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Assessment of soil heavy metal pollution using stochastic site indicators

Junyu He[a], Yong Yang[b,*], George Christakos[a,c], Yajun Liu[b], Xue Yang[b]

[a] Ocean College, Zhejiang University, Zhoushan, China
[b] College of Resources & Environment, Huazhong Agricultural University, Wuhan, China
[c] Department of Geography, San Diego State University, California, USA

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ABSTRACT

Improving the understanding and characterization of spatial soil heavy metal distribution is becoming an important component of risk assessment and environmental policy. In this work, 213 soil samples collected from Daye (Hubei Province, China) were used as the empirical dataset. First, maps of soil heavy metal distributions, including Cd, Co, Cr, Cu, Mn, Ni, Pb and Zn, were obtained using the ordinary Kriging method. Then, the pollution index (PI) and integrated pollution index (*IPI*) were calculated based on the ordinary Kriging maps to obtain a comprehensive quantitative pollution characterization of the eight heavy metals in the Daye soil. The results showed that 46.1%, 32.1%, and 0.5% of the soil in the study region are moderately, highly and extremely polluted, respectively. Finally, the one- and two-point stochastic site indicators of *IPI* were used to assess quantitatively the uncertainties and risks associated with soil heavy metal distributions in the polluted regions. These results showed that the *IPI* values exceeding a specified threshold increased almost linearly with increasing threshold value, whereas the relative area of excess pollution decreased steadily with increasing threshold. Among the site pairs considered in the study region, about 70% and 26% of them simultaneously experienced moderate and high pollution risk, respectively.

1. Introduction

Soils located in the interactive zone among the lithosphere, atmosphere hydrosphere and biosphere constitute the main part of the terrestrial ecosystem (Coskun et al., 2006). With strong stability and toxicity, heavy metals in soils can migrate and transfer by means of natural processes or anthropogenic activities in environmental and ecological chains on a large scale, threatening the safety of water resources (both surface and subsurface) and food, and even human health (Chabukdhara and Nema, 2013). At present, soil pollution is a serious problem in many cities, especially in industrialized and mining regions (Kodirov and Shukurov, 2009). Accordingly, many studies have focused on regional soil heavy metals, including pollution assessment, spatial distribution, and source apportionment (Yang et al., 2016; Xu and Zhang, 2017; Xiao et al., 2017). In these studies, a number of quantitative indices have been employed to evaluate pollution level in soils due to single or multiple heavy metal elements. These indices include the geoaccumulation index (Müller, 1969), the enrichment factor (Sutherland, 2000), the Nemerow pollution index (Zhong et al., 2010), the potential ecological risk (Håkanson, 1980), the contamination security index (Pejman et al., 2015), the pollution index and the integrated pollution index (Chen et al., 2003). Based on these indices, heavy metal pollution assessment in urban soils (Sun et al., 2010; Rizo et al., 2011), road dusts (Zhu et al., 2008), agricultural soils (Cui et al., 2014; Marrugo-Negrete et al., 2017), industrial or mining area soils (Machender et al., 2011; Ogunkunle and Fatoba, 2013; Yang et al., 2016), and soils in other kind of regions (Sheng et al., 2012) have been studied around the world.

However, in most of the existing studies the values of those indices are calculated only at the soil sampling points, meaning that the seriousness of soil heavy metal pollution is not assessed at unsampled areas of considerable size. In such cases, spatial interpolation methods (e.g., ordinary Kriging) can provide a useful tool to estimate the concentrations of soil heavy metals at unsampled sites. However,

Abbreviations: SSI, stochastic site indicators; PI, pollution index; IPI, integrated pollution index; OK, ordinary Kriging; SRF, spatial random filed; RAEP, relative area of excess pollution; MEP, mean excess pollution; MEDP, mean excess differential pollution; CMEP, conditional mean excess pollution; PID, pollutant indicator dispersion; NIC, noncentered indicator covariance; CIC, centered indicator covariance; NEC, noncentered excess covariance; EDC, excess differential covariance; CEC, conditional excess covariance; IR, interaction ratio; SD, standard deviation; CV, coefficients of variation; BGV, background values; PSPBB, percentage of sampling points beyond BGV; MAE, mean absolute error; ER, error rate

* Corresponding author at: Huazhong Agricultural University, No.1 Shizishan Street, Hongshan District, Wuhan, Hubei Province 430070, China.
E-mail address: yangyong@mail.hzau.edu.cn (Y. Yang).

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interpolation methods operate in conditions of in situ uncertainty, and, thus, environmental decisions are also subject to uncertainty (Christakos, 1985; Christakos and Killam, 1993; Smith et al., 1993). Therefore, it is necessary to identify risk regions and reduce uncertainties with some quantitative indicators.

In view of the above considerations, the present work has two main goals: the first goal is to assess heavy metal contamination in the soils of the study region (Daye city, Hubei province, China) using the pollution and integrated pollution indices of eight different heavy metal elements estimated by spatial interpolation (ordinary Kriging); and the second goal is to assess quantitatively the pollution risk in the study region based on one-point and two-points stochastic site indicators.

2. Materials and methods

2.1. Study region and sample collection

The city of Daye (latitude 29°40′—30°15′N, longitude 114°31′—115°20′E) is located in the southeast of Hubei province and the south bank of the middle reaches of the Yangtze River. It is a famous mining city in China –its mining and smelting history can be traced back to 3000 years ago. Now, due to the rich mineral resources, there are still a larger number of mines (coal, copper, iron, and gold) and smelting factories inside and outside Daye city. Unlike other industrial and mining areas, the industrial and mining enterprises in Daye city are mixed with commercial, residential and agricultural area. Thus, heavy metals in the soils can have a great impact on the health of the local population.

