

Use of portable X-ray fluorescence spectroscopy and geostatistics for health risk assessment



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ABSTRACT

Laboratory analysis of trace metals using inductively coupled plasma (ICP) spectroscopy is not cost effective, and the complex spatial distribution of soil trace metals makes their spatial analysis and prediction problematic. Thus, for the health risk assessment of exposure to trace metals in soils, portable X-ray fluorescence (PXRF) spectroscopy was used to replace ICP spectroscopy for metal analysis, and robust geostatistical methods were used to identify spatial outliers in trace metal concentrations and to map trace metal distributions. A case study was carried out around an industrial area in Nanjing, China. The results showed that PXRF spectroscopy provided results for trace metal (Cu, Ni, Pb and Zn) levels comparable to ICP spectroscopy. The results of the health risk assessment showed that Ni posed a higher non-carcinogenic risk than Cu, Pb and Zn, indicating a higher priority of concern than the other elements. Sampling locations associated with adverse health effects were identified as 'hotspots', and high-risk areas were delineated from risk maps. These 'hotspots' and high-risk areas were in close proximity to and downwind from petrochemical plants, indicating the dominant role of industrial activities as the major sources of trace metals in soils. The approach used in this study could be adopted as a cost-effective methodology for screening 'hotspots' and priority areas of concern for cost-efficient health risk management.

1. Introduction

Substantially increased industrial activities have resulted in soil trace metal contamination (Chen et al., 2015). Exposure to trace metals above certain concentrations can pose significant risks to human health (Z. Li et al., 2014; Peña-Fernández et al., 2014; Xiao et al., 2015). It is important that the necessary data are available for health risk assessments and to inform the decision-making regarding whether soil remediation is required or not; one barrier to effective decision-making is the lack of cost-effective approaches for assessment (Horta et al., 2015).

Conventional laboratory analyses (e.g., atomic absorption spectroscopy (AAS) or inductively coupled plasma (ICP) spectroscopy) of soil trace metals are time consuming, are costly and can result in sample destruction, limiting further analysis (Shuttleworth et al., 2014). Consequently, there is a need for fast and cost-effective analytical methods to measure concentrations of metals in soils. Portable X-ray fluorescence (PXRF) spectroscopy exactly meets these requirements (Parsons et al., 2013; Shuttleworth et al., 2014). Compared with conventional

laboratory analyses, elemental concentrations in soils measured via PXRF spectroscopy have acceptable quality (Parsons et al., 2013; Radu and Diamond, 2009). PXRF spectroscopy is often used for screening toxic elements in soils (e.g., Guerra et al., 2017; Lagerström et al., 2016; McComb et al., 2014; Peinado et al., 2010; Rouillon et al., 2017) or in combination with spatial interpolation techniques for identifying contaminated areas (e.g., Carr et al., 2008; Dao et al., 2014; Paulette et al., 2015; Suh et al., 2016; Weindorf et al., 2012).

The pollution sources of trace metals have different emission modes and intensities; therefore, each source has its own spatial scale of influence (Aboal et al., 2006). Some sources may only affect the local area, but other sources may affect the entire area (Buttafuoco et al., 2017; Guagliardi et al., 2016; Lacarce et al., 2012). These nested pollution processes form a hierarchical spatial structure of trace metals (Aboal et al., 2006; Li et al., 2013; Ordóñez et al., 2015; Yang et al., 2014). Therefore, for health risk assessment, it is not enough to assume that the entire population is exposed to the same soil contaminant level (Gay and Korre, 2006). It is essential to capture the heterogeneous

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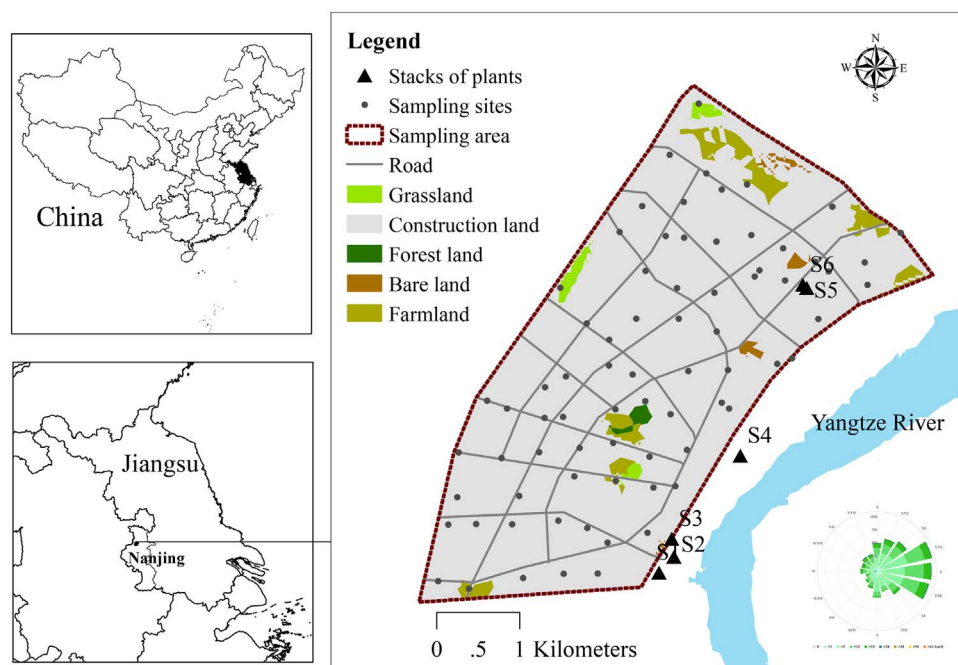


