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Simulated annealing and kriging method for identifying the spatial patterns and variability of soil heavy metal

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**SIMULATED ANNEALING AND KRIGING METHOD
FOR IDENTIFYING THE SPATIAL PATTERNS AND
VARIABILITY OF SOIL HEAVY METAL**

Key Words: Simulated annealing, soil heavy metal, spatial pattern and variability, Geographic Information Systems

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ABSTRACT

Environmental data, including information regarding soil heavy metals, may contain significant uncertainty and exhibit a skewed distribution with a complex and unexplainable spatial variation. This study identified the spatial patterns and variations of soil heavy metals (Cu, Cd, Hg, and Pb) in the northern part of Changhua County in Taiwan to clarify the characteristics and pollution of soil heavy metals. The spatial maps of soil heavy metals were also estimated and simulated using kriging and simulated annealing methods. Correlation analysis indicated that Pearson and Spearman correlation coefficients among these four heavy metals were significant; between Cd and Cu, these two correlation coefficients were strongly significant. As expected, the spatial maps of estimation and simulation of soil heavy metals revealed high concentration areas

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of Cd, Cu along the main irrigation-ditch system and surrounding industrial plants in the area of study. In addition to reproducing the spatial variation of the investigated Cd, Cu, Hg and Pb, simulated annealing could also identify the global spatial continuity and discontinuity patterns of soil heavy metals. Kriging and simulated annealing methods can both be applied to identify pollution sources and patterns for monitoring and remedy.

INTRODUCTION

Soil pollution data occasionally display a complicated spatial variation (uncertainty) and a widely skewed distribution over the investigated site. Furthermore, the spatial patterns reveal that these environmental data occasionally display a small-scale discontinuity or heterogeneity formation. Therefore, accurately identifying the spatial characteristics of pollutants ensures the effectiveness of risk assessment and remedy. Kriging has recently been applied to analyze the spatial variability of pollutants. Notable examples include Keck et al. (1993), Samra and Gill (1993), Litaor (1995), Steiger et al. (1996), Couto et al. (1997), Juang and Lee (1998), Wang (1998), Wang and Zhang (1999), White et al. (1999) and Chang et al. (1999). Kriging, a linear interpolation procedure, provides a best linear unbiased estimator (BLUE) for quantities that vary in space. The procedure then, supports estimates at unsampled locations. The estimated values based on kriging display less variation than the investigated values, implying that kriging estimates may fail to preserve the high spatial variability and widely skewed distribution evident from investigation. However, generating many realizations that closely match the same sample statistics and conditioning data provides a visual and quantitative measure of spatial uncertainty (Goovaerts, 1996).

Conditional simulation, such as by simulated annealing, attempts not only to generate a set of values with some specified mean and covariance, but also to reproduce observed data at several locations. For instance, measurements of the

investigation can be used, along with simulated values, to analyze the spatial distribution of the variable in question. Varljen and Shafer (1991) established a conditional simulation to determine a cap by simulating transmissivity. Meanwhile, Rouhani et al. (1995) used geostatistical estimation and simulation to assess in-situ and engineering properties. Furthermore, Eggleston et al. (1996) used geostatistical simulations and ordinary kriging to reproduce the hydraulic conductivity structure in natural log formation and the sensitivity of this structure to limited data. Later, Mowrer (1997) used Sequential Gaussian Simulation to create maps of potential old-growth forest conditions across a 121 hectare first-order subalpine watershed. Also, Sterk and Stein (1997) applied a simulated annealing method to create multiple realizations of wind-blow mass transport, to overcome the smoothing effect of kriging estimation. Similarly to Mowrer, Kentwell et al. (1999) used a Sequential Gaussian fractal simulation to enhance the prediction accuracy of the grade tonnage curve. Finally, Wang and Zhang (1999) used a turning band method to reproduce multiple realizations for heavy metals.

This study performed estimation by ordinary kriging and simulation by simulated annealing techniques to produce maps and realizations of soil heavy metals in a real case study. The descriptive statistics, spatial structure (experimental variogram), and spatial patterns of simulation results were also discussed and compared. Moreover, the spatial maps of soil heavy metals in geographic information systems (GIS) were presented to elucidate the spatial patterns of these soil heavy metals.

