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Three-dimensional spatial variability of arsenic-containing soil from geogenic source in Hong Kong: Implications on sampling strategies

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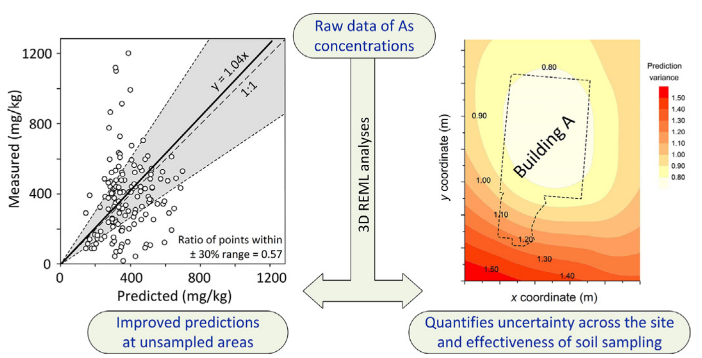
HIGHLIGHTS

- Spatial variations of geogenic As concentrations can be highly anisotropic.
- REML approach with 3D correlation structure is proposed to analyse such variations.
- Uncertainty at unsampled locations can be rationally quantified by the approach.
- The uncertainty estimates are useful indicators for site investigation/remediation.

GRAPHICAL ABSTRACT



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ABSTRACT

Soil contamination by trace elements such as arsenic (As) can pose considerable threats to human health, and need to be carefully identified through site investigation before the soil remediation and development works. However, due to the high costs of soil sampling and testing, decisions on risk management or mitigation strategies are often based on limited data at the site, with substantial uncertainty in the spatial distributions of potentially toxic elements. This study incorporates the restricted maximum likelihood method with three-dimensional spatial autocovariance structure, to investigate the spatial variability features of As-containing soils of geogenic origin. A recent case study in Hong Kong is presented, where >550 samples were retrieved and tested for distributions of As concentrations. The proposed approach is applied to characterize their spatial correlation patterns, to predict the As concentrations at unsampled locations, and to quantify the uncertainty of such estimates. The validity of the approach is illustrated by utilizing the multi-stage site investigation data, through which the advantages of the approach over traditional geostatistical methods are revealed and discussed. The new approach also quantifies the effectiveness of soil sampling on reduction of uncertainty levels across the site. This can become a useful indicator for risk management or mitigation strategies, as it is often necessary to balance between the available resources for soil sampling at the site and the needs for proper characterization of contaminant distributions.

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1. Introduction

Soil contamination by metals/metalloids poses an increasing threat to human health and environmental quality across the globe (Tóth et al., 2016; Xia et al., 2017; Plessl et al., 2017). Among the potentially toxic elements, arsenic (As) has received considerable attention over the last few decades due to its high toxicity and environmental risk (Bundschuh et al., 2013; Ehlert et al., 2016; Sandhi et al., 2017). Geochemical properties of As are complex because of its various chemical species and amphoteric nature (Yang et al., 2017; Wu et al., 2017), which makes it challenging for proper assessment and remediation (Rahman et al., 2017). Soil can naturally possess high concentrations of As due to weathering of the parent materials, volcanic eruptions, and forest fires (Beiyuan et al., 2017; Li et al., 2017). In recent years, industrialization and urbanization have also transferred As from used products into the environment, resulting in many industrial contaminated sites (Tsang et al., 2014; Gallego et al., 2016; Wcisło et al., 2016). Consequently, many studies have focused on urban contaminated soils with As from anthropogenic sources, such as agricultural activities and industrial and mining processes (Rieuwerts et al., 2014; Cao et al., 2016; Yoon et al., 2016; González-Fernández et al., 2018). Although geogenic As-containing soil/sediment is a common problem worldwide (Fendorf et al., 2010; Yang et al., 2014), including Hong Kong (Li et al., 2017; Cui et al., 2018), there has been limited discussion on the characterization of spatial variations of geogenic As and the corresponding implications on management/remediation of As-containing sites.

To develop cost-effective management and risk mitigation recommendations, the concentration distribution and variability features (or spatial correlation patterns) of the trace elements should be established foremost through identifying, mapping, and monitoring processes (Bednářová et al., 2016; Pan et al., 2017). It is because the remediation processes, especially for extraction-based approaches, are often expensive and highly dependent on the types of contaminants and estimated amounts of contaminated soils that need to be treated (Bolan et al., 2014; Tsang and Yip, 2014; Wan et al., 2016). In practice, however, it is difficult to accurately predict the concentrations of metals/metalloids due to their complex spatial distribution patterns, including the occasional occurrence of 'hotspots' with high levels of anthropogenic contamination or geogenic formation. Legislations in various countries (e.g., China, the United States, the Netherlands, Australia, and New Zealand) advocate the use of probabilistic sampling schemes (e.g., square grid, simple random, stratified random) for contaminated site assessments (Waterhouse, 1980; Bell et al., 1983; Gilbert, 1987; Horta et al., 2015), which is similarly applied in Hong Kong (HK EPD, 2011). However, these methods focused on detecting high concentration regions of anthropogenic contaminants and quantifying the extent of such hotspots. This is considered appropriate when there is prior knowledge about the contaminants involved, their transport mechanisms, and the human activities causing the contamination.

For trace elements of geogenic nature, the determination of sampling strategies and characterization of their spatial distributions may require different techniques because their existence are not caused by anthropogenic activities (Li et al., 2015; Cui et al., 2018). To this end, various methods such as Geostatistics, multivariate methods, and Geographic Information System (GIS) mapping have been applied to identify and reveal the distributions of these trace elements (Lark, 2000; Lark and Cullis, 2004; Santra et al., 2012; Antunes and Albuquerque, 2013; Hao et al., 2016; Chakraborty et al., 2017; Boente et al., 2017). Geostatistics has been developed for application in various disciplines, and is represented by techniques including various types of kriging (ordinary/disjunctive/indicator kriging), global/local polynomial interpolation (G/LPI), inverse distance weighting (IDW), nearest neighbour interpolation (NNI), radial basis functions (RBF), sequential Gaussian simulation (SGS), etc. Each of them involves different statistical assumptions. Despite the growing literature of these methods, there are major limitations associated with their application for site investigation in an urban setting. For instance, many development sites in densely-populated cities may span across hundreds of metres to tens of kilometres, within which a large number (i.e., hundreds to thousands) of samples may be needed to provide adequate precision for meaningful geostatistical analyses to aid the site development plans. Meanwhile, the concentrations of trace elements may display three-dimensional spatial variations across the subsurface soil domain, which should be properly accounted for in such analyses. Many previous studies (e.g., Santra et al., 2012; Chakraborty et al., 2012; Chakraborty et al., 2017; Zhang and Yang, 2017) discussed the accuracy of various geostatistical approaches through cross validation measures and indicators such as root-mean-square-error (RMSE) and mean percentage error (MPE). However, the uncertainty associated with As distribution across the site is rarely discussed in detail. While quantification of uncertainty is essential from the project management perspective, such estimates are often difficult to verify.

This study extends the integrated framework for spatial variability analyses from our recent studies (Liu et al., 2017; Liu and Leung, 2017), incorporating the restricted maximum likelihood (REML) method with a three-dimensional, anisotropic autocorrelation structure, tailored for analysing the concentrations of trace elements in soils. Effectiveness of the approach is illustrated by the implementation on a major development site in Hong Kong, where borehole sampling of As is performed in multiple stages. The current study articulates the spatial extent of the geogenic As, and proposes a rational approach to quantify the associated uncertainty, hence improving the effectiveness of geoenvironmental sampling strategy for site assessment and remediation.

2. Methodology

2.1. Soil sampling and analysis

A new development site located in the New Territories in Hong Kong is discussed in this study to illustrate the three-dimensional variations of As concentrations, and how the variability can be characterized by the proposed framework in this study. Two stages of geoenvironmental investigation were performed within this development area, with their key information summarized in Table 1. In order to identify potential contamination at the site, the Stage 1 investigation was performed (Fig. 1a) at an early stage of the project, which included drilling of 35 boreholes, with 388 soil samples retrieved from different depths for the testing of As concentrations (HK CEDD, 2015). The locations of boreholes had been strategically selected for broad coverage across the development area (approximately 1600 m × 2700 m on plan), considering both site accessibility and the locations of future structures. Within the development area, there was a smaller site (around 100 m × 200 m) of particular concern in the project. The site was where one of the first structures (Building A shown in Fig. 1) would be constructed, and the As concentrations at this location had to be assessed to formulate appropriate mitigation measures. However, during the Stage 1 investigation, no boreholes had been drilled within this site due to accessibility issues at that stage. Predictions were therefore made by the proposed approach (Section 2.2), utilizing all 388 sample values obtained from the Stage 1, and their corresponding spatial information.

Table 1
Site area and sampling information for Stages 1 and 2 investigations.

	Stage 1 (Fig. 1a)	Stage 2 (Fig. 1b)
Target area	Entire development area (1600 m × 2700 m)	Building A site (100 m × 200 m)
No. of boreholes	35	12
No. of samples	388	205
Average spacing of boreholes	870 m	57 m
Vertical sampling interval	0.5–2 m	0.5–2 m