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MAPPING SOIL MERCURY IN TAIWAN USING GEOSTATISTICS AND GEOGRAPHIC INFORMATION SYSTEMS

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ABSTRACT

A map illustrating the total Hg content in soils of Taiwan was developed using the geostatistics and geographic information systems. Data were collected from Environmental Protection Administration's study targeting at agricultural soils in the 1980s. The range and arithmetic mean of the Hg contents in the topsoil (0 to 15 cm) of the study samples were as follows:0.01 to 2.61, 0.16 (mg/kg dry soil). An exponential variogram model was fitted using least squares and used to krige a grid covering Taiwan. This study emphasized that the map delineated gross trends in total soil Hg on the scale of conterminous Taiwan. Small-scale regional or localized spatial variability might be inadequately represented. The results showed that Soils in Toufen, Changhua and Pingtung areas tended to contain higher levels of Hg than the average. The map will be useful in future research in determining the geographic distribution of the regional patterns of fish Hg content, the relationship between Hg and parent soil material, and industrial emission.

INTRODUCTION

Mercury is widely used in amalgams, scientific instruments, batteries, arc lamps, the extraction of gold and silver, and the electrolytic production of chlorine. Its salts are used as fumigants in combating plant diseases and insect pests. Mercury may occur in several forms ranging from elemental to dissolved organic and inorganic species; however, some microorganisms have the ability to convert less hazardous organic and inorganic forms of mercury to the highly toxic methyl and dimethyl mercury [1]. The most devastating incident of mercury poisoning in human history resulted from the ingestion of sea food taken from Minamata Bay, Japan, during the late 1950s. Out of the total 111 cases reported, 43 people died. Babies born of afflicted mothers suffered congenital Defects [2]

Map illustrating the geographic distribution of soil Hg in Taiwan should contribute to the better understanding of the correlation between the soil Hg content and the emission of mercury from industries. In addition, the map will be useful in considering the consequences of Hg content in fish.

Geostatistics provides a means to characterize and quantify spatial variability, to use this information in rational interpolation, and to estimate the variance of the interpolated values. Variance estimation provides valuable information on the sampling density and configuration necessary for estimating a property to a specified precision.

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Geostatistics has been used to characterize spatial variability and to map a variety of soil properties at scales ranging from centimeters to kilometers, and it may prove useful across even greater distances [3,4]. Examples of other works include those of Burgess and Webster [5], Bierkens and Burrough [6], Bourennane *et al*. [7], Davies [8], and Smith *et al*. [9] among others.

Studies by the Environmental Protection Administration (EPA) in a collaborative research program initiated in 1983 aimed to determine the As, Cd, Cu, Cr, Hg, Ni, Pb and Zn trace element contents in soils and other soil properties, such as cation-exchange capacity and pH. Soils were sampled from 897 sites representing important agricultural production areas across Taiwan as described in reports on the elemental contents of soils in Taiwan. Total soil Hg ranged from 0.01 to 2.61 mg kg^{-1}, with an estimated arithmetic mean of 0.16 mg kg^{-1}. In another study, Chen and Lee [10] have detailed the Hg contents of 101 soils in 21 representative agricultural soil profiles in Taiwan, ranging from 0.04 to 1.62 mg Hg kg^{-1} with an arithmetic mean of 0.23 mg kg^{-1}.

The primary objective of this study was to develop a map illustrating the geographic distribution of Hg in surface soil horizons of Taiwan using geostatistics and geographic information systems (GIS). We used variography, linear regression, and sensitivity analysis to characterize the spatial variability of soil Hg using based on the data from EPA-ROC. We used geostatistical interpolation, i.e., punctual kriging, to estimate and map the total soil Hg content throughout Taiwan

MATERIALS AND METHODS

1. Data

Data were derived from the EPA-ROC studies described above. The sampling sites for the study are shown in Fig. 1. The sampling was conducted from 1983 through 1986. Samples were taken from geographically well-distributed sites at a target interval of 4 km network. Soils were sampled at a depth of 0-15 cm. Total soil Hg for 897 samples was determined by mean of acid digestion, followed by cold vapor atomic absorption technic.

