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Heavy metals in soils from a typical county in Shanxi Province, China: Levels, sources and spatial distribution

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HIGHLIGHTS

- Heavy metals enriched in soils with different degrees, especially for Hg and Cd.
- Agricultural production and vehicular transport were the primary sources of heavy metals.
- Spatial distribution of heavy metals in soil was closely related with human activities.

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ABSTRACT

The concentrations of As, Cd, Cr, Cu, Pb, Ni, Zn, and Hg in 128 surface soil samples from Xiangfen County, Shanxi Province, China were measured. The concentrations of these eight heavy metals were lower than the critical values in the national soil quality standard. However, these concentrations were found to be slightly higher than their background values in soils in Shanxi Province, indicating enrichment of these metals in soils in Xiangfen County, especially for Hg and Cd. Principal component analysis coupled with cluster analysis was used to analyze the data and identify possible sources of these heavy metals; the results showed that the eight heavy metals in soils from Xiangfen County came from three different sources. Lead, Cd, Cu and Zn mainly arose from agricultural practices and vehicle emissions. Arsenic and Ni arose mainly from parent materials. Industrial practices were the main sources of Cr and Hg. The spatial distribution of the heavy metals varied greatly, and was closely correlated to local anthropogenic activities. This study will be helpful not only for improving local soil environmental quality but will also provide a basis for effectively targeting policies to protect soils from long-term heavy metal accumulation.

1. Introduction

Soil heavy metal pollution has become a severe problem in many parts of the world. Due to advancing industrialization and urbanization over the past few decades, soil pollution by heavy metals has been both serious and widespread in China (He et al., 2013). Heavy metals do not easily migrate or break down through natural degradation processes, but instead accumulate within the soil over time, which can directly affect the physical and chemical properties of the soil by inhibiting soil microbial activity and hindering effective nutrient supply (Nicholson et al., 2003; Khan et al., 2008). In addition, the presence of heavy metals in soils presents an increasing risk to human health, since metal ions can be absorbed through dust inhalation and food intake (Tipping et al., 2003).

Heavy metals are natural components of the earth's crust, and natural concentrations of heavy metals in soils tend to remain low. However, anthropogenic inputs of several heavy metals to soils greatly exceed natural inputs from pedogenesis (Facchinelli et al., 2001). A 6-year soil pollution study conducted by the Chinese government found that the country's soil had been heavily polluted by human activities (CSC (China State Council), 2012), and the dominant sources of soil heavy metal pollution in China were fertilization, sewage irrigation, sludge application, and metallic ore mining and smelting operations (Cox et al., 2004).

Shanxi Province is well-known for its coal resources. In 2003, Shanxi Province produced 300 million tons of coal (Zhang et al., 2009). Mining, including coal mining, is considered to be one of the most significant sources of heavy metal contamination (Acosta et al., 2011). In addition, other potential sources of heavy metals are

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present in the region, such as coking industry, smelting industry, and so on (Wang et al., 2001). Shanxi Province is also an important farming region in China, and agricultural practices can also result in heavy metal enrichment in soils (Nicholson et al., 2003). To better understand soil heavy metal pollution in Shanxi Province, a study was undertaken in Xiangfen County, located in the south of Shanxi Province, with the objective of investigating concentrations, potential sources and spatial distribution of heavy metals in soils. This study will provide a useful reference for environmental protection in this region.

2. Materials and methods

2.1. Soil sampling

Xiangfen County is located in southern Shanxi Province, China, which has a total population of approximately 0.5 million and an area of 1034 km^2. The county's terrain is mostly covered with flat land, and the terrain slopes gently from north to south. The climate belongs to warm temperate semi-arid continental monsoon climate zone, characterized by hot and humid summer and generally cold and dry winter. The average annual temperature and rainfall is 12.4 °C and 508.6 mm, respectively. Cinnamon soil is the main soil type in the study area. Xiangfen County has developed agricultural activities, with the main production of grain, cotton, vegetable and fruit. The main industries including coking, iron and steel, cement, foundry etc. (XACC (Xiangfen Annals Compilation Committee), 2011).

The soil sample collection method used was that described by Pan et al. (2015). Briefly, in May 2012, a total of 128 surface (0–20 cm) soil samples were collected from across Xiangfen County using a gridded sampling design with a grid spacing of approximately 3 km to represent the whole area (Fig. S1 in Supplementary Material). Some sites in mountainous or other unsuitable regions were modified based on convenience for vehicular access. At each sampling site, five subsamples were taken from the same area (approximately 100 m^2) and pooled together to form one composite sample.

