Assessing the risk costs in delineating soil nickel contamination using sequential Gaussian simulation and transfer functions

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Abstract

Geostatistical simulated realization maps can represent the spatial heterogeneity of the studied spatial variable more realistically than the kriged optimal map because they overcome the smoothing effect of interpolation. The difference among realizations indicates spatial uncertainty. These realizations may serve as input data to transfer functions to further evaluate the resulting uncertainty in impacted dependent variables. In this study, sequential Gaussian simulation was used to simulate the spatial distribution of soil nickel (Ni) in the top soils of a 31 km² area within the urban-rural transition zone of Wuhan, China. Simulated realizations were then imported into transfer functions to calculate the health risk costs caused by Ni polluted areas ignored in remediation due to underestimation of the Ni contents and the remediation risk costs caused by unnecessary remediation of unpolluted areas due to overestimation of the Ni contents. The uncertainty about the input Ni content values thus propagated through these transfer functions, leading to uncertain responses in health risk costs and remediation risk costs. The spatial uncertainty of the two forms of risk costs were assessed based on the response realizations. Because the risk of exposure of soil Ni to humans and animals is generally greater in contaminated arable lands than in industrial and residential areas, the effect of land use types was also taken into account in risk cost estimation. Results showed that high health costs mainly appear in the southwest part of the study area, while high remediation costs mainly occur in the east, middle and northwest of the study area, and that most of the south part of the study area was delineated as contaminated according to the minimum expected cost standard. This study shows that sequential Gaussian simulation and transfer functions for assessing risk costs of soil contamination delineation and associated spatial uncertainty.

1. Introduction

Accumulation of heavy metals, such as lead and nickel, in soils may impact soil properties, reduce soil biological activity and hinder the effective supply of nutrients. More importantly, heavy metals can be strongly enriched through the food chain and other ways, directly or indirectly threatening human health. Therefore, effectively delineating contaminated areas of soil heavy metals is of great importance to environmental management such as remediation decision making. Geostatistical interpolation, or kriging, has been increasingly used to estimate the spatial distribution of trace elements in soils. However, the smoothing effect, commonly found in the maps generated by optimal interpolation, results in less variation in estimated values than in observed values. This problem causes low values to be overestimated and high values to be underestimated, thus impacting the delineation of polluted areas. However, simulated realization maps by geostatistical stochastic simulation algorithms can represent the spatial distribution more realistically than the kriged map, because they overcome the smoothing effect of kriging.

One effective method in delineating contaminated areas of soil heavy metals for remediation consists of evaluating the economic impact of the two possible decisions — remediation and non-remediation — using the concept of loss functions. Each location is classified as safe or contaminated so as to minimize the resulting expected loss. For example, Goovaerts et al. (1997) estimated the risk costs of contamination by cadmium, copper and lead in the topsoil of a 14.5 km² region in the Swiss Jura; Cattle et al. (2002) assessed the spatial distribution of lead contamination within an area of 2.3 km² in the inner Sydney suburb of Glebe; and Amini et al. (2005) mapped the risk of cadmium and lead contamination to human health in soils of Central Iran in a large study area of 6800 km². All of these studies combined loss functions and indicator kriging together, where indicator kriging was used to estimate the threshold exceeding probabilities and loss functions were used to calculate the expected loss. An alternative method is to combine a sequential simulation algorithm and transfer functions for evaluating the economic risk costs of remediation decision, because alternative simulation realizations generated by stochastic simulation may serve as an input to transfer functions. Such an idea was initially suggested in...
Nickel (Ni) is an essential element of lives. While in small quantities nickel is necessary, with high uptake it also can be harmful to human health (Nieboer and Nriagu, 1992). Studies found that uptakes of large quantities of nickel may cause various consequences on human bodies, such as inflammation, cancer, neurasthenia, system disorders, lower fertility, teratogenic, mutagenic, and heart disorders (Das et al., 2008; Kargacin et al., 1993; Nieboer and Nriagu, 1992). Humans may be exposed to nickel through food, drinking water, smoking, or polluted air (Nieboer and Nriagu, 1992). Therefore, soil nickel should be treated seriously in environmental management.

In this study, soil Ni concentration spatial distribution was simulated using sequential Gaussian simulation (SGS), and the simulated realizations were then used as input to transfer functions to compute the health risk costs caused by ignoring the areas with high soil Ni contents due to underestimation and the remediation risk costs caused by remediating the areas with low soil Ni contents due to overestimation. The response realizations were used for uncertainty assessment of the two forms of risk costs. Because land use types usually have apparent impacts on the risk of soil Ni exposure to humans and animals, we took land use types into account. The objectives of this study are to: (1) explore the Ni spatial distribution pattern in the study area; (2) explore the spatial distribution patterns of remediation risk costs and health risk costs caused by wrong decisions on pollution remediation using simulated realizations and transfer functions accounting for land use types; and (3) delineate contaminated areas using the standard of minimum expected costs.