MATERIALS AND METHODS

Data

The study area was a rice paddy field located in the north of Changhua County, Taiwan, as illustrated in Fig. 1. The two main irrigation ditches included first, a curve from the southeast to the northwest of the study area, and second, a straight line in the northeast of the study area. Industrial plants

FIGURE 1
Study area.

surrounded both of these main irrigation-ditch systems. Furthermore, the wastewater from industrial plants might be discharged into irrigation ditches and applied for agricultural purposes.

The Environmental Protection Administration (EPA) of the Republic of China initiated a collaborative research program in 1983 to measure As, Cd, Cu, Cr, Hg, Ni, Pb and Zn trace elements in the soil. This program also examined other soil properties such as cation-exchange capacity, and pH value. In these

FIGURE 2
Sampling points of (a) Cd, (b) Cu.

studies, soils were sampled from 878 sites representing important agricultural production areas across Taiwan.

This investigation obtained data from the EPA studies described above. This study used 194 sampling points (Figs. 2 and 3) to simulate the spatial distribution of soil heavy metals (Cu, Cd, Hg, and Pb). The soil heavy metals were measured between 1983 and 1986. Samples were taken from sites at target

FIGURE 3
Sampling points of (a) Hg, (b) Pb.

intervals of 100 m and 50 m in a network formation. Topsoil was sampled at depths of 0-15 cm.

Figures 2 and 3 display the spatial maps of the measured values for these heavy metals. Meanwhile, Fig. 4 displays the histograms of the measured soil heavy metal data, and Table 1 summarizes the statistics of the investigated data. Mean Cd, Cu, Hg and Pb concentrations for the complete set of data were 0.743(mg/kg), 54.46(mg/kg), 0.551(mg/kg) and 14.826(mg/kg), respectively.

FIGURE 4
Histogram of measured (a) Cd, (b) Cu, (c) Hg, (d) Pb.

TABLE 1
Descriptive Statistics of Investigated Data of Soil Heavy Metals

	Cd	Cu	Hg	Pb
Mean (mg/kg)	0.743	54.455	0.551	14.826
Median (mg/kg)	0.570	32.845	0.470	13.685
Variance ((mg/kg) ²)	0.331	4502.211	0.1176	16.960
Minimum (mg/kg)	0.120	11.370	0.160	8.100
Maximum (mg/kg)	3.600	556.000	2.100	31.020
25 th Percentile (mg/kg)	0.380	23.000	0.350	12.000
75 th percentile (mg/kg)	0.878	61.043	0.613	17.000
Skewness	2.032	4.387	2.572	1.121

The variance of these soil heavy metals were 0.331((mg/kg)²), 4502.121((mg/kg)²), 0.1176((mg/kg)²) and 16.960((mg/kg)²). According to these figures and Table 1, the Cd, Cu, Hg and Pb data exhibited skewed distributions.

Ordinary Kriging

Geostatistical techniques such as kriging, incorporate the spatial or temporal characteristics of actual data into statistical estimation. Geostatistics provide a variogram of data within a statistical framework, including spatial and temporal covariance functions. As expected, these variogram models are referred to as spatial or temporal structures, and are defined in terms of the correlation between any two points separated by either spatial or temporal distances.

Ordinary kriging, as applied within moving data neighborhoods, is a non-stationary algorithm that corresponds to a non-stationary random function model with varying mean but stationary covariance (Deutsch and Journel, 1992). Kriging estimates are calculated as weighted sums of the adjacent sampled concentrations. These weights depend on the exhibited correlation structure.

For illustration, if data appear highly continuous in space, the points closer to the estimated points receive higher weights than those farther away. The criterion for selecting these weights is minimization of the estimation variance. In this framework, kriging estimates can be regarded as the most accurate among all linear estimators (i.e., Best Linear Unbiased Estimator). Thus, at an unsampled location and for a given variogram, a kriging estimate can be thought of simply as an optimally weighted average of the surrounding sampled data (Cressie, 1990).