2. Geostatistical Methodology

Geostatistics [11-13] consists of a collection of techniques for the analysis of spatially correlated data. Such geostatistical techniques as kriging incorporate the spatial or temporal characteristics of actual data into statistical estimation processes. These techniques can be linear, such as point kriging, ordinary kriging and block kriging.

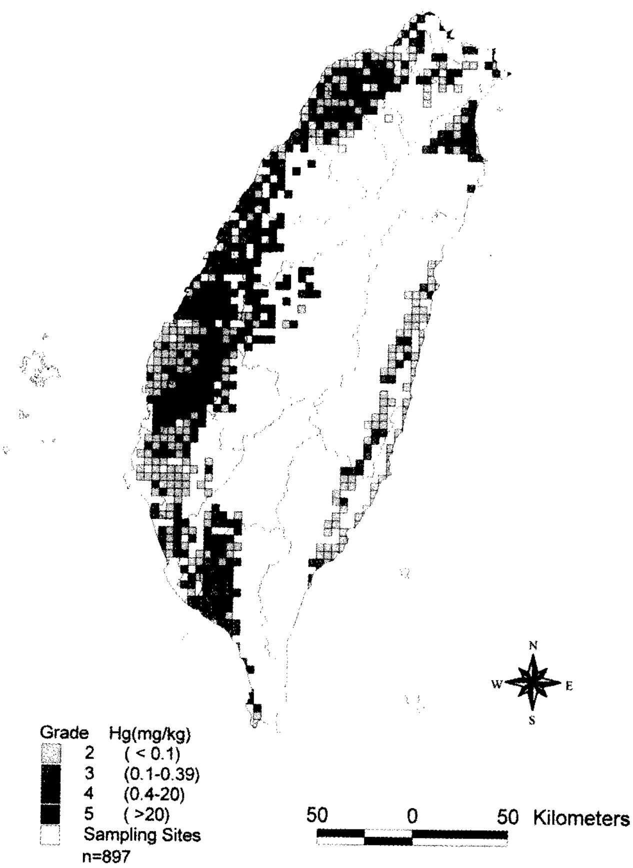

Fig. 1. General map showing the soil sampling sites and Hg grades in Taiwan.

Geostatistics provides a model for the spatial correlation of data within a statistical framework, including spatial and temporal covariance functions. Not surprisingly, these models are generally 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. A great deal of collected environmental data indicate that points which are closer in a given direction display higher correlation values than do those that are separated farther.

Geostatistical estimates are calculated as weighted sums of the adjacent sampled concentrations. These weights depend on the exhibited correlation structure. To illustrate, if data appear to be highly continuous in space, those points closer to the estimated points receive higher weights than do those farther away. The criterion for the selection of these weights is the minimization of the estimation variance. In this framework, geostatistical estimates may be regarded as most accurate among all linear estimators (i. e., the Best Linear Unbiased Estimator).

3. Geostatistical Structural Analyses

The first task in any geostatistical investigation is to identify the variogram of the investigated variable in space or time. This task, referred to as variography, is usually performed by determining the estimated variogram of the data collected. Variography is initiated by grouping the available pair-values into a number of lags or distance classes in accordance with their in-between distances. Variograms provide a means of quantifying the commonly observed relationship where samples close together tend to have more similar values than samples farther apart. The variogram $\gamma(h)$ is defined as

$$\gamma(h) = \frac{1}{2} Var[Z(x) - Z(x+h)] \qquad (1)$$

where (h) is the lag distance separating pairs of points and Var is the variance of the argument. $Z(x)$ is the value of the regionalized variable of interest at location x, and $Z(x+h)$ is the value at the location $x + h$. An experimental variogram $\gamma^*(h)$, is given by

$$\gamma^*(h) = [1/2n(h)] \sum_{i=1}^{n(h)} [Z(x_i + h) - Z(x_i)]^2 \quad (2)$$

where $n(h)$ is the number of pairs separated by the lag distance h.