2. Materials and methods

2.1. Study area and data

A study area of approximately 31 km² is chosen in the urban–rural transitional zone of Wuhan City, Hubei Province. The study area is an important vegetable production base and has a history of vegetable planting of 30 to 40 years. Most of the area is surrounded by iron and steel corporations and power plants. In this study, lands are divided into two types — arable lands, mainly used for vegetable production, and non-arable lands, mainly used for residence and industry (Fig. 1). The climate represents a typical subtropical humid monsoon with an average annual temperature of 15.9 °C and an average annual rainfall of 1300 mm.

An investigation of soils was performed in October, 2010. 124 non-rhizosphere topsoil samples (0–20 cm depth) were collected in the study area (Fig. 1). Sampling was carried out within the intervals of vegetation to reduce the impact of plant uptake. At each sampling point, four to six sub-samples were randomly taken and then mixed to obtain a composite soil sample. All samples were air-dried at room temperature (20–22 °C), crushed after stones and other debris were removed, and then sieved to the particle size of less than 2 mm. Portions of soil samples (about 50 g) were ground in an agate grinder and sieved through 0.149 mm mesh. The prepared soil samples were then stored in polyethylene bottles for measuring total concentrations of nickel (Ni). About 0.5 g of the prepared soil samples were digested in Teflon beakers with a mixture of nitric acid (HNO₃) and perchloric acid (HClO₄) using hot plane (Agricultural Chemistry Committee of China, 1983). Total concentrations of Ni in the digested solution were measured using inductively coupled plasma mass spectrometry (X7 ICP-MS, TMO, USA). For quality assurance and quality control, we analyzed duplicates, method blanks, and standard reference materials.

2.2. Geostatistical methods

Geostatistics provides the methods to predict values at unsampled locations from nearby data at sampled locations by taking into account the spatial correlation of sampled points. It can minimize the variance of estimation errors and investigation costs (Ferguson et al., 1998; Qu et al., 2012; Saito et al., 2005). The variogram — the spatial correlation measure for kriging — is an effective tool for evaluating spatial variability (Boyer et al., 1991; Cahn et al., 1994). A variogram may describe the spatial auto-correlation structure of a continuous variable and provide some insight into possible factors that affect data distribution (Webster and Oliver, 1990). Spatial patterns of soil attributes following the intrinsic stationarity assumption can be described using the following experimental variogram

\[
\hat{\gamma}(h) = \frac{1}{2N(h)} \sum_{i=1}^{N(h)} [Z(x_i) - Z(x_i + h)]^2
\]

where \(Z(x_i)\) is the measured value at location \(x_i\), \(h\) is the lag distance, and \(N(h)\) is the number of data pairs at the lag distance \(h\).
where $N(h)$ is the number of data pairs separated by distance $h$, and $z(x_i)$ and $z(x_i + h)$ are the measured values for regionalized variables $z(x)$ at the locations of $x_i$ and $x_i + h$, respectively.

Ordinary kriging (OK) was chosen as one of the methods to create the spatial distribution maps of soil Ni contents, with the maximum search radius being set to the auto-correlation range. Kriging is also known as Best Linear Unbiased Predictor (BLUP) and OK can be expressed as a linear-weighted average function of observations in the neighborhood of the unsampled location $x_0$:

$$\hat{y}(x_0) = \sum_{i=1}^{N} \lambda_i y(x_i)$$

where $\hat{y}(x_0)$ is the predicted value at location $x_0$, $y(x_i)$ is the measured value for soil property at position $x_i$, $\lambda_i$ is the weight of the corresponding measured value obtained from the ordinary kriging system, and $N$ is the number of measured samples within the neighborhood.