Simulation by Simulated Annealing

The annealing algorithm requires the image to be perturbed by simulating thermal perturbation (Deutsch and Cockerham, 1994). However, simulated annealing is an optimization technique for generating an initial field by drawing random values from a given histogram. Swapping the values in pairs of grid nodes not involving a conditioning datum sequentially modifies this initial field. A swap is allowed if the objective function is lower (Deutsch and Journel, 1992). The objective function is defined as an average squared difference between the experimental and given variograms.

A temperature function (the Boltzman distribution) in simulated annealing procedure controls the reduction speed of the optimization function by allowing some switches that increase the optimization function (Deutsch and Journel, 1992; Eggleston et al., 1996). The parameter, t , of the temperature function is called the temperature in the annealing procedure. A higher temperature increases the probability that an unfavorable swap will be accepted (Deutsch and Journel, 1992).

This study performed statistical analysis and correlation of concentrations of the four heavy metals using SPSS (Norusis, 1996). The variogram models of heavy metals were also fitted within GS+ (Gamma Design, 1995). Meanwhile, OKB2DM and SASIM in GSLIB conducted ordinary kriging and simulated annealing for heavy metals. The simulations and estimates were performed in a square thirty eight column by a twenty eight row grid consisting of 1064 50m by 50m cells. Three simulations from simulated annealing were performed in these

TABLE 2
Pearson Correlation Coefficient Matrix for Soil Heavy Metals

		Cd	Cu	Hg	Pb
Cd	Pearson Coefficient	1.000	0.611**	0.170*	0.282**
	Significant (2-tailed)		0.000	0.018	0.000
Cu	Pearson Coefficient	0.611**	1.000	0.178**	0.293**
	Significant (2-tailed)	0.000		0.009	0.000
Hg	Pearson Coefficient	0.170*	0.178**	1.000	0.167*
	Significant (2-tailed)	0.018	0.009		0.020
Pb	Pearson Coefficient	0.282**	0.293**	0.167*	1.000
	Significant (2-tailed)	0.000	0.000	0.020	

** : correlation is significant at the 0.01 level.

* : correlation is significant at the 0.05 level.

1064 cells. The results were transferred into Arcview 3.0 (ESRI, 1996) for display, to identify the spatial patterns of soil heavy metals.

RESULTS AND DISCUSSION

Correlation of Soil Heavy Metals

A nonparametric measure of association between two variables requires no assumptions regarding the normality of the distribution of variables (Walpole and Myers, 1985). However, a nonparametric measure, such as the Spearman correlation coefficient, is more reliable than a conventional measure, when data shows a distinct curvilinear relationship (Walpole and Myers, 1985). Therefore, this work calculated both Pearson and Spearman correlation coefficients for 194 pairs to identify the interrelationships of the four soil heavy metals.

Correlation analysis revealed that significant Pearson correlation coefficients among the four heavy metals. Meanwhile, Table 2 lists those coefficients. The correlation coefficient between the Cd and Cu measured data was 0.611, revealing a strong linear correlation, as confirmed in Table 2. These two heavy metals apparently shared a common origin and possibly similar spatial patterns. These results also implied that Cd and Cu might originate in the same pollution sources

TABLE 3
Spearman Correlation Coefficient Matrix for Soil Heavy Metals

		Cd	Cu	Hg	Pb
Cd	Spearman Coefficient	1.000	0.743**	0.144*	0.150*
	Significant (2-tailed)		0.000	0.046	0.037
Cu	Spearman Coefficient	0.743**	1.000	0.129	0.163*
	Significant (2-tailed)	0.000		0.073	0.023
Hg	Spearman Coefficient	0.144*	0.129	1.000	0.302**
	Significant (2-tailed)	0.046	0.073		0.000
Pb	Spearman Coefficient	0.150*	0.163*	0.302**	1.000
	Significant (2-tailed)	0.037	0.023	0.000	

** : correlation is significant at the 0.01 level.

* : correlation is significant at the 0.05 level.

as wastewater from industrial plants in the study area. Moreover, according to the 2-tailed test, the correlation coefficients of Cd/Cu, Cd/Pb, Cu/Hg, and Cu/Pb were significant at the 0.01 probability level. At the 0.05 level, using the 2-tailed test, the correlation coefficients of Cd/Hg and Hg/Pb were also significant. All of these four heavy metals might originate in the same irrigation-ditch systems.