The main features of a typical variogram are three-fold: the (1) range, (2) sill, and (3) nugget effects. Range is the distance at which the variogram reaches its maximum value. A pair of samples whose in-between distance is greater than the range is uncorrelated. This means that the range is regarded as a measure of the spatial continuity of the investigated variable. Sill, as the upper limit of the variogram which tends to level off at large distances, is a measure of the population variability of the investigated variable; generally, the higher the sill, the greater the variability in the population. The nugget effect is exhibited by the apparent jump in the variogram at the origin, a phenomenon which may be attributed to the small-scale variability of the investigated process and/or to measurement errors. The most accurateestimations can be determined if the investigated variable is well structured. Such a variable will have a variogram with a long range (i. e., high continuity), low sill values (i.e., small population variance) and a small nugget effect (i.e., insignificant small-scale variabilities or measurement errors).

Exploratory data analysis by linear regression were done using STATISTICA, Windows Version [14]. Geostatistical analysis was done using GS+ Version 3.1 [15]. We calculated variograms at lag intervals ranging from 1 km to 10 km across

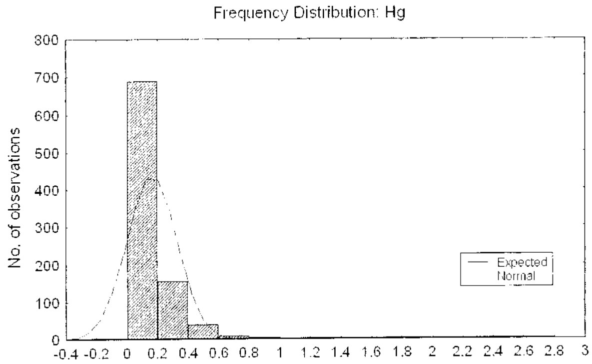

Fig. 2. Frequency distributions of soil Hg for conterminous Taiwan.

Table 1. Statistical summary of soil Hg data set and kriged estimates for conterminous Taiwan

	Units	Original	Kriged
n		879	8969
min	mg Hg kg^{-1}	0.01	0.02
max	mg Hg kg^{-1}	2.61	0.48
media	mg Hg kg^{-1}	0.12	0.13
mean	mg Hg kg^{-1}	0.16	0.14
std	mg Hg kg^{-1}	0.16	0.07
KS-c		0.027	0.009
KS		0.182	0.114
KS/KS-c		6.768	13.353
Skewness		5.503	1.212
kurtosis		61.96	1.491

the full extent of the data (382.8km). Ordinary kriging was carried out into a rectangular 106 column by 211 row grid consisting of 22366 2 by 2 km cells. Sixty-four neighboring data points were used to krige each cell. Kriged estimates and estimated standard deviations were transferred to Microsoft Excel 97 [16], sorted to conform with the vector display format of the GIS, Arc/Info [17], and then transferred to Arcview 3.0 [18] for analysis, reclassing, and display.

RESULTS AND DISCUSSION

Descriptive statistics and frequency distribution histograms for the data sets are shown in Table 1 and Fig. 2. The kriged estimates of total soil Hg are shown on the interpolated map (Fig. 3).

1. Variography

A global experimental variogram calculated at a representative lag interval of 3 km across the full extent (251.4 km) of the data (n = 897) is shown

Table 2. Least squares best-fit variogram model parameters at various lag intervals. In all cases, the best fit was an exponential model: $\gamma(h) = C_0 + C[1 - \exp(-h/A_0)]$, where $\gamma(h)$ = variance at lag interval h, C_0 = nugget variance, C = tructural variance, and A_0 = range parameter

Maximum lag distance (MLD) (km)	Lag interval (h) (km)	Nugget (C_0) mg^2Hgkg^{-2}	Sill ($C+C_0$) mg^2Hgkg^{-2}	Range[a] parameter (A_0) (km)	Estimated[b] range ($3A_0$) (km)	Model reduced sum of squares $\times 10^{-4}$	Model R^2
251.4	1.0	0.000	0.026	3.7	11.1	57.460	0.168
251.4	1.5	0.002	0.026	5.6	16.8	28.200	0.212
251.4	2.0	0.003	0.026	4.5	13.5	18.170	0.200
251.4	2.5	0.003	0.027	5.3	15.9	12.890	0.275
251.4	3.0[c]	0.005	0.027	7.8	23.4	10.390	0.289
251.4	3.5	0.004	0.027	6.5	19.5	7.852	0.287
251.4	4.0	0.005	0.026	2.4	7.2	6.593	0.044
251.4	8.0	0.006	0.027	3.5	10.5	2.764	0.052
251.4	10.0	0.006	0.027	6.2	18.6	2.061	0.123
382.8	4.0	0.000	0.023	3.6	10.8	0.205	0.015

[a] in the exponential model is not the range but a parameter indicative of the range.

