

Spatial distribution and sources of trace elements in surface soils, Changchun, China: Insights from stochastic models and geostatistical analyses



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ABSTRACT

Trace elements As, Cd, Cu, Hg, and Pb in soils originate from both natural and anthropogenic sources, and intense anthropogenic activity can lead to excessive trace element levels, which are harmful to the environment. We investigated the trace elements in the topsoil of Changchun, China, using stochastic models and the indicator kriging method. The influence of anthropogenic and natural variables on the distribution of trace elements was evaluated, with the proportions of background and anthropogenic sources identified using stochastic models. High-risk maps were constructed using indicator kriging. The results showed that the anthropogenic variables significantly influenced the spatial variations of trace elements. The relative influence of natural variables differed for the different elements. The median values of background As, Cd, Cu, Hg, and Pb were 10.55 mg kg⁻¹, 0.114 mg kg⁻¹, 21.80 mg kg⁻¹, 0.036 mg kg⁻¹, and 25.88 mg kg⁻¹, respectively; while the threshold values were 12.90 mg kg⁻¹, 0.200 mg kg⁻¹, 29.40 mg kg⁻¹, 0.046 mg kg⁻¹, and 29.49 mg kg⁻¹, respectively. The relative contributions of background values to total values of As, Cd, and Cu were greater than 95%, while those for Pb were 91%. In contrast, Hg showed serious pollution, with less than 75% of the total Hg derived from background sources. The geographically extensive Hg pollution is likely influenced by emissions from coal-fired plants and boilers, while other trace elemental pollutants primarily originate from factory emissions. Furthermore, the atmospheric diffusion of Pb and Hg is significant, resulting in their migration to agricultural areas.

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1. Introduction

Trace elements As, Cd, Cu, Hg and Pb, in soils are toxic, biologically available, and non-degradable (Lee et al., 2001; Li et al., 2001; Möller et al., 2005; Gutiérrez-Caminero et al., 2015; Xiao et al., 2015); therefore, it is necessary to understand their behavior, spatial variation, pollution condition, and sources. Trace elements in surface soils originate from both natural and anthropogenic processes (e.g., traffic and industrial emissions) (Nriagu and Pacyna, 1988; Baize and Sterckeman, 2001; Hojdova et al., 2008; Li et al., 2013), which lead to spatial variations in trace element levels (Bocchi et al., 2000; Zhao et al., 2009; Lin, 2002; Burak et al., 2010; Zhao et al., 2010).

Soils originate from the weathering of bedrock; therefore, the concentrations of elements in soils are inherited from the bedrock source (Alloway, 2012; Ballesta et al., 2010). When there are multiple soil parent materials (SPM), the concentrations of natural trace elements in soil may show marked spatial variation (Facchinelli et al., 2001). The physical and chemical properties of soils are controlled by climate, soil formation, land use and landform (Fritz and Mohr, 1984;

Middelburg et al., 1988; Breemen and Buurman, 2002; De Temmerman et al., 2003; Sariyildiz et al., 2005; Seibert et al., 2007; Cai et al., 2010), with different conditions leading to variations in the contents of trace elements. Soil pH is the main index representing soil properties, as it significantly influences many chemical and physical reactions (e.g., oxidation–reduction, adsorption, desorption, and coordination) (Zeng et al., 2011). Soil organic carbons depend on climate, overlying plants, terrain features, soil structure, etc. Soils with high organic carbon content are more preservative; thus, they accumulate more trace elements (Chen, 2005). From the perspective of anthropogenic activities, population density, road density, and the distribution of industrial sites are all thought to influence the distributions of trace elements in surface soils.

Due to the different geochemical properties of trace elements, they are influenced differently by different environmental variables. Quantitative research into the factors controlling elemental spatial variations is of great significance for understanding elemental geochemical characteristics. However, few studies have investigated the topsoil in Changchun, China. In this study, we applied the random forest method to fulfill the quantitative evaluation on elemental distribution. It is a powerful, predictive, model-average technique. In this method, random samples of predictive variables are used to generate the “forest,” which

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can be used to calculate the relative importance of predictive variables (Johnstone et al., 2014; Vitale et al., 2014).

In areas of intense human activity (e.g., urban and industrial areas), trace element accumulation can be significant (Rehman et al., 2008; Kien et al., 2010). Determining the geochemical backgrounds and thresholds of trace elements in soils and identifying the concentrations of point source pollutions are key processes in soil environmental quality assessment. The soil geochemical background traditionally refers to concentration of trace element formed naturally (Salminen and Gregorauskien, 2000); however, as it is usually difficult to find soils that have not been impacted by the atmospheric transportation of pollutants (Chen et al., 1999), it has become common to consider an artificial background (i.e., the sum of the natural background and the unavoidable diffuse anthropogenic concentration) (Gough et al., 1988; Holmgren et al., 1993; Tack et al., 1997). In contrast, point source pollution is the combination of the background with intensive trace element contamination.

The probability density functions of natural background, artificial background, and point source pollution differ. Finite mixture distribution models (FMDM), which assume that observed data come from random observations of a mixed distribution, can be used to calculate the proportion of a subpopulation (McLachlan and Peel, 2004; Lin et al., 2010). The total probability density functions of elements can be obtained by collecting samples from topsoils in both urban and agricultural regions. Using an FMDM, the different probability distributions can be separated, providing information about geochemical backgrounds and point source pollution, as well as threshold values (Yang and Chang, 2005).