Table 3 lists Spearman correlation coefficients among the four heavy metals in which the correlation between Cd and Cu revealed a significant coefficient (0.743) at the 0.01 level. Meanwhile, the correlation between Hg and Pb was significant at the 0.01 level, and the correlation coefficients of Cd/Hg, Cd/Pb and Cu/Pb were significant at the 0.05 level. The nonparametric correlation results indicated that Cd and Cu might both exhibit similar spatial patterns. The spatial patterns of Hg and Pb might resemble each other as well.

Variography

Experimental variograms were calculated for the soil heavy metals at various active lags and lag intervals. Least squares model fitting of these variograms generated a relatively consistent set of best-fit models with the lowest RSS (Model Reduced Sum of Squares) and highest r^2 values. Table 4 lists the best-fit models for Cd, Cu, Hg and Pb.

TABLE 4
Variogram Model of Cd, Cu, Hg and Pb Data

Heavy Metal	Model	Nugget Effect (mg/kg) ²	Sill (mg/kg) ²	Range (m)	Nugget/ (Nugget + Sill)
Cd	Exponential	0.174	0.175	1044	0.499
Cu	Exponential	2960.0	2340.0	714	0.559
Hg	Spherical	0.053	0.058	613	0.478
Pb	Exponential	6.300	12.200	850	0.351

For Cd, the investigated values revealed an exponential model with nugget effect=0.174(mg/kg)², sill=0.175(mg/kg)² and range=1044m, provided the best fit of all models (Spherical, or Gaussian) available in the software. The best-fit variogram of measured values for Cu was an exponential model with nugget effect=2960 (mg/kg)², sill=2340(mg/kg)² and range=714m. The variogram of Hg was a spherical model with nugget effect=0.053 (mg/kg)², sill=0.058 (mg/kg)² and range=613m. Meanwhile, the best-fit model for Pb data was an exponential model with nugget effect=6.300(mg/kg)², sill=12.200(mg/kg)² and range=850m. For distances below 613 to 1044 m, all investigated soil heavy metals exhibited a spatial structure.

The variography results indicated that the variogram models of the soil heavy metals with a high nugget effect illustrate a high small-scale variation or measurement error of the investigated data, over the study area. Table 4 lists the ratios ($R_c = \text{nugget effect} / (\text{Nugget effect} + \text{Sill})$) of Cd, Cu, Hg and Pb. The ratios of Cd, Cu and Hg were close to each other, also indicating high small-scale variations in Cd, Cu and Hg data. However, these variations might be attributed to wastewater from local industrial plants.

Statistics concerning Estimation and Simulation

The ordinary kriging estimates and simulations were based on the above variogram models and 194 observations of soil heavy metals. Tables 5, 6, 7 and 8 summarize the descriptive statistics related to ordinary kriging and simulated annealing simulation results. Tables 5 and 6 present the statistics on kriging

TABLE 5
Descriptive Statistics of Kriging estimates and Simulations of Cd

Statistics	Kriging	Simulation1	Simulation2	Simulation3
Mean (mg/kg)	0.770	0.741	0.750	0.748
Median (mg/kg)	0.729	0.570	0.570	0.570
Variance ((mg/kg) ²)	0.084	0.314	0.338	0.352
Minimum (mg/kg)	0.193	0.120	0.120	0.120
Maximum (mg/kg)	1.926	3.60	3.60	3.60
25 th Percentile (mg/kg)	0.575	0.380	0.380	0.380
75 th percentile (mg/kg)	0.874	0.894	0.900	0.870
Skewness	1.028	2.038	1.885	2.045

