Science of the Total Environment 512–513 (2015) 143–153

Contents lists available at ScienceDirect

Science of the Total Environment

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

Contamination features and health risk of soil heavy metals in China

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HIGHLIGHTS

- Soil contamination with heavy metals in China was systematically studied.
- Spatial distribution patterns of heavy metals in Chinese soils were identified.
- Monte-Carlo simulation was used to analysis the uncertainty of health risk model.

ARTICLE INFO

Article history:
Received 26 August 2014
Received in revised form 9 January 2015
Accepted 12 January 2015
Available online xxxx

Editor: F.M. Tack

Keywords:
Heavy metal
Soil contamination
Health risk assessment
Pollution index
Enrichment factor
Geoaccumulation index

ABSTRACT

China faces a big challenge of environmental deterioration amid its rapid economic development. To comprehensively identify the contamination characteristics of heavy metals in Chinese soils on a national scale, data set of the first national soil pollution survey was employed to evaluate the pollution levels using several pollution indicators (pollution index, geoaccumulation index and enrichment factor) and to quantify their exposure risks posed to human health with the risk assessment model recommended by the US Environmental Protection Agency. The results showed that, due to the drastically increased industrial operations and fast urban expansion, Chinese soils were contaminated by heavy metals in varying degrees. As a whole, the exposure risk levels of soil metals in China were tolerable or close to acceptable. Comparatively speaking, children and adult females were the relatively vulnerable populations for the non-carcinogenic and carcinogenic risks, respectively. Cadmium and mercury have been identified as the priority control metals due to their higher concentrations in soils or higher health risks posed to the public, as well as, arsenic, lead, chromium and nickel. Spatial distribution pattern analysis implied that the soil metal pollutions in southern provinces of China were relatively higher than that in other provinces, which would be related to the higher geochemical background in southwest regions and the increasing human activities in southeast areas. Meanwhile, it should be noticed that Beijing, the capital of China, also has been labeled as the priority control province for its higher mercury concentration. These results will provide basic information for the improvement of soil environment management and heavy metal pollution prevention and control in China.

1. Introduction

In general, heavy metals, such as cadmium (Cd), chromium (Cr), mercury (Hg), lead (Pb), copper (Cu), zinc (Zn) and nickel (Ni), refer to metals having densities greater than 5 g/cm^3. Due to similarities in chemical properties and environmental behavior, metalloid arsenic (As) often falls into the heavy metal category. Heavy metals and their compounds are naturally ubiquitous throughout the soil environment (Hu and Cheng, 2013). They are introduced naturally through the weathering of the parent materials, and also resulted from a variety of human activities such as mining, smelting, electroplating, and other industrial processes that have metal residues in their waste streams (Alloway, 1995; Nriagu, 1996). When heavy metals in soils have been transformed from solid form into either ionic moieties or through biomethylation to organometallic moieties, they could have caused threat to the health of animals and human beings (Madrid et al., 2002). For instance, the chronic effects of Cd dust or aerosol particulate matter through soil ingestion consist of lung cancer, pulmonary adenocarcinomas, prostatic proliferative lesions, bone fractures, kidney dysfunction, and hypertension, while chronic exposure to As dust can have adverse effects such as dermal lesions, peripheral neuropathy, skin cancer, and peripheral vascular disease (Żukowska and Biziuk, 2008). Another example, the excessive intake of Pb (PM2.5, Pb^{2+} ions or organolead) can damage the nervous, skeletal, circulatory, enzymatic, endocrine, and immune systems (Li et al., 2014). Due to their potential toxic, persistent and irreversible characteristic, the heavy metals such as

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http://dx.doi.org/10.1016/j.scitotenv.2015.01.025
0048-9697/© 2015 Published by Elsevier B.V.

Cd, Cr, As, Hg, Pb, Cu, Zn and Ni have been listed as priory control pollutants by the United States Environmental Protection Agency (USEPA) and caused more and more attention in many part of the world (Giller and McGrath, 1988; Abrahams, 2002; Rodrigues et al., 2013).

With the developing industrial operations and fast urban expansion since the economic reform and opening that began in the late 1970s, soil pollution by heavy metals has been both serious and widespread in China (Chen et al., 1999; Wang et al., 2001; Cheng, 2003). According to the State Environmental Protection Administration, China faces serious soil metal pollution (CSC, 2012). In China, about 10 million ha of arable land have been polluted, and ~12 million tons of grains are contaminated each year by heavy metals in soils (Teng et al., 2010). The first national census of pollution sources in China also indicated that high levels of metal pollution in soils raised widespread concerns for food safety, particularly of rice (Brian, 2011).

However, until 2005, no national census data are available for the contents and distributions of main pollution components in soils of China, including the potentially toxic metals such as Cd, Cr, As, Hg, Pb, Cu, Zn and Ni. This deficiency is particularly serious considering the risk accumulation features of waste effluent present in soils and difficult to develop fit policies on soil pollution prevention and control. Therefore, Chinese government launched the first national soil pollution survey during April 2005 to December 2013 backed by a budget of about 125 million dollars (Teng et al., 2010). The purpose of the program conducted collaboratively by the Ministry of Environmental Protection and the Ministry of Land and Resources were to investigate soil pollution status in China and to establish regulations and standards for soil pollution management (Teng et al., 2014).