SGS is the most frequently used sequential simulation algorithm for simulating continuous variables. A detailed introduction on this method can be found in Goovaerts (1997) and Remy et al. (2009). After a regularly spaced grid covering the study area is defined, the SGS procedure involves the following steps:

1. Transform the sample data of Ni into standard normal data using the normal score transformation.
2. Compute and model the experimental variogram of the normal score transformed data.
3. Establish a random path through all of the grid nodes, in a way that each node is visited only once in each sequence.
4. At each node $x_i$:
   a. Estimate the parameters (mean and variance) of the Gaussian conditional cumulative distribution function (ccdf) of Ni by simple kriging estimator with the normal score variogram model. The conditioning data includes both the original data and already simulated data within the defined neighborhood of the location to be simulated.
   b. Draw a simulated normal score value from the estimated ccdf and then add it to the conditional data set to be used for simulating other nodes.
   c. Proceed to the next grid node along a random path and repeat steps (a) and (b) until the entire set of grid nodes are simulated.
   d. Back-transform the simulated normal score values into the content values of Ni in the original data space.

These sequential steps build up only the first realization, $\{z^{(1)}(x_0), j = 1, ..., N\}$, which is only one model of Ni spatial distribution. To generate multiple, say $K$ realizations, $\{z^{(k)}(x_0), j = 1, ..., N, k = 1, ..., K\}$, steps 3 to 5 should be repeated with different random paths passing through all nodes. In this study, geostatistical interpolation and simulation used a grid having a pixel resolution of 100 m $\times$ 100 m.

### 2.3 Transfer functions

When decision-makers delineate polluted areas where remedial measures should be taken, they have to face two types of misclassification risks. One is the underestimation of pollution in some polluted areas, which may result in these underestimated areas being ignored in remediation and consequently cause health problems leading to insurance claims and lawsuits. In this study we define this type of costs as health risk costs. The other is the overestimation of toxic pollutants in some unpolled (or low-polluted) area, which may result in unnecessary remediation of those overestimated areas leading to extra costs. In this study we define this type of costs as remediation risk costs. In reality, risk costs may be a complicated function of pollutant concentration $z(\mathbf{u})$ and other factors. Following Goovaerts (1997), we determined the risk costs associated with misclassifying a location $\mathbf{u}$ as safe or contaminated according to a specific threshold ($z_e$) as

$$L_1(z(\mathbf{u})) = \begin{cases} 0 & \text{if } z(\mathbf{u}) \leq z_e \\ \omega_1 & \text{if } z(\mathbf{u}) > z_e \end{cases}$$

and

$$L_2(z(\mathbf{u})) = \begin{cases} 0 & \text{if } z(\mathbf{u}) > z_e \\ \omega_2 & \text{otherwise} \end{cases}$$

respectively. In above Eq. (3), $\omega_1(z(\mathbf{u}))$ is the relative health risk cost of underestimating the pollutant concentration at location $\mathbf{u}$ with a unit of money per concentration (i.e., dollar/(mg kg$^{-1}$)) at here. Because the risks of exposure to humans and animals are different between contaminated arable lands and non-arable lands (i.e., industrial and residential areas in this study), the health risk cost $\omega_1(z(\mathbf{u}))$ is modeled as a function of the land use type prevailing at $\mathbf{u}$. In above Eq. (4), the remediation risk cost $\omega_2$ is modeled as a constant value with a unit of money only. This is because the remediation procedure amounts to removing the upper layer of soils and hence the risk cost is independent of the actual concentration.

In this study, we chose $z_e = 50$ (mg kg$^{-1}$) for the whole study area, $\omega_1(z(\mathbf{u})) = 10$ dollar per (mg kg$^{-1}$) for arable lands or 5 dollar per (mg kg$^{-1}$) for non-arable lands, and $\omega_2 = 100$ dollar for the whole study area. All of these $\omega$ values were based on an area of a single pixel (i.e., 100 m $\times$ 100 m). These parameters were determined approximately for the study area. One may choose them differently for different situations. For example, Goovaerts et al. (1997) chose 1 and 2.5 for $\omega_1$ and $\omega_2$, respectively, in estimating the risk costs of contamination by soil cadmium, copper and lead; Cattle et al. (2002) used values of 2.5 and 10 for $\omega_1$ and 1 for $\omega_2$ in assessing the spatial distribution of soil lead contamination; and Amini et al. (2005) took the values of 2.5 for $\omega_1$ and 1 for $\omega_2$ in mapping the risk of soil cadmium and lead contamination to human health. In reality, however, these parameters are not constants and may be complicated functions of concentration and other factors. The decision of a value to assign to either $\omega_1$ or $\omega_2$ was subjective in these studies, and also beyond the scope of this research.