TABLE 6
Descriptive Statistics of Kriging Estimates and Simulations of Cu

Statistics	Kriging	Simulation1	Simulation2	Simulation3
Mean (mg/kg)	56.731	53.700	55.836	54.245
Median (mg/kg)	48.318	32.964	32.560	32.690
Variance ((mg/kg) ²)	858.167	4233.320	5214.420	4223.182
Minimum (mg/kg)	17.329	11.370	11.370	11.370
Maximum (mg/kg)	167.881	556.000	556.000	556.000
25 th Percentile (mg/kg)	37.877	23.000	23.000	23.000
75 th percentile (mg/kg)	66.149	61.136	61.624	63.788
Skewness	1.498	4.484	4.353	4.343

TABLE 7
Descriptive Statistics of Kriging Estimates and Simulations of Hg

Statistics	Kriging	Simulation1	Simulation2	Simulation3
Mean (mg/kg)	0.559	0.544	0.535	0.550
Median (mg/kg)	0.526	0.466	0.460	0.468
Variance ((mg/kg) ²)	0.034	0.107	0.101	0.111
Minimum (mg/kg)	0.276	0.160	0.160	0.160
Maximum (mg/kg)	1.400	2.100	2.100	2.100
25 th Percentile (mg/kg)	0.444	0.350	0.340	0.350
75 th percentile (mg/kg)	0.617	0.610	0.607	0.610
Skewness	1.934	2.530	2.548	2.508

estimates and simulations of Cd and Cu. Their comparison revealed that simulated annealing could reproduce the statistics of Cd and Cu better than usual, including the mean values listed in Tables 1, 5 and 6. Tables 7 and 8 list statistics related to kriging estimates and simulations of Hg and Pb. The simulation method also showed a better fit with the Hg data, as displayed in Table 7. Table 8 reveals that simulation could produce the empirical data related to investigated Pb, except the mean value better than kriging could.

The median values of the Cd, Cu, Hg and Pb simulations were almost identical to those of the measured heavy metal data as listed in Tables 1, 5, 6, 7 and 8. Simulation results also indicated that the 25th percentiles of the Cd, Cu and Hg simulations were identical to those of the investigated Cd, Cu and Hg data. Meanwhile, the 25th and 75th percentiles of the simulations were identical to those of the empirical Pb data.

The mean values of ordinary kriging estimated values over the area of investigation were close to the mean empirical values of Cd, Cu, Hg and Pb,

TABLE 8
Descriptive Statistics of Kriging Estimates and Simulations of Pb

Statistics	Kriging	Simulation1	Simulation2	Simulation3
Mean (mg/kg)	14.814	14.770	14.886	14.847
Median (mg/kg)	14.300	13.693	13.761	13.669
Variance ((mg/kg) ²)	7.065	15.357	16.315	16.991
Minimum (mg/kg)	10.958	8.10	8.10	8.10
Maximum (mg/kg)	23.172	31.020	31.020	31.020
25 th Percentile (mg/kg)	12.553	12.000	12.000	12.000
75 th percentile (mg/kg)	16.623	17.000	17.000	17.000
Skewness	0.734	1.026	1.009	1.020

particularly Pb. The median, variance, kurtosis, and skewness of simulations were extremely close to those of the investigated soil metals data, as indicated in Tables 5, 6, 7 and 8. Above results demonstrate that the ordinary kriging process might not preserve the variability and skewness of the investigated process. Moreover, the minimization of the prediction error variance involved smoothing the actual variability. Simulated annealing could reproduce the statistics for empirical data.

Figures 5, 6, 7, and 8 show the histograms of estimates and simulations. According to these figures, the simulation method closely matched the right and left hand tails of the distribution for heavy metals containing the highest and lowest concentration values. However, simulated annealing better reproduced the distribution of the soil heavy metals than kriging. These figures also indicated that the kriging producing smoothed data could not capture the distributions or spatial changes of the measured Cd, Cu, Hg and Pb data.

FIGURE 5

Histogram of Cd of (a) kriging estimates, (b) simulation 1, (c) simulation 2, (d) simulation 3.