In order to assess soil heavy metal pollution, a total of 213 topsoil samples (0–20 cm depth) were collected in the industrial, mining and population concentrated areas of Daye city during September 2016 (Fig. 1). The sampling sites were determined randomly, with an average distance between sites approximately equal to 500 m. Following the pre-processing stage (Yang et al., 2016), the concentrations of each one of the eight heavy metal elements (Cd, Co, Cr, Cu, Mn, Ni, Pb, Zn) were measured using inductively coupled plasma mass spectrometry (ICP-MS).

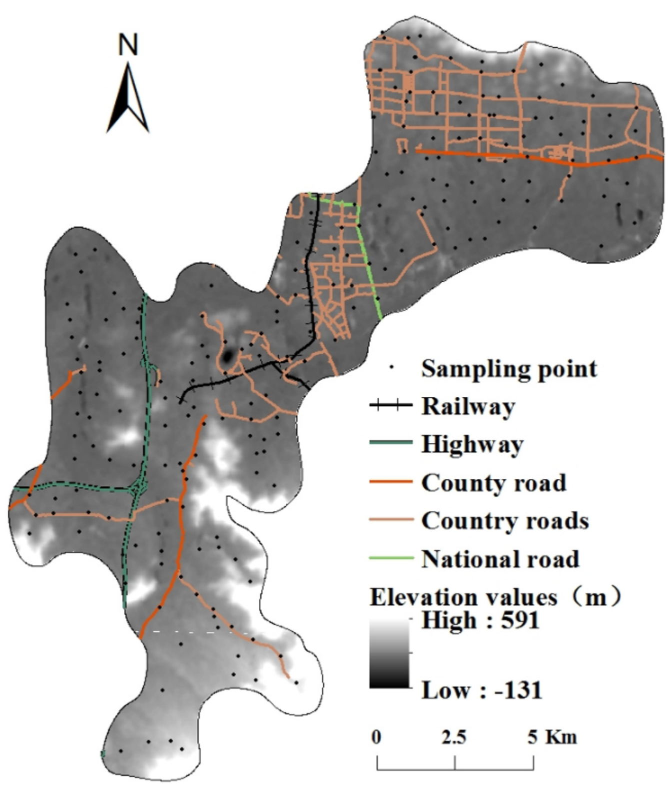

Fig. 1. Map of soil sampling sites, roads and elevation.

2.2. Data analysis and spatial distribution maps

After removing the abnormal values, a standard statistical analysis was carried out to describe soil heavy metal contents. The Kolmogorov-Smirnov (K–S) test was used to determine whether the original data of various heavy metals followed a normal distribution. Data transformation was applied in the original data if the K–S value was < 0.05. Then, the ordinary Kriging method (OK, Webster and Oliver, 2007) was used to obtain the spatial distribution of each heavy metal in the soil. In order to assess the spatial interpolation (OK) accuracy, the standard sample-based cross-validation technique was implemented. Based on the leave one out cross validation technique, two accuracy indicators were computed from the pairs of "estimated-observed" soil heavy metal concentrations at the sampling points: the Pearson correlation coefficient (r), and the mean absolute error (MAE). The r and MAE were used to measure the strength of the linear relation and mean absolute deviation between the estimated and the observed soil heavy metal concentrations, respectively. For an accurate spatial interpolation, the r should be close to 1 and the MAE should be as small as possible. In addition, in order to remove any quantitative difference among the various heavy metals in soils, an error rate (ER) was defined as the ratio of the MAE over the mean value of the heavy metals.

2.3. Heavy metals pollution assessment

Based on the spatial distribution of each heavy metal in soils, the pollution index (PI) and the integrated pollution index (*IPI*) were used to assess the pollution impact for each heavy metal separately and the eight heavy metals as a whole, respectively. The PI was defined as the ratio

$$PI(\boldsymbol{s}) = \frac{C(\boldsymbol{s})}{B} \tag{1}$$

where $C(\boldsymbol{s})$ is the concentration of each heavy metal at the spatial location $\boldsymbol{s}$ with coordinates s_1 and s_2, i.e., $\boldsymbol{s} = (s_1, s_2)$, and B is the corresponding background value. Then, based on the PI results, the pollution levels for each metal were divided into three classes: $PI(\boldsymbol{s}) \le 1$, low pollution at location $\boldsymbol{s}$; $1 \le PI(\boldsymbol{s}) \le 3$, moderate pollution at location $\boldsymbol{s}$; $PI(\boldsymbol{s}) \ge 3$, high pollution at location $\boldsymbol{s}$. The IPI was defined as the mean value of the PIs of the eight metals considered,

$$IPI(\boldsymbol{s}) = \frac{1}{N} \sum_{i=1}^{N} PI_i(\boldsymbol{s}) \tag{2}$$

(in this study $N = 8$). Based on the IPI value, and following the classification proposed by Wei and Yang (2010), the soils were classified into four categories: $IPI \le 1$ (slightly polluted soil); $1 < IPI \le 2$ (moderately polluted soil); $2 < IPI \le 5$ (highly polluted soil); $IPI > 5$ (extremely polluted soil).

2.4. Stochastic site indicators

Stochastic site indicators (SSI) describing the pollution state of a region under conditions of uncertainty were proposed by Christakos and Hristopulos (1996a, 1996b, 1997). In this work, the goal is to assess quantitatively the soil heavy metal risks in the polluted region of interest, in which case the SSI of the IPI variation were calculated as follows.

Let us define $IPI(\boldsymbol{s}) = IPI(s_1, s_2)$ as the spatial random filed model (SRF, Christakos, 1992) representing the spatial variation of each IPI within the domain D. A spatial indicator random field is defined in terms of $IPI(\boldsymbol{s})$ as