Science of the Total Environment 472 (2014) 407–420

Contents lists available at ScienceDirect

Science of the Total Environment

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

The identification of ‘hotspots’ of heavy metal pollution in soil–rice systems at a regional scale in eastern China

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HIGHLIGHTS

- GIS and Moran's I clearly explored the spatial variability and pollution hotspots.
- Family-sized industry presents potential heavy metal damages.
- Both soil metals and physico-chemical properties affected metal uptake by rice.

ARTICLE INFO

Article history:
Received 31 July 2013
Received in revised form 7 November 2013
Accepted 9 November 2013
Available online 1 December 2013

Keywords:
Heavy metals
Pollution hotspots
Spatial variability
Geostatistics
Moran's I

ABSTRACT

Chinese agricultural soils and crops are suffering from increasing damage from heavy metals, which are introduced from various pollution sources including agriculture, traffic, mining and especially the flourishing private metal recycling industry. In this study, 219 pairs of rice grain and corresponding soil samples were collected from Wenling in Zhejiang Province to identify the spatial relationship and pollution hotspots of Cd, Cu, Ni and Zn in the soil–rice system. The mean soil concentrations of heavy metals were 0.316 mg kg^{-1} for Cd, 47.3 mg kg^{-1} for Cu, 31.7 mg kg^{-1} for Ni and 131 mg kg^{-1} for Zn, and the metal concentrations in rice grain were 0.132 mg kg^{-1} for Cd, 2.46 mg kg^{-1} for Cu, 0.223 mg kg^{-1} for Ni and 17.4 mg kg^{-1} for Zn. The coefficient of variability (CV) of soil Cd, Cu and rice Cd were 147%, 146% and 180%, respectively, indicating an extensive variability. While the CVs of other metals ranged from 23.4% to 84.3% with a moderate variability. Kriging interpolation procedure and the Local Moran's I index detected the locations of pollution hotspots of these four metals. Cd and Cu had a very similar spatial pattern, with contamination hotspots located simultaneously in the northwestern part of the study area, and there were obvious hotspots for soil Zn in the north area, while in the northeast for soil Ni. The existence of hotspots may be due to industrialization and other anthropogenic activities. An Enrichment Index (EI) was employed to measure the uptake of heavy metals by rice. The results indicated that the accumulation and availability of heavy metals in the soil–rice system may be influenced by both soil heavy metal concentrations and soil physico-chemical properties. Cross-correlograms quantitatively illustrated that EIs were significantly correlated with soil properties. Soil pH and organic matter were the most important factors controlling the uptake of heavy metals by rice. As results, positive measures should be taken into account to control soil pollution and to curtail metal contamination to the food chain in the areas of Wenling, which were the most polluted by toxic metals.

1. Introduction

The potential impact of climate change, heavy metals, pesticide residues and numerous adulterants on food security is one of the most important public health issues worldwide (Akhtar, 2013; Miraglia et al., 2009). Soil pollution is the most critical factor that leads to the problem of food safety (Akhtar, 2013; Micó et al., 2006). When considering the different kinds of contaminants, heavy metals are especially dangerous because of their toxicity, non-biodegradability and persistence (Yan et al., 2013). They can cause problems in agriculture, the aerosphere, and water, and may be finally accumulated in the human body, causing serious health problems (Fu and Wei, 2013; Wei and Yang, 2010). With the development of industrialization and consequential food security accidents, soil heavy metal pollution has also became of great concern to the public (He et al., 2013; Liu et al., 2006; Rogan et al., 2009; Römkens et al., 2009), especially in eastern China with its rapid economic growth.

Rice production is the dominant agricultural activity in China, with rice forming the staple diet of more than 65% of the Chinese population (Zhang et al., 2005). China ranks as the second largest rice producer in the world. The mean consumption of rice grain in China is 206 kg per

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http://dx.doi.org/10.1016/j.scitotenv.2013.11.046

year per person, and as much as 50% of food intake may be assumed to be grown on contaminated soil (Dudka et al., 1996). Long term consumption of local rice would pose a potentially high health risk caused by heavy metal exposure to consumers (He et al., 2013). Therefore, both the qualities of soil and rice produced are extremely closely related to local public health. Rice cultivated on soils polluted with Cd, Pb, Cr, Hg, As, Cu, Ni and Zn has been reported to accumulate the metals in root, leaf, stem and grain causing food-chain and ecotoxicological problems (Wu and Chen, 2013; Dudka and Miller, 1999). For example, chronic exposure to Ni may cause many diseases such as cardiovascular and kidney diseases, as well as allergic dermatitis (Khlifi and Hamza-Chaffai, 2010). Among all toxic heavy metals, Cd ranks the highest in terms of damage to plant growth and human health (Dong et al., 2006). This heavy metal is not an essential element for plants, but its uptake and accumulation in plants pose a serious health issue to humans through the food chain, such as the "itai-itai" disease that caused more than 100 deaths in Japan from 1922 to 1965 (Sun et al., 2013). Therefore, Cd is receiving the most attention as a pollutant among all the metals (Römkens et al., 2009; Sriprachote et al., 2012).