[b] Range in the exponential variogram model is usually assumed to be the point at which the model attains about 95 % of the sill ($C + C_0$), which can be estimated as 3 A_0.

[c] Parameters of the adjusted least squares best-fit model at a lag interval of 3 km, with C_0 and A_0 adjusted to provide better fit at the smallest lags. This is the model used for kriging.

in Fig. 4. At short to moderate lags, i.e., 1 to 10 km, the variogram was relatively stable. The variance increased from a nugget variance of approximately 0.000 mg^2 Hg kg^{-2} to approach a sill of about 0.006 mg^2 Hg kg^{-2}, which remained relatively constant up to a lag distance of about 251.4 km. Beyond 251.4 km, the variance increased and then became extremely large. Instability in the variogram at long lag distances has been noted by others [19], and as a result, variograms are typically calculated only to one-half the maximum distance between points [20].

Experimental variograms were calculated for the data at a variety of lag intervals to a maximum lag of 251.4 km. Least squares model fitted of these variograms generated a relatively consistent set of best-fit models. Parameters from representative models are shown in Table 2. An exponential model always had the best fit compared with the other models (linear, linear with sill, spherical, or Gaussian) available in the software. The exponential model is of the form

$$\gamma(h) = C_0 + C[1 - \exp(-h/A_0)] \quad (3)$$

where C_0 is the nugget variance ($C_0 \geqq 0$), C is the structural variance ($C \geqq C_0$), and A_0 is a range parameter, different from the true range in a spherical or linear-sill model. Range in the exponential variogram model is usually assumed to be the point at which the model attains about 95 % of the sill ($C + C_0$), which can be estimated as 3 A_0.

The modeled nugget variance ranged from 0.000 to 0.006 mg^2 Hg $^{-2}$corresponding to a standard deviation of from 0.111 to 0.181 mg Hg^{-1}. The experimental error in Hg determinations was usually less than this, suggesting the presence of spatial variability at lags smaller than those in the data set. The soil element content exhibited different levels of spatial variability at different scales, and the variogram across the extent probably contained localized spatial variation nested within it [21]. The modeled sill was very consistent, ranging from 0.023 to 0.027 mg^2 Hg kg^{-2}, less than the sample standard deviation of 0.16 mg Hg kg^{-1}. The range estimated from the model range parameter varied from 2.4 to 7.8 km, beyond which the soil Hg exhibited no significant spatial correlation.

2. Interpolation via Kriging

Kriging from several of the variogram models shown in Table 2 produced only minor differences in the results, so we only present here results obtained with a representative lag interval of 3 km. The least squares best-fit model for the variogram calculated with a 3 km lag interval is shown superimposed on the variogram in Fig. 5; model parameters appear in Table 2. Kriging is particularly sensitive to model parameters at the shortest lags, but a least squares best-fit model may not fit the variogram well at the smallest lag intervals [11,19]. To obtain a better fit for the nugget variance and the initial portion of the variogram, the best-fit model nugget variance (C_0) and range parameter (A_0) were adjusted slightly. Weighted least squares methods giving greater emphasis on points at smaller lags are an alternate approach