Delineating the spatial distribution patterns of elements and determining the positions and ranges of over-limit samples in soils is important for environmental management. The indicator kriging method is a nonlinear technique that can be used to estimate the cumulative frequency of data larger than a threshold value (Kuisi et al., 2009; Dash et al., 2010; Hangen et al., 2010; Arslan, 2012; Lin et al., 2011). The method is robust to outliers and skewed distributions, and for this reason, we applied this method to construct high-risk maps.

2. Materials and methods

2.1. Overview of the study area

Changchun, the capital of Jilin province, is located in the northeastern plain of China (Fig. 1), on the eastern Eurasia continent (E124°18'–

127°02', N43°05'–45°15'), and represents the old industrial zone of northeast China. Industrial infrastructure in the downtown area includes petrochemical factories, machine factories, and smelting plants. The surrounding agricultural area is an important base for commodity grains, including corn and soybean. The region has a temperate continental monsoon climate with an annual mean temperature of 4.5 °C, and an annual mean rainfall of 668.4 mm.

2.2. Sampling and analysis

A total of 300 surface soil samples (0–20 cm) were collected using grid-based sampling at 1 sample per 16 km². Each sample was composed of four sub-samples collecting randomly within each grid. Each sample weighed more than 1 kg. After natural drying and the removal of gravel and plant roots, samples were sieved using a 20-mesh screen and then sent to a laboratory to be further processed to 200 mesh.

All analyses were carried out in the Ministry of Land and Resources Supervision and Inspection Center, Changchun. The concentrations of As and Hg were analyzed by atomic fluorescence spectrometry (AFS) and the detection limits were 0.9 mg kg⁻¹ and 0.003 mg kg⁻¹, respectively. Samples were prepared by mixing 0.5 g of soil with 5.0 ml of nitric and hydrochloric acid in a 25 ml colorimetric tube and shaken well. Samples were then dissolved in boiling water for 2 h, during which they were shaken every 15 min. Next, samples were cooled and then diluted to scale. Finally, the solution was filtered through dry filtration and the filtrate was used for the determination of As and Hg.

Concentrations of Cd were determined using graphite furnace atomic absorption spectrometry (GF-AAS) with a detection limit 0.02 mg kg⁻¹. Samples were prepared by placing 0.3 g of soil into a 50 ml polytetrafluoroethylene crucible, wetting with water, and then adding 5 ml of hydrochloric acid. Samples were then decomposed on an electric heating plate under a fume hood on low heat. Following evaporation to ~2–3 ml, samples were removed and cooled, before 5 ml of nitric acid, 4 ml of hydrofluoric acid, and 2 ml of perchloric acid were added. Samples were then returned to the electric heating plate at high temperature for 1 h. After 1 h, the cover was opened and heating was continued in order to remove Si. According to the condition of digestion, 2 ml of nitric acid, 2 ml of hydrofluoric acid, and 1 ml of perchloric acid were added and the above process was repeated. The mixture was then dissolved in 1 ml of nitric acid and transferred to a 25 ml flask, where 3 ml of diammonium phosphate solution was

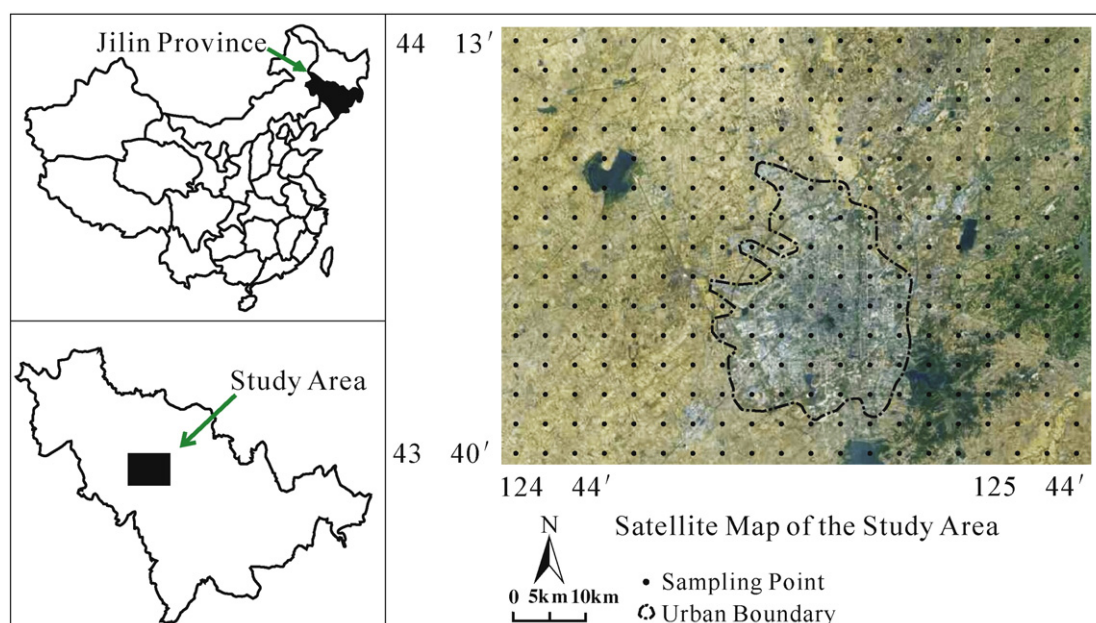


Fig. 1. Sampling locations and map of the study area.

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