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Assessment of terrain factors on the pattern and extent of soil contamination surrounding a chemical industry in Chongqing, Southwest China

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ABSTRACT

In order to determine the pattern and spatial distribution of polycyclic aromatic hydrocarbons (PAHs) in contaminated soil at a site of an organic chemical plant with irregular terrain, we developed a method of soil sampling by subarea that considers topographic features. We divided the site into the west subarea, which had the highest elevation; the southeast subarea (second-highest elevation), and the northeast subarea (lowest elevation). Two- and three-dimensional kriging and principal component analysis (PCA) models were employed to interpret the contamination pattern and spatial distribution of PAH contamination and to calculate the volume of soil contaminated with PAHs. The results show that the total concentrations of 16 priority PAHs in each borehole ranged from 0.7 to 4889.5 mg/kg, with an average of 130 mg/kg. The pollutants mainly accumulated in the first layer of the site (0–1 m). Compared with the pollution levels at other chemical plants reported previously, the soil PAH concentrations measured in this study were higher. PCA results show that the explained variations of the first and second components were 88% and 0.9%, respectively. The samples had good separation characteristics between the different subareas and correlation and pertinence within the same subarea are strong. Volumes of soil contaminated with benzo[*a*]pyrene calculated using four different interpolation models were 131,019, 135,365, 142,741, 146,783 m^3. Three-dimensional kriging that takes into consideration the surface-elevation change showed the lowest root-mean-square error and mean error, as well as higher accuracy compared with the other models. These results are important for defining remediation boundaries and the volume of contaminated soil for sites subjected to risk assessment and remediation.

1. Introduction

Because of rapid social and economic development across China, many large-scale industrial facilities have been moved out of cities, and land is being redeveloped into commercial or residential areas (Ding and Hua, 2012; Zhong et al., 2013). Unfortunately, many of these industrial sites are contaminated. Since the industry of organic chemicals has been an important part of the Chinese economy, many sites for organic chemical manufacture in cities across the country are contaminated. Contaminants most often encountered in organic chemical sites are polycyclic aromatic hydrocarbons (PAHs) (Jiao et al., 2009; Ma et al., 2013). PAH contamination not only causes severe environmental effects, but also represents a serious human health hazard (Wilcke, 2007; Cai et al., 2008).

Before the remediation of contaminated sites can be initiated, the spatial distribution of contaminants should be understood in detail in order to define the scope of pollution at such sites (Critto et al., 2003; Zhihuan et al., 2008). Spatial interpolation of data from limited soil samples is typically used to predict soil contaminant concentrations and to map the spatial distribution of pollutants across sites (Carlon et al., 2001; Goovaerts et al., 2008; Yenilmez et al., 2011; Liénard et al., 2014). However, low geographic densities of soil sampling data as well as high variability and skewed statistical distributions usually limit our understanding of the distribution of contaminants in soils (Wu et al., 2006).

Geostatistics can provide an unbiased estimate of variables at unmeasured locations. It has been widely used in describing the spatial distribution of pollutants in contaminated soils and in determining the area of the site that exceeded a critical threshold value (Meuli et al., 1998; Cattle et al., 2002; McGrath et al., 2004; Xie et al., 2011). Many improvements have been made in spatial interpolation models to improve the prediction accuracy and to reduce the uncertainty of

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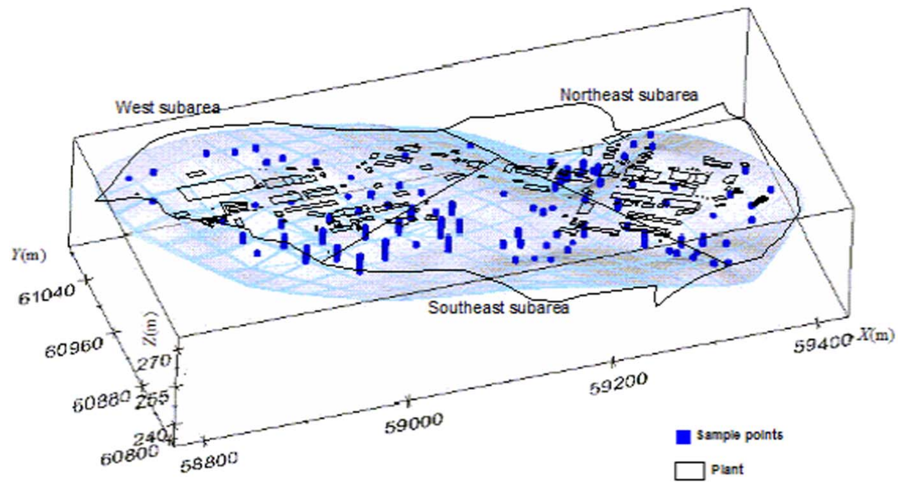

Fig. 1. Locations of sampling sites.

spatial interpolation results. For example, a geographically weighted regression procedure and a Bayesian maximum entropy model have been used to describe the variability characteristics of data (Christakos, 1990; Brunsdon et al., 1996). Other studies have considered normal transformations of data, as well as soil map units, geographical factors, sampling plans, and so forth, to improve prediction accuracy (Juang et al., 2008; Wu et al., 2008; Yuan et al., 2010; Dragović et al., 2014).