### 2.4 Delimitation of contaminated areas

The criterion for classifying a location as contaminated is as follows: the expected cost associated with declaring a location contaminated should be smaller than the expected cost associated with classifying a location as safe. The expected cost associated with misclassifying a location as safe (health risk caused by pollution hazard to human health) and the expected cost associated with wrongly declaring a location contaminated (remediation risk caused by undue cleaning of a safe location) are approximated, respectively, as

$$\varphi_1(\mathbf{u}) = E[L_1(Z(\mathbf{u}))) \approx \frac{1}{K} \sum_{k=1}^{K} L_1(z^k(\mathbf{u}))$$

and

$$\varphi_2(\mathbf{u}) = E[L_2(Z(\mathbf{u}))) \approx \frac{1}{K} \sum_{k=1}^{K} L_2(z^k(\mathbf{u}))$$

where $z^k(\mathbf{u})$ is the $k$th simulation value at location $\mathbf{u}$. $K$ is the total number of simulated realizations. The location $\mathbf{u}$ is declared safe or

<table>
<thead>
<tr>
<th>Soil elements</th>
<th>Minimum</th>
<th>Maximum</th>
<th>Mean</th>
<th>S.D.</th>
<th>C.V. (%)</th>
<th>Background Ratio$^a$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ni (mg kg$^{-1}$)</td>
<td>26.46</td>
<td>90.58</td>
<td>52.87</td>
<td>13.20</td>
<td>24.97</td>
<td>37.3</td>
</tr>
</tbody>
</table>

$^a$ Natural background value in Hubei (Wei et al., 1990). $^b$ Ratio = Mean/Background value.
contaminated so as to minimize the resulting expected loss using the following rules:

\[ \phi_1(u) > \phi_2(u) \Rightarrow u \text{ is classified as contaminated;} \]

\[ \phi_1(u) < \phi_2(u) \Rightarrow u \text{ is classified as safe.} \]

In above Eqs. (5) and (6), we used SGS to generate a number of simulated realizations for approximately estimating the expected risk costs, rather than using indicator kriging as done in earlier studies (Amini et al., 2005; Cattle et al., 2002; Goovaerts et al., 1997). Such an idea was proposed in Goovaerts (1997, p. 373–375), but rarely used later, probably due to the computation load. Consequently, in this study Eqs. (3) and (4) were called transfer functions, rather than loss functions, as suggested by Goovaerts (1997). In addition, the effect of land use types was also incorporated.

3. Results and discussion

3.1. Statistics of soil Ni sample data

Descriptive statistics and the histogram of soil Ni concentration data are provided in Table 1 and Fig. 2, respectively. One can see that the mean concentration of soil Ni is higher than its background value in Hubei Province (Wei et al., 1990), which means Ni is enriched in the top soils. Our data show that the concentrations of Ni in some samples exceed the level of second-grade pollution based on Chinese Environmental Quality Standard for Soils (State Environmental Protection Administration of China, 1995), and thus exhibit a pollution trend. Therefore, monitoring this heavy metal element is necessary in the study area, and remediation measures should be taken to prevent its harm to the environment and human health.

Because no apparent anisotropy was found for Ni data, experimental variograms were estimated omni-directly. Experimental variograms with parameters based on the original Ni data and the corresponding normal score transformed data are shown in Fig. 3. Both the experimental variogram of the original Ni data (for use in OK) and that of the normal score transformed Ni data (for use in SGS) are well fitted with exponential models. The nugget-to-sill ratio \( C_0/(C_0 + C) \) is usually used as a criterion to define the spatial auto dependence of a variable. Ratio values lower than 25% and higher than 75% correspond to strong and weak spatial dependencies, respectively, while ratio values between 25% and 75% indicate moderate spatial dependence. Generally, strong spatial dependence of soil properties may be attributed to intrinsic factors and weak spatial dependence may be attributed to extrinsic factors (Cambardella et al., 1994). The nugget-to-sill ratio \( C_0/(C_0 + C) \) of the variogram model for original Ni data is 17%, exhibiting strong spatial auto dependency which may be attributed to intrinsic factors such as other soil properties.

3.2. Spatial distribution of soil Ni concentration

Five hundred realizations of soil Ni spatial distribution were generated using SGS. Two randomly selected realizations are displayed in Fig. 4a and b. Each realization represents a possible spatial distribution of Ni without the smoothing effect. The E-type estimate from SGS simulated realizations maps (Fig. 4c) is, however, similar (but not the same) to the OK interpolation map (Fig. 4d). This is expected because the E-type estimate is a point-by-point average map of a number of
simulated realizations and such a map should converge approximately to a kriged map when the number of realizations is large (Chilès and Delfiner, 1999). All the four maps, especially the SGS E-type and OK estimates show similar trends, with high Ni values appearing in the south part and low values mainly occurring in the middle part of the study area. Among the three kinds of distribution maps of soil Ni, the SGS E-type and OK estimates are obviously smoother than the two randomly selected realizations. This smoothing effect is a known characteristic of kriging interpolation that may cause low values to be overestimated and high values to be underestimated (Lark and Webster, 2006).