Geostatistics and spatial analysis, together with Geographic Information System (GIS) techniques, have been widely used to quantify the spatial distribution of soil properties, reduce uncertainties, minimize investigation costs, and to identity pollution sources (Liu et al., 2006; Zhang and McGrath, 2004; Zhang et al., 2009). Compared to the few methods proposed for hotspot identification, such as Getis's G index (Getis and Ord, 1992), spatial scan statistics (Ishioka et al., 2007) and Tango' C index (Tango, 1995; Zhang and Lin, 2006), the Local Moran's I index is a very popular tool because of its ability to explicitly identify spatial outliers and its accurate (Anselin, 1995; Zhang et al., 2008; Sugumaran et al., 2009). The Local Moran's I index examines the individual locations, enabling hotspots to be identified based on a comparison with the neighboring samples. It can be used to estimate the statistical significance of spatial correlations (Fu et al., 2011; Overmars et al., 2003; Zhang and McGrath, 2004).

The combination of geostatistical methods with Moran's I analysis would provide more accurate results and evidence to characterize the heavy metal problems. In this study, geostatistics, GIS and Local Moran's I index were applied to explore the spatial variation of Cd, Cu, Ni and Zn. The primary objectives were (1) to investigate the concentration of heavy metals in soil and rice grain, (2) to determine the spatial correlations between heavy metal pollution hotspots in soil–rice systems, and (3) to identify the main soil properties influencing heavy metal concentrations in rice grain.

2. Materials and methods

2.1. Study area

This research was performed in Wenling (121°10′ ~ 121°44′E, 28°13′ ~ 28°32′N), which is located in the southeast of Zhejiang Province, China (Fig. 1). It stretches from the borders with Yuhuan, Leqing and the Leqing gulf. Wenling has an area of approximately 926 km^2, covering 11 towns and 5 urban sub-districts with a population of 1.2 million.

Wenling has a subtropical climate with an average annual air temperature of 17.3 °C and annual precipitation of 1693 mm. The main soil types in the study area are Argosols, Ferrosols, Cambosols, Anthrosols and Halosols accounting for 0.06%, 48.29%, 2.35%, 43.85% and 5.45%, respectively. Rice is the dominant agricultural crop. There are over 10,000 private industrial enterprises including a flourishing electronic recycling industry and 1725 large state-owned enterprises in Wenling County.

2.2. Soil sampling and chemical analysis

In October of 2011, 219 pairs of rice grain samples and their corresponding soil samples were collected in Wenling. When sampling, five topsoil samples were collected within a 10 m radius circular area using an "S" sampling procedure (Liu et al., 2013) and then bulked to provide an individual composite sample. The rice grain was collected similarly. All the soil and rice samples were quartered separately to provide sub-samples for further analysis (e.g., soil physical and chemical analysis), with the redundant abandoned. The distribution of sampling points is shown in Fig. 1.

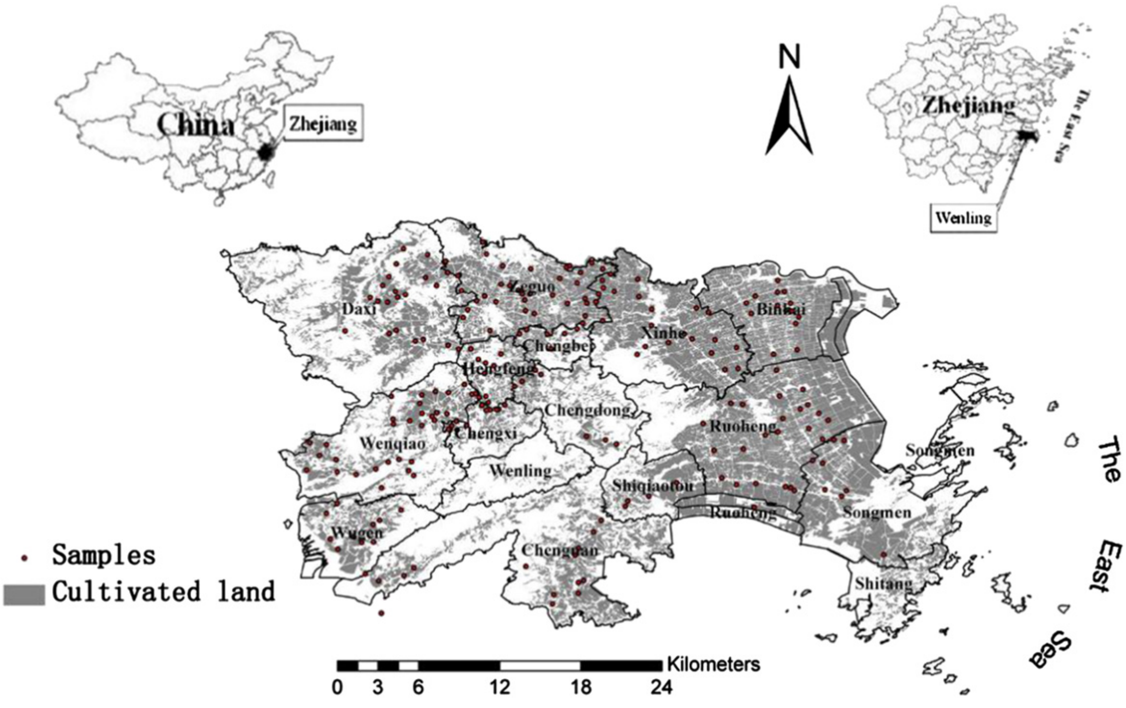

Fig. 1. Location of the study area and distribution of sampling points.