In the remediation of contaminated sites, accurate calculation of the volume of contaminated soil is essential, as this affects the rehabilitation costs and the choice of the remediation technology. The main objective of the previous study was to obtain a two-dimensional spatial distribution of contaminants and to determine the site subarea that exceeded the critical threshold value (10 mg/kg for Cd and 15 mg/kg for Pb; Juang and Lee, 1998). In site remediation fieldwork, we usually multiply the contaminated area that exceeds a critical threshold value by the depth of soil to obtain the volume of contaminated soil. Variation in pollutants in the three-dimensional stratum and topographic features are not considered in this method; thus, its results often fail to coincide with the field situation (Liu et al., 2014). Some researchers have used three-dimensional methods to describe the spatial distribution of pollutants or geological variables (Liu et al., 2002; Van Meirvenne et al., 2003; Perroy et al., 2014). However, few studies have been conducted on the volume of contaminated soil at industrial sites with irregular terrain.

Identifying the contamination pattern (including the congener variation and concentration) adds value to the site-specific risk assessment and to the conceptual model of the site in terms of understanding the distribution and sources of contamination (Henriksson et al., 2013). Multivariate statistical analysis, trend analysis, and spatial autocorrelation analysis are typically employed in analyzing data from contaminated sites with regular terrain and for determining the interrelation and pattern of contaminant concentrations in soil samples (Carlon et al., 2001; Critto et al., 2003; Zhang, 2006). The contaminant concentrations of soil samples in different subareas and strata belong to multivariate data sets; however, contamination pattern identification is always based on univariate categorical data. Nonetheless, multivariate data analysis such as principal component analysis (PCA) is generally suitable for its use in the interpretation of multivariate data sets to identify contamination in environmental samples (Colombo et al., 2011).

The study site was an organic chemical plant situated on mountainous terrain in Chongqing, an industrialized city and municipality in southwest China. The city is located in the southeastern Sichuan Basin. The plant is located on irregular terrain, and the contaminant distribution in the soil has strong spatial heterogeneity affected by the irregular terrain and the manufacturing process. Traditional methods of spatial interpolation and contamination pattern identification have typically not considered terrain characteristics, making it exceptionally difficult to obtain accurate calculations of the volume of contaminated soil and spatial patterns of contaminants in the irregular terrain. In this study, two- and three-dimensional kriging were compared to obtain the spatial distribution of pollutants and to calculate the volume of contaminated earth at the target site. Multivariate statistics was employed to identify the spatial patterns of polluted samples.

The objectives of this study were (1) to characterize the distribution of contaminants in soils, (2) to calculate the volume of soil contaminated with specific pollutants in the whole stratum, and (3) to interpret the contamination pattern of soil samples at the site.

2. Materials and methods

2.1. Site characterization

The study site was located at a former organic chemical plant on the banks of the Yangtze River within the city of Chongqing in southwest China. The plant was one of the most important chemical enterprises when it began to operate in the region. The area has a subtropical monsoon climate with an average annual temperature of 17.6 °C and a mean annual precipitation of approximately 1360 mm. The plant is located on irregular terrain, with the northwest area at 280 m in ground elevation and the southeast area at 230 m (Fig. 1). Phenol, benzene, methylbenzene, solvent oil, and unsaturated polyester resin were produced in this plant for > 50 years. The site has an area of 200,000 m^2 and contains a phenol workshop, resin workshop, sewage outfall, office area, and warehouse. The sources of PAHs at the site were exhaust emissions, wastewater, and solid-waste pollution produced during the overall manufacturing process. Production of refined benzene from crude benzene generated lower-boiling hydrocarbons and benzo[*a*]pyrene (Bap). Wastewater produced by separation of oily water also had these contaminants. Poor production technology and pollution controls during its operational period caused serious environmental pollution at the site, resulting in severe damage to the plant site and to the surrounding environment.

2.2. Sampling and chemical analyses

According to historical production, technological layout, and topographical features, we divided the site into three regions: the west subarea (highest elevation), the southeast subarea (second-highest elevation), and the northeast subarea (lowest elevation). Ninety-one soil sample locations within the site were included, and three soil samples were collected from each of the three layers in each borehole