Understanding the contamination characteristics of heavy metals in soils and identifying their environmental exposure risks not only are the basic preconditions for soil pollution prevention and control, but also provide important information for making decisions for remediation of contaminated soils. In this study, based on the data set of the first national soil pollution survey, several pollution indicators (pollution index, geoaccumulation index and enrichment factor) and exposure risk model recommended by the US Environmental Protection Agency have been put forward for evaluating the pollution level of soil metals in China on a national scale and quantifying their exposure risks posed to human health. The additional objectives of this research are to screen the priority control soil metals and to identify the spatial distribution pattern of metal pollution in Chinese soils. The results presented here illustrate the general accumulation and risk features of heavy metal contamination in soils, and will be helpful for policy makers to formulate effective pollution control strategies.

2. Materials and methods

2.1. Sample collection and analysis

The scope of the first national soil pollution survey included the territory of the People's Republic of China (excluding the Hong Kong, Macao, and Taiwan areas). The systematic sampling pattern with regularly spaced intervals was employed, and the sampling network was designed to cover a wide range of determinants based on a regular grid which reasonably represented the nationwide soils. The randomly distributed sampling points covered the main land use types, including the cultivated land, woodland, grassland, construction land, and unused land, with the actual survey area of about 6.3 million km^2. More than 38,000 topsoil samples were collected during April 2005 to December 2013. All of soil samples were collected using a bamboo or stainless spade and stored in sealed kraft packages to avoid contamination. Preservation and transportation of the soil samples were executed according to the Technical Specification for SEPAC (State Environmental Protection Agency of China) (2004).

In the laboratory, soil samples were always air-dried at room temperature, and then sieved and pulverized. In general, soil samples were digested with a concentrated acid mixture (HNO_3, HF, and $HClO_4$) and then were treated by a variety of analytical methods (CEPA, 1995). A necessary analytical quality control method was designed and followed during the analysis through careful standardization, procedural blank measurements, and spiked and duplicate samples (Cheng et al., 2014).

The concentrations of the heavy metal ions of the 12 species (Cd, Hg, Pb, Cr, As, Cu, Zn, Ni, Mn, Co, Se, and V), TOC, and pH were measured for each soil sample, resulting in approximately 480 thousand analytical data points. In this study, 8 trace elements (Cd, Hg, Pb, Cr, As, Cu, Zn, and Ni) were chosen as analysis variables to identify the contamination characteristics of soil metals in China and evaluate their exposure risks posed to human health. Summary statistics of heavy metal concentrations are presented in Table 1.

2.2. Pollution indicators

Pollution index can be used to provide a relative ranking of contamination levels. On the other hand, geochemical approaches including geoaccumulation index and enrichment factor can be employed to assess the pollution status and to estimate the impact of anthropogenic activities (Lee et al., 2006; Loska et al., 2004).

2.2.1. Pollution index

According to Lee et al. (2006), pollution index (PI) is defined as: $PI = C_i/C_{0i}$, where C_i is the concentration of a given ith element in soil samples (mg/kg), and C_{0i} is its corresponding reference concentration (mg/kg). In addition, to give an assessment of the overall pollution status for a sample, the integrated pollution load index (PLI) or the Nemerow integrated pollution index (NIPI) (Nemerow, 1985) can be employed (Tomlinson et al., 1980; Lu et al., 2014; Luo et al., 2012). The PLI and NIPI can be calculated using $(PI_1 \times PI_2 \times PI_3 \times \ldots \times PI_n)^{1/n}$ and $[0.5 \times (I_{Avg}^2 + I_{Max}^2)]^{1/2}$, respectively, where I_{Avg} is the mean value of all pollution indexes of the metals considered, I_{Max} is the maximum value, n is the number of heavy metals.

According to Zhang et al. (2011), the PLI is divided into seven levels from none to high pollution to indicate the contamination degree and classified as either background concentration (PLI = 0), unpolluted ($0 < PLI \leq 1$), unpolluted to moderately polluted ($1 < PLI \leq 2$), moderately polluted ($2 < PLI \leq 3$), moderately to highly polluted ($3 < PLI \leq 4$), highly polluted ($4 < PLI \leq 5$), or very highly polluted ($PLI > 5$). However, the classification of NIPI is slightly different from the PLI levels, and can be graded as either safe ($NIPI \leq 0.7$), precaution ($0.7 < NIPI \leq 1$), slight pollution ($1 < NIPI \leq 2$), moderate pollution ($2 < NIPI \leq 3$), or heavy pollution ($NIPI > 3$) (Cheng et al., 2014).

2.2.2. Geoaccumulation index

The geoaccumulation index (I_{geo}) is a geochemical criterion to evaluate pollution level in soils or sediments and has been used since the late 1960s (Muller, 1969). It can be calculated using $I_{geo} = log_2(C_n/1.5B_n)$, where C_n is the measured concentration of the metal ions in

Table 1
Summary statistics of heavy metal concentrations in Chinese soils (n = 38,393) (mg/kg).

	Cd	Cr	As	Hg	Pb	Cu	Zn	Ni
10th	0.058	32.1	4.5	0.015	16.7	12.0	42.5	12.5
25th	0.092	48.9	6.8	0.026	20.9	17.8	55.1	20.3
50th	0.14	65.0	9.7	0.047	25.6	23.2	69.8	27.4
75th	0.214	79.8	13.1	0.093	32.4	30.0	88.9	34.3
90th	0.505	115.0	25.7	0.256	59.5	53.4	141.5	53.4
Mean	0.225	68.5	12.1	0.087	31.2	27.1	79.0	29.6
Average background values in China	0.097	61.0	11.2	0.065	26.0	22.6	74.2	26.9
Chinese soil guidelines (Grade I)	0.2	90	15	0.15	35	35	100	40
Chinese soil guidelines (Grade II)	0.6	200	30	0.5	300	200	250	50