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Source apportionment and health risk assessment of heavy metals in soil for a township in Jiangsu Province, China

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HIGHLIGHTS

- Soil heavy metals caused unacceptable health risks, mainly through homegrown food.
- Arsenic and chromium were the predominant hazardous elements.
- Waste incineration, textile/dyeing industries were the main anthropogenic inputs.
- Electroplating and livestock/poultry industries produced the highest health risks.

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ABSTRACT

Human activities contribute greatly to heavy metal pollution in soils. Concentrations of 15 metal elements were detected in 105 soil samples collected from a typical rural-industrial town in southern Jiangsu, China. Among them, 7 heavy metals—lead, copper, zinc, arsenic, chromium, cadmium, and nickel—were considered in the health risk assessment for residents via soil inhalation, dermal contact, and/or direct/indirect ingestion. Their potential sources were quantitatively apportioned by positive matrix factorization using the data set of all metal elements, in combination with geostatistical analysis, land use investigation, and industrial composition analysis. Furthermore, the health risks imposed by sources of heavy metal in soil were estimated for the first time. The results indicated that Cr, Cu, Cd, Pb, Ni, and Co accumulated in the soil, attaining a mild pollution level. The total hazard index values were 3.62 and 6.11, and the total cancer risks were 9.78×10^{-4} and 4.03×10^{-4} for adults and children, respectively. That is, both non-carcinogenic and carcinogenic risks posed by soil metals were above acceptable levels. Cr and As require special attention because the health risks of Cr and As individually exceeded the acceptable levels. The ingestion of homegrown produce was predominantly responsible for the high risks. The potential sources were apportioned as: a) waste incineration and textile/dyeing industries (28.3%), b) natural sources (45.4%), c) traffic emissions (5.3%), and d) electroplating industries and livestock/poultry breeding (21.0%). Health risks of four sources accounted for 23.5%, 32.7%, 7.4%, and 36.4% of the total risk, respectively.

1. Introduction

Along with the remarkable achievement of rapid economic development, environmental costs are also significantly increasing. Soil contamination by heavy metals (HM) has been increasing worldwide (Facchinelli et al., 2001; Frangi and Richard, 1997; Giller and McGrath, 1988; Huang et al., 2007; Li et al., 2014; Solgi et al., 2012) and has become the focus of attention in recent years (Chen et al., 2016; Wang et al., 2016; Zou et al., 2015). Heavy metals have generally high toxicity with low concentration thresholds, long residence times (often exceeding decades), and persistent bioavailability (Alloway, 2013). They could be hazardous to human health and ecosystems at a trace level due to their ubiquity, toxicity, and persistence (Burges et al., 2015; Guney et al., 2010). The United

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States Environmental Protection Agency (USEPA) considers heavy metals such as Cd, Cr, As, Hg, Pb, Cu, Zn, and Ni as priority control pollutants (USEPA, 2014). Many studies have focused on pollution levels and risk assessments of HM in soil environments (Chabukdhara and Nema, 2013; Li et al., 2014; Zhao et al., 2012; Zheng et al., 2010). According to the first National Survey of Soil Contamination conducted by the Ministry of Environmental Protection and Ministry of Land and Resources of China, concentrations of heavy metals in 82.8% of the soil samples exceeded the standard limit (MEPPRC and MLRPRC, 2014). It is necessary to evaluate human health risks from exposure to soil heavy metals, and identify the contamination sources to improve the soil environment and protect human health.

Compared with research involving pollution investigation, risk assessment, or remediation of soil heavy metals, few have been conducted for qualitative source identification, and even fewer for quantitative source apportionment. Qualitative methods used in previous source studies have included geostatistical models based on geographic information systems (Davis et al., 2009; Facchinelli et al., 2001; Nanos and Rodríguez Martín, 2012; Sun et al., 2013; Zhang, 2006), multivariate statistics analyses (principal component analysis, PCA; cluster analysis, CA) (Qu et al., 2013), and isotopic signatures (Cheng and Hu, 2010; Luo et al., 2011). Quantitative methods have included mostly receptor models, such as a Chemical Mass Balance (CMB) model, PCA related methods (Absolute Principal Component Scores, APCS; Multiple Linear Regression, PCA-MLR; (A)PCS-MLR; UNMIX model), and Positive Matrix Factorization (PMF) (Luo et al., 2014; Mijić et al., 2010; Wang et al., 2016). PMF has been widely applied for source apportionment of pollutants in the atmosphere (Amil et al., 2016; Hsu et al., 2016; Kim et al., 2007; Lee et al., 1999), water (Li et al., 2015; Li and Zhang, 2011; Rodenburg et al., 2011; Soonthornnonda and Christensen, 2008), and sediment (Bzdusek et al., 2006; Chen et al., 2013; Comero et al., 2014; Sundqvist et al., 2010). There are also some successful cases of source apportionment used for soil heavy metals (Schaefer and Einax, 2016; Vaccaro et al., 2007; Xue et al., 2014). Apportioning sources of soil heavy metals could help in understanding the characteristics and contributions of different sources, so that appropriate control measures can be effectively targeted to reduce anthropogenic metal inputs to soil. However, the composition of metals in emissions differs depending on the source, and different metals have different toxicities. Thus, the sources should be prioritized for control and management based on the potential health risk of each source, rather than the contribution of each source to the environmental levels of heavy metals, which we proposed here for the first time.

Jiangsu Province has a highly developed economy, being located in the eastern coastal region of China. The study area, atypical rural-industrial town in southern Jiangsu, has a complex industry structure, with agricultural activities of planting, feeding, and aquaculture, and various industrial activities, such as production of photovoltaic electronics, bio-pharmaceutical products, mechanical metallurgy, electronic components, textile clothing, as well as printing and dyeing industries. The town also has the advantage of good transportation, with two first class roads passing through the core of the territory, connecting the commercial cities of Suzhou and Wuxi. Diverse industries and heavy traffic, which provide various heavy metal emission sources, made challenging to quantify the appropriate sources. Fortunately, the contamination history and related information of source changes were captured during our 10 years of researching this area (Cao et al., 2010; Jiang et al., 2015). Combining this knowledge with auxiliary methods, such as spatial analysis, correlation analysis, and others, apportioned sources could be correctly interpreted. Previous investigations have found that inhabitants were confronted with potential health risks from exposure to heavy metals in soils and self-planted rice and garden vegetables (Cao et al., 2010; Jiang et al., 2015). However, these samplings were conducted in 2008 and 2009. Several policies, such as rectification of polluted industries, transformation of the energy structure, and the termination or emigration of heavily polluting enterprises have been implemented since September 2010. In order to measure the reform effect, heavy metal concentrations in soil samples needed to be analyzed, and the relative health risks estimated.

The three main objectives of this study were: (1) to investigate the concentration distribution of major and trace metals in soil in the study area; (2) to assess the health risk of residents exposed to seven heavy metals—lead, copper, zinc, arsenic, chromium, cadmium, and nickel—in soils; and (3) to apportion potential sources of these common heavy metals and quantify their contributions using a PMF model combined with geostatistical analysis, land use investigation, and historical industrial information, and further to evaluate the health risks imposed by each source.

2. Materials and methods

2.1. Study area

The studied town, X, with an area of 104.26 km^2, belongs to Changshu City, Jiangsu Province, China. The region has a north subtropical humid monsoon climate with an average annual temperature of 16.6 °C and mean annual precipitation of about 1320 mm. The predominant and secondary prevailing wind directions are east-southeast and east-northeast, respectively. The annual average wind speed is 3.7 m/s. The main soil type is gleyic clayey paddy soil derived from lacustrine deposits. The study area belongs to the well-developed regions in the east of China. The area has been undergoing rapid and intense industrialization and urbanization over the past two decades. Primary, secondary, and tertiary industries currently coexist in the area.

2.2. Soil sampling and chemical analysis

A total of 105 topsoil samples (0–20 cm) were taken from the centers of the 1 km grid squares in October 2014. The average temperature was 19.6 °C during sampling period. Soil samples were collected on non-rainy days. The distribution of all samples is presented in Fig. 1. Each sample (about 1 kg dry weight) was a composite of five subsamples from nearby sites (approximately 5 m apart). Samples were packed into polyethylene bags and brought back to the lab. The soil samples were then air-dried, ground, sieved, and digested with a typical concentrated acid mixture (HNO_3, HF, and $HClO_4$). The details of the laboratory analyses of soil were the same as described in our previous research (Cao et al., 2010; Jiang et al., 2015). Concentrations of 15 elements (Mg, K, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Se, Cd, Sb, and Pb) were measured by inductively coupled plasma-mass spectrometry (ICP-MS; Agilent 7500a, USA). For quality control (QC) and quality assurance (QA), blank control, duplicate samples, and standard reference soils (GBW07419; Center for Certified Reference Materials, China) were used. The detection limits of Mg, K, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Se, Cd, Sb, and Pb were 0.228, 0.218, 0.010, 0.070, 0.009, 0.230, 0.010, 0.018, 0.023, 0.020, 0.009, 0.021, 0.010, 0.023, and 0.023 mg/kg, respectively. The results measured for the standard reference soils were within uncertainty ranges of the certified values. The relative standard deviations (RSD) for soil properties of duplicates were <3%.