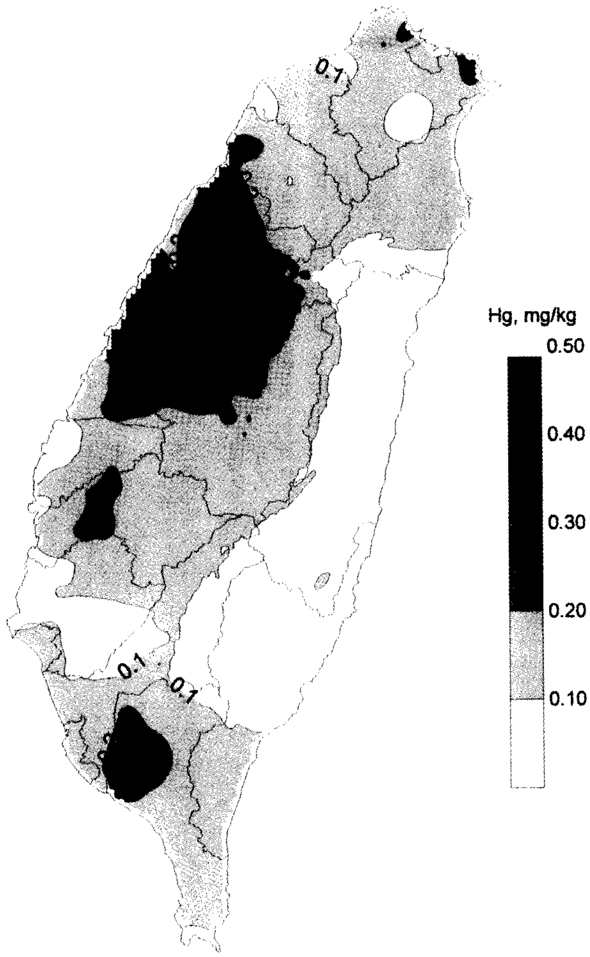

Fig. 3. Map of the Kriged estimates of the total soil Hg for conterminous Taiwan.

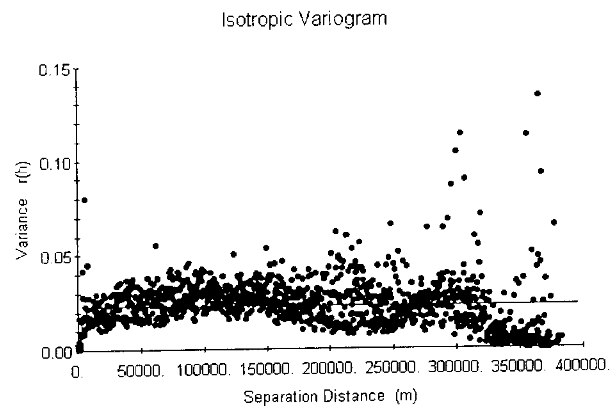

Fig. 4. Global experimental variogram of total soil Hg calculated using a lag interval of 3 km across the full extent of the data.

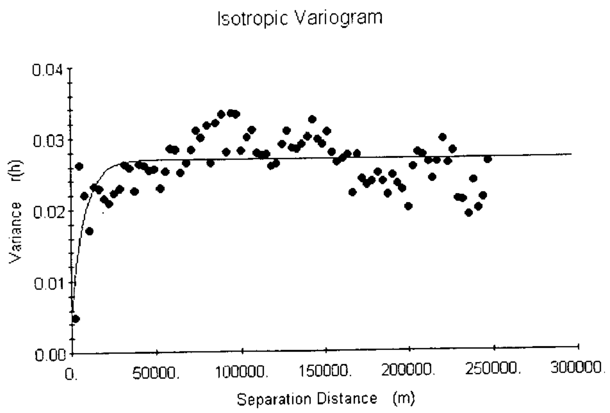

Fig. 5. Global variogram of total soil Hg calculated using a lag interval of 3 km and a maximum lag distance of 251. 4 km, and using the least squares best-fit variogram model.