The SGS conditional variance map and the OK variance map for Ni are presented in Fig. 5. The predictive variance map provided by SGS showed more certainty in the middle part of the study area and less certainty in the bottom part than that generated by OK. The OK variance varies much less across the study area (Fig. 5b). The smallest predictive variance, which reflects the smallest uncertainty, can be seen at sampling locations and nearby, whereas the largest uncertainty belongs to areas without samples and boundary areas. In Fig. 5a, the SGS conditional variance map shows that higher uncertainty appears mostly in high-valued areas of Ni contents, for example, the south part of the study area (see Figs. 4c and 5a, bottom areas). These areas are usually considered to need additional sampling. These results mean that the drawback of kriging variance being independent of actual sample values is overcome or mitigated by simulation algorithms.

3.3. Risk costs of delineating soil Ni contamination

For a given variogram and search rule for neighboring data, OK could obtain only one estimate, while stochastic simulation, such as SGS, could obtain many simulated realizations. With these realizations, the uncertainty of estimates could be quantified. Therefore, conditional simulation...
is clearly superior to interpolation, in dealing with the complexity and uncertainty of the problems which we face. These spatial simulations and uncertainty assessment could be further used in the simulation of risk costs in delineating soil Ni contamination. Health risk costs and remediation risk costs corresponding to the 20th and 80th simulated realizations in the spatial distribution of Ni concentration are presented in Fig. 6a–d, respectively. It could be seen that the location where there is a great health risk cost always has a small remediation risk cost, and vice versa. High value areas for health risk costs have a similar spatial distribution pattern to high Ni content areas, but there are some differences between the SGS E-type estimate of Ni and the mean estimate of health risk costs (see Figs. 4d and 6e). As different land use types were taken into account in estimating health risk costs through Eq. (3) which assigns less costs to non-arable lands compared with arable lands, health risk costs of non-arable lands was sharply reduced compared with that of arable lands, resulting in a similar pattern for low health risk costs and non-arable lands (see Figs. 1 and 6e).

Predictive variance maps corresponding to health risk costs and remediation risk costs are presented in Fig. 7. It can be seen that these two predictive variance maps have different spatial distribution patterns, caused by the different spatial patterns of corresponding risk costs (see Fig. 6). In the predictive variance map of remediation risk costs, the locations where the contents of Ni approximate to the chosen threshold value (i.e., 50 mg kg$^{-1}$) have greatest uncertainty (Fig. 7b) due to the binary partition function of Eq. (4) used for counting remediation costs.

### 3.4. Delineating soil Ni contamination

The classification map of locations regarded as contaminated by Ni, on the basis that the expected remediation risk cost (unnecessary cleaning) is smaller than the health risk cost associated with misclassifying a location as safe, is presented in Fig. 8. It is found that most of the south part...
of the study area was delineated as contaminated according to the minimum expected risk cost standard.

4. Conclusion

Sequential Gaussian simulation (SGS) and transfer functions were used to assess the remediation risk costs and health risk costs of soil Ni contamination in the top soils of a 31 km² area within the urban-rural transition zone of Wuhan, China. The spatial uncertainties of these two kinds of risk costs were also analyzed. Results showed that high health risk costs mainly appear in the southwest part of the study area, while high remediation risk costs mainly occur in the east, middle, and northwest parts of the study area. Most of the south part of the study area was delineated as contaminated according to the minimum expected cost standard. As land use type data were taken into account in estimating health risk costs through transfer functions, health risk costs in non-arable lands are reduced compared with that in arable lands because non-arable lands have lower health risk, thus resulting in similar patterns for the areas with little health risk costs and the areas of non-arable lands. Predictive variance maps for health risk costs and remediation risk costs show different spatial distribution patterns, due to the different spatial patterns of corresponding risk costs.

Geostatistical simulated realization maps can represent the spatial heterogeneity of soil Ni concentration more realistically than the kriged optimal map because simulated realizations do not have the smoothing effect of interpolation. Simulated realizations not only can indicate the spatial uncertainty of soil Ni through the differences among them, but also can serve as input data to transfer functions for further evaluating the result uncertainty in impacted dependent variables—the health risk cost and the remediation risk cost. This study proves that SGS and transfer functions are valuable tools for assessing risk costs of soil contamination delineation and associated spatial uncertainty.

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References


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