FIGURE 6
Histogram of Cu of (a) kriging estimates, (b) simulation 1, (c) simulation 2, (d) simulation 3

FIGURE 7

Histogram of Hg of (a) kriging estimates, (b) simulation 1, (c) simulation 2, (d) simulation 3

FIGURE 8
Histogram of Pb of (a) kriging estimates, (b) simulation 1, (c) simulation 2, (d) simulation 3

FIGURE 9
Cd spatial map of (a) kriging estimates, (b) simulation 2, (c) simulation 3

Spatial Pattern of Heavy Metals

Ordinary kriging estimates and simulated annealing simulation were mapped using Arcview 3.0a, as indicated in Figs. 9, 10, 11, and 12. The kriged maps displayed the spatial pattern of the four heavy metals. Figure 9(a) illustrates two

FIGURE 10
Cu spatial map of (a) kriging estimates, (b) simulation 2 (c) simulation 3

high concentration areas along the main irrigated ditch, with industrial plants running from southeast to northwest and east to central parts of the study area. The kriged map of Cu showed a similar spatial pattern to that in the map of Cd. Above results improved the correlation analysis and also identified the pollution sources of heavy metals. The concentration of the eastern part in the kriged Cu

FIGURE 11
Hg spatial map of (a) kriging estimates, (b) simulation 2, (c) simulation 3

map was higher than the southwestern part, as shown in Fig. 10(a). The high concentration areas of the Hg kriged map surrounded industrial plants located in the east and northwest of the study area, as shown Fig. 11(a). Figure 12(a) presents the kriged map of Pb, in which the high concentration area was in the

FIGURE 12

Pb spatial map of (a) kriging estimates, (b) simulation 2, (c) Simulation 3

southeast. However, according to Figs. 9(a), 10(a), 11(a), and 12(a), maps of ordinary kriging estimates revealed that kriging tends to smooth out extreme values of the empirical data set. Despite indicating the spatial patterns of these soil heavy metals, kriging results might overestimate high and low concentration

areas of soil heavy metals, as well as underestimate extremely high and low concentration areas of soil heavy metals as indicated in Figs. 2, 3, 9(a), 10(a), 11(a) and 12(a).

Figures 9(b), 9(c), 10(b), 10(c), 11(b), 11(c), 12(b) and 12(c) display the simulation maps of Cd, Cu, Hg and Pb contents. The simulated Cd maps also indicated that the groups of industrial plants (Fig. 1) surround the high concentration areas displayed in Figs. 9(b) and 9(c). The spatial continuity and discontinuity patterns of the simulated Cu maps resembled those of the simulated Cu maps presented in Figs. 10(b) and 10(c). These two maps also revealed continuity and discontinuity patterns that might be caused by the irrigation ditches and random distribution of the industrial plants. The simulation maps of Hg (Figs. 11(b) and 11(c)) identified the continuity area with high Hg content in the northwest. Figures 12(b) and 12(c) show a high continuity area with a high Pb concentration in the southeast. Above simulated maps emphasized the strong variation across short distance and provide a measure of spatial uncertainty.

Kriging provides an optimal estimate of soil heavy metals at unsampled sites, thus illustrating the spatial patterns of soil heavy metals. However, it does not reproduce the statistics and spatial variability for the measured data collected in this study. Simulated annealing can not only reproduce the spatial variation exhibited by the investigated data, but can also identify the global spatial continuity and discontinuity patterns of soil heavy metals. The simulation maps of soil heavy metals illustrated that the large-scale and small-scale continuity patterns produced by simulation appear to be similar to those seen in the map of empirical data. The high concentration sites in all simulations might have neighboring points with low concentrations. These maps also confirmed that kriging estimates are much smoother than the simulations.

CONCLUSION

This study has demonstrated that Pearson and Spearman correlation coefficients among Cd, Cu, Hg and Pb heavy metals were significant. In

addition, between Cd and Cu, these two correlation coefficients were extremely significant. The variography results illustrated high small-scale variations in measured data concerning Cd, Cu, Hg and Pb over the area of interest. The spatial maps of estimates and simulation of soil heavy metals confirmed expectations that the high concentration areas of Cd and Cu were located along the main irrigation-ditch system and around the groups of industrial plants. In addition to reproducing the spatial variations shown in the measured Cd, Cu, Hg and Pb data, simulated annealing could also identify the global spatial continuity and discontinuity patterns of soil heavy metals. Kriging and simulated annealing methods can be applied to identify pollution sources and patterns for political purposes and remediation.

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