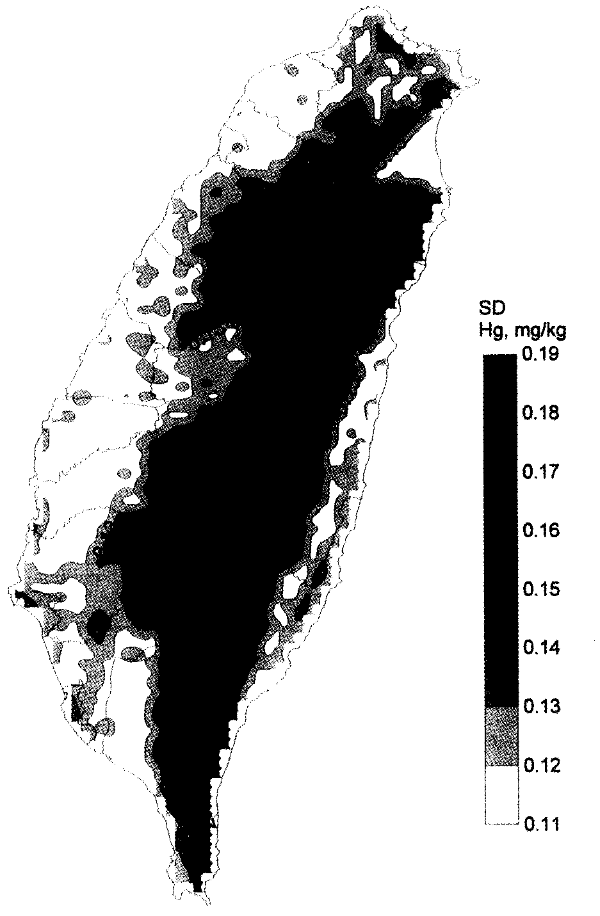

Fig. 6. Map of the estimated standard deviations of the kriged estimates of total soil Hg for conterminous Taiwan.

[22]. The adjusted model is shown superimposed on the variogram in Fig. 5, and the adjusted model parameters are listed in Table 2.

Kriging from the adjusted least squares best-fit model (Fig. 5) and classifying the results by means of deciles produced the map of total soil Hg shown in Fig. 3. We chose decile classification because it provided a geographical representation of the frequency distribution of the kriged estimates. No other statistical differentiation between these classes is meant, nor should it be inferred. Descriptive statistics for the 22366 Hg estimates included in this map are shown in Table 1, and the estimated standard deviations of the kriged estimates are mapped in Fig. 6. The mean total soil Hg for Taiwan estimated from kriging was similar to that from the original data, 0.16 vs. 0.14 mg Hg kg^{-1}, respectively. The kriged estimates had a smaller range than the original data, with both the minimum and maximum truncated slightly. Kriging also normalized the data somewhat, reducing the skewness and kurtosis compared with the original data.

The kriging estimated standard deviations of Hg ranged from 0.111 to 0.181 mg Hg kg^{-1}, with a mean of 0.14 mg Hg kg^{-1}. These estin ,ted standard deviations reflect both the variability of soil Hg, as indicated by the standard deviation of the original data (0.16 mg Hg kg^{-1}), and the uncertainty inherent in interpolating from widely dispersed sites. The nugget variance comprises both of these sources of variability, and represents, in effect, the minimum variance that can be expected for a kriged estimate. Expressed as a standard deviation, the modeled nugget variance was equivalent to 0.005 mg Hg kg^{-1}. In comparison, the estimated standard deviations for the kriged estimates were quite reasonable. The highest estimated standard deviations (Fig. 6) occurred where data were sparsest (Fig. 1). This consequence of the fact that variances estimated from kriging depend only on the variogram model and the sample site configuration; they are not a direct measure of the variance of the neighboring data used to estimate an unsampled point. Acquiring additional data in undersampled areas would reduce estimation variance.

The kriged map of total soil Hg indicated that there were three hot spots in Taiwan around Toufen, Changhua and Pingtung areas. The major source c the mercury emission was suspected to be from chlor-alkali plants employing electrode, metal finishing and industry park discharge respectively.

Kriging is a statistically based interpolation method that has proven effective in mining, soil analysis, ecology, and other disciplines. Kriging is considered to be very sensitive to violations of the basic assumptions, yet it has proven very robust in many circumstances. Simple sensitivity analysis was useful here in judging the effects of anisotropy estimation via kriging. Other studies have shown that simple punctual kriging following careful exploratory statistical analysis and proper variography can yield reasonable results despite the violations of the assumptions. Statisticians are working to develop more robust geostatistical methods to handle difficult data [19,20,22,23].

Geostatistical analysis of total soil Hg data from 897 sites across Taiwan produced an interpretable exponential variogram (Fig. 5). Soil Hg showed spatial correlation at distances up to 23.4 km, a much greater range than reported for other soil properties. Interpolation by kriging produced digital maps showing the geographic distribution of soil Hg (Fig. 3) and estimates of the standard deviations of the interpolated values (Fig. 6). Estimