrated method of ecological and health risk index, which takes consideration of both anthropogenic emission and cropping system can provide a practical tool for evaluating of agricultural soil in the peri-urban area regrading different risk factors.

1. Introduction

Heavy metal pollution has become a worldwide environmental problem (Facchinelli et al., 2001; García-Carmona et al., 2017; Solgi et al., 2012). Soil pollution by heavy metals has been accelerated in China in the last two decades due to a rapid economic development and industrialization (Chen et al., 1999). Heavy metals in agricultural soils can exert health risk to human attributed to consumption of crops and chronic exposure to soil particles (Boim et al., 2016; Liu et al., 2017; Oliver, 1997). Chronic exposure to Cd was reported to cause pulmonary adenocarcinomas, lung cancer, kidney dysfunction, and bone fractures (Żukowska and Biziuk, 2008). Long-term high dose exposure to zinc (Zn) affects cholesterol balance and fertility (Zhang et al., 2012). Lead (Pb) has adverse effects on blood enzymes and central nervous system (Kaufmann et al., 2003). Chronic arsenic (As) exposure may result in hyperkeratosis, skin lesions, and cancer of lung, bladder, and kidney (Hughes, 2002). Mercury (Hg) can accumulate in fatty tissues after long-term exposure and can damage the human central nervous system (Lee et al., 2006; Clarkson, 1987). Copper (Cu), chromium (Cr) and nickel (Ni) also have adverse effects on human health when exposures exceed the tolerable dose levels (USEPA, 2000).

Heavy metals in soil originate from both natural and anthropogenic sources. Natural sources are mainly attributed to lithogenesis, weathering, erosion, and other geological processes (Stafilov et al., 2010). Anthropogenic sources include mining, industrial activities, traffic emissions, and agricultural activities (mainly the use of pesticides and chemical fertilizers) (Anju and Banerjee, 2012; Gu et al., 2014; Han et al., 2006). Mining, industrial activities and traffic emission were considered the major anthropogenic sources for heavy metal pollution of soils. In the

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process of mining, heavy metals from geological origin can be released into the environment. Rodríguez et al. (2009) reported that contents of Pb, Zn, Cd near mining areas were significantly higher than those in the unaffected area. Heavy metals and their compounds are frequently used as catalysts and chemical additives in industrial processes. Therefore, soils could be polluted by industrial emissions (Gowd et al., 2010; Krishna and Govil, 2008). Studies reported that traffic emission is a major contributor to Pb and Cd pollution, because of the exhausts from gasoline, brakes, and tyres (Kim et al., 2004; Kumar et al., 2001). Contents of heavy metals in soil and crops were found to be higher at nearby roadsides (Hamurcu et al., 2010). A national soil monitoring by the Chinese government also clearly showed that agricultural soils in China have been widely polluted by industrial activities, mining, and farming (China State Council, 2014). Peri-urban areas are regions located at the intersections of urban and rural regions and are characterized by highly varied agricultural and industrial activities. It plays an important role in providing food to local residents and protecting ecosystems. Agricultural soil pollution by heavy metals is more severe and complex in peri-urban areas as such areas are typically exposed to multiple emission sources. Therefore, it is important to evaluate soil heavy metal pollution in such an area.

Besides the variation in anthropogenic emissions, there are various cropping systems in peri-urban area, and nearly all crops are consumed locally. Different cropping systems can have different impacts on heavy metal uptake from soil to plant, and therefore can lead to different health risks to residents through food chain (Antoniadis et al., 2017; Liu et al., 2013; Zhuang et al., 2009). Typical crops in the peri-urban areas of southeast China are rice, vegetable and tea. Rice is the most important staple food for Chinese, especially for citizens in south China. High concentration of Pb and Cd in rice were found all over China, exceeding the maximum allowable concentrations (MAC) (Huang et al., 2009; Zhao et al., 2010). Praveena and Omar (2017) found obvious health risk presenting for both adult and children of Malaysian population through cooked rice grain ingestion. Vegetables are sources of essential human nutrients, and peri-urban areas play a central role in vegetable supply to the local residents. Huang et al. (2014) analyzed heavy metal contents in vegetables from Zhejiang, and found that health risks of Hg and Cd were higher than threshold levels, whereas As and Pb posed only minimal risks to local residents. Pan et al. (2016) claimed that there were only low health risks to human related to vegetable intake. Except for rice and vegetables, tea is a contributor to human health risk, specific for southern China. Tea is a popular beverage to Chinese, and high concentrations of heavy metals in tea may pose a health threat to tea drinkers (Cao et al., 2010; Qin and Chen, 2007). Obviously, in this kind of peri-urban areas with various cropping systems, ecological evaluation methods like Nemerow's synthetical pollution index (P_n) and Potential ecological risk index (RI), which are only based on heavy metal contents in soils may not be reliable for the comprehensive assessment of heavy metal pollution. Health risk to residents through food intake of different cropping systems should be quantified and compared, so that effective management can be developed to maintain food safety and to protect human health.

This study was attempted to develop an integrated pollution evaluation method for assessing ecological and health risk of heavy metal pollution under different anthropogenic emissions and cropping systems in peri-urban areas. Such a method can result in effective environmental management to protect ecosystem and human health. The ecological risk of heavy metal pollution in soil was assessed by Nemerow's synthetical pollution index (P_n) and Potential ecological risk index (RI). The spatial distribution of heavy metal and risk index could be help for pollution assessment (Papadopoulou-Vrynioti et al., 2013, 2014). Health risk to human was quantified using a risk exposure model, which is recommended by United States Environmental Protection Agency (USEPA). The main aims of this study were: 1) to assess heavy metals pollution in soil and crops in a typical peri-urban area in southeast China; 2) to explore the relative impacts of different anthropogenic sources and cropping systems; 3) to analyze the potential health risks to local residents through chronic soil exposure and food consumption.

2. Materials and methods

2.1. Sample collection

The selected study area is located in southeast China, with a total area of 12 × 10 km^2 and administered by four towns. It is about 15 km away from the city center, and is considered as a production area for crops and vegetables. There were more than 700 km^2 of arable land, and more than 80% of the residents were peasants. The major crops planted in this area were rice, vegetables and tea. The potential heavy metal emission sources included traffic emission, a Pb-Zn mine, and industrial plants (like dyeing mills, textile mills and metal-processing plants). In addition, a large industrial hub was located in the western part of the study area, containing several plastic factories and a printing company (Fig. 1). Soil samples and crop samples were collected from December 2014 to January 2015. One hundred and fourteen topsoil samples (0–15 cm) were collected with a stainless steel auger. The sampling sites were randomly selected with a sampling grid of about 1 × 1 km^2. Each sample (about 1.5 kg fresh soil) consisted of five top-soil cores collected at the central and four additional points towards North, East, South and West. The crop samples (rice, vegetable, tea) were randomly collected in the sampled fields. Edible part of rice and vegetables and top tea leaves were taken and packed into polyethylene bags. During the collection, five random samples from each plot were combined to make one composited sample (3.0 kg fresh weight for rice and vegetable, and 1.0 kg fresh weight for tea).

2.2. Sample analysis

After transported to the laboratory, the soil samples were air-dried, ground and passed through a 100-mesh polyethylene sieve. Rice samples were air-dried and ground to <1.0 mm. Edible part of vegetables and tea leaves were washed and dried in an oven at 105 °C for half an hour and then at 60 °C for 72 h until constant weight was obtained. The dried vegetables and tea leaves were ground to <1.0 mm. All ground soil and crop samples were stored in a desiccator prior to analysis. Soil samples were digested in a closed poly-tetrafluoroethylene system with a mixture of HNO_3 (5 mL) – HF (1 mL) – $HClO_4$ (1 mL) at 180 °C for 10 h, cooled to room temperature, and diluted with deionized water to 30 mL. Concentrations of Pb, Cr, Cu, Zn, and Ni were determined using inductively coupled plasma optical emission spectrometry (ICP-OES, Thermo122 6300, USA), and Cd was determined using inductively coupled plasma-mass spectrometry (ICP-MS, Agilent 7500a, USA). Mercury (Hg) and arsenic (As) were analyzed using atomic fluorescence spectrometry (AFS, AFS-230E Haiguang Analytical Instrument Co., Beijing, China) after being heated on water bath at 100 °C for 4 h with 5 mL aqua regia and 5 mL deionized water. Crop samples were digested in a closed poly-tetrafluoroethylene system with a mixture of HNO_3 (5 mL) – H_2O_2 (1 mL) at 120 °C for 10 h, and then analyzed together with soil samples. Sample replicates, reagent blanks, and standard reference materials (GBW07429, the National Research Center for Certified Reference Materials of China) were included in each batch of analyses to ensure analytical quality. The limit of determinations (LOD) of As, Hg, Cd, Pb, Cu, Ni, Zn, Cr