2.2. Sample preparation and analysis

All samples were air-dried for 1 week at room temperature in a storage room, sieved into 60-mesh size particles after removing stones, residual roots, and other unwanted materials, and then sealed in brown glass bottles and conserved in a refrigerator at −4 °C until analysis. Samples were analyzed following methods 3050B and 6010C suggested by USEPA (1996a, b), respectively. In brief, approximately 10 g of sample were air-dried, gently ground with an agate pestle, and passed through a 100-mesh nylon sieve. 1 g of sample was put into Anton PVC digestion vessels along with 9 mL concentrated nitric acid (HNO_3) and 3 mL hydrogen peroxide (H_2O_2), then sealed and heated at 180 °C for 15 min. The concentrations of eight heavy metals, arsenic, Cd, Cr, Cu, Pb, Ni, Zn and Hg, in the digestion solution were determined using inductively coupled plasma mass spectrometry (ICP-MS, POEMS3, Thermo Electron, USA).

2.3. Quality assurance and quality control

Soil standard reference material (GBW07401, GSS-1) obtained from the Center of National Standard Reference Material of China was used for quality assurance and quality control (QA/QC). Accepted recoveries ranged from 81.0 to 109%. Analysis methods were evaluated in blank ($n = 7$) and duplicate samples ($n = 13$) for each set of samples. The relative deviation of the duplicate samples was <7% for all batch treatments.

2.4. Data analysis

Data analysis was performed using SPSS version 18.0 (IBM, Chicago, Illinois, USA). Descriptive statistics used for the analysis of the heavy metals in the soil samples included maximum, minimum, median, mean, standard deviation (SD). We used Spearman correlation analysis, principal component analysis (PCA) and cluster analysis (CA) of the classic multivariate statistical method to process data and identify potential sources of the eight heavy metals. In the PCA, varimax was used as the rotation method in the analysis following standardization of the data. Spearman correlation coefficients were obtained using the bivariate procedure in SPSS. A two-tailed P value of <0.05 was considered to indicate statistical significance. After log-transformation and normalization of all measured samples, the spatial distribution of heavy metals in Xiangfen County was visualized using ArcGIS version 9.0 (ESRI, Redlands, California, USA).

3. Results and discussion

3.1. Degree of heavy metal pollution

The descriptive statistical results for eight heavy metals in soil samples from Xiangfen County were presented in Table 1 and Fig. 1.

As shown in Table 1 and Fig. 1, the concentrations of eight heavy metals varied greatly; Znic was found in the highest concentrations, followed by Cr. Measured concentrations of Cu, Pb and Ni were relatively similar. Concentrations of Cd and Hg were considerably lower than those of other metals. The average concentrations of As, Cd, Cr, Cu, Pb, Ni, Zn and Hg were 14, 0.20, 71, 30, 23, 32, 82, and 0.13 mg kg^{-1}, respectively. Background concentrations of these heavy metals in the soils of Shanxi Province are 9.2, 0.07, 53, 20, 24, 23, 68 and 0.04 mg kg^{-1} for As, Cd, Cr, Cu, Pb, Ni, Zn and Hg, respectively (CNEMC (China National Environmental Monitoring Center), 1990). Enrichment factor (EF) can be utilized to evaluate the degree of anthropogenic influence on soil contamination by heavy metals, and to differentiate the metals originating mainly from human activities and those from natural sources (Ye et al., 2011). In the present study, EFs for the metals were 1.52, 2.86, 1.34, 1.50, 0.96, 1.39, 1.21 and 3.25 for As, Cd, Cr, Cu, Pb, Ni, Zn and Hg, respectively. EF values of these metals follow a descending order as Hg>Cd>As>Cu>Ni>Cr>Zn>Pb. According to Sutherland (2000), EF values are always divided into several grades: EF<1 indicates no pollution, 1<EF<2 indicates slight pollution, 2<EF<5 indicates moderate pollution. Hence, the results suggested slightly pollution of As, Cr, Cu, Ni, and Zn, moderate pollution of Hg and Cd in the study area, which indicate significant human input of heavy metals to soils in Xiangfen County, especially for Hg and Cd. The Chinese Environmental Quality Standard for Soils (CEPA (Chinese Environmental Protection Administration), 1995) provides critical

Table 1
Descriptive statistics of heavy metal concentrations in soil samples (mg kg^{-1}).

	Maximum	Minimum	Median	Mean	SD
As	31.0	9.0	14.0	14.0	2.71
Cd	0.65	0.03	0.19	0.20	0.09
Cr	113	54	70	71	8.38
Cu	64	21	29	30	5.70
Pb	66	16	23	24	5.19
Ni	43	26	32	32	3.04
Zn	139	64	79	83	12.95
Hg	3.28	0.00	0.06	0.13	0.36