Fig. 1. Study area, soil sampling locations and stacks of industrial plants. S1 is a steel plant, S2 and S3 are power plants, and S4, S5 and S6 are petrochemical plants.

spatial distribution of soil trace metals (Huang et al., 2016b; Li et al., 2017).

Geostatistical methods provide a valuable tool for studying the spatial structure of trace metals and for mapping their spatial distribution (Guagliardi et al., 2015; Minasny and McBratney, 2016). These methods allow one to isolate sources of variation acting at different spatial scales, including local anthropogenic processes that may contribute to spatial outliers (Aboal et al., 2006; Buttafuoco et al., 2017; Guagliardi et al., 2013). Spatial outliers or anomalies exhibit extreme values of soil trace metals in their spatial contexts (Sun et al., 2013; Buttafuoco et al., 2017), and they often cannot be discarded since they may indicate ‘hotspots’ (Saby et al., 2011; Zhang et al., 2009). Trace metal contaminant data commonly have highly skewed distributions due to outliers, which are challenging to analyze and map (Meklit et al., 2009; Saby et al., 2011; Zhang et al., 2009). Care should be taken when geostatistics is used in cases where outliers are seen because, when using kriging to estimate the value of soil properties, outliers may result in incorrect variances (Lark, 2000; Saby et al., 2011). Robust geostatistical methods can identify and process these outliers and, thus, avoid a smoothing effect due to directly discarding the outliers (Lark, 2002; Saby et al., 2011). Robust estimators of variograms provide us good protection against the effects of outlying data (Hawkins and Cressie, 1984; Lark, 2000). Lark (2002) proposed a procedure for identifying spatial outliers. Hawkins and Cressie (1984) proposed a robust median kriging procedure that winsorized the observations so that the component due to anomalous contaminating processes was removed. These methods have been widely used in the study of trace metal pollution in soils (Lacarcé et al., 2012; Marchant et al., 2010; Saby et al., 2011; Zhao et al., 2007).

Based on the spatial distribution of soil trace metals, priority regions of concern and ‘hotspots’ can be identified. This is often done by comparing the calculated results with a critical threshold that might cause concern for human health (Buttafuoco et al., 2016; Guagliardi et al., 2015; Huang et al., 2016a). This process is critical because it provides reference for cost-efficient risk management policy-making (Huang et al., 2016a; Li et al., 2017).

Horta et al. (2015) proposed that integrating field spectroscopy and spatial analysis can enhance the assessment of soil contamination. Considering this, we combined PXRF spectroscopy and geostatistics for the health risk assessment of exposure to trace metals in soils in this

study. To our knowledge, there are few reports using PXRF spectroscopy for the assessment of human health risk. Itai et al. (2014) assessed non-cancer risk according to the ingestion of a soil/ash mixture from e-waste recycling sites. However, they did not use spatial interpolation techniques to identify high-risk areas. Recently, Urrutia-Goyes et al. (2017) made a map of non-carcinogenic risk, but they did not consider the influence of spatial outliers.

Considering these backgrounds, for health risk assessment, we suggest using PXRF spectroscopy to replace ICP spectroscopy for metal analysis in combination with robust geostatistical methods to identify spatial outliers and map trace metal distributions. A case study was carried out around an industrial area in Nanjing, China. The major objective of this study was to assess whether this suggestion is promising or not from three aspects: assessing the accuracy and precision of PXRF spectroscopy for the analysis of trace metals (Cr, Cu, Ni, Pb and Zn) in soils; identifying high-risk sites and areas that need to be investigated in detail; and determining potential pollution sources. If deemed promising, the results of this study could be adopted as a cost-effective methodology for screening high-risk sites and areas from pre-investigated regions.

2. Materials and methods

2.1. Study area and soil sampling

The study area is located in the northern part of Nanjing, Jiangsu Province, China (Fig. 1). The geomorphic unit of the study area is the alluvial plain of the Yangtze River with an elevation typically no greater than 10 m above sea level (Zhao et al., 2010). The geologic setting in the study area is Holocene Yangtze River alluvium (Wang et al., 2011). The soil parent material is the alluvium from the Yangtze River (Zhao et al., 2010), and the main soil type is anthrosol (Zhang et al., 2007). The study area covers approximately 30 km², and the resident population is approximately 190,000. It is one of the earliest chemical industrial bases in China, with its first chemical plant having been built in the 1930s. The plants are located along the eastern side of the study area (Fig. 1). The stacks of plants, as elevated emission sources, emit pollutants that enter the soil via atmospheric deposition, and thus, they are potential pollution sources of soil trace metals. The study area is characterized by a subtropical monsoon climate, and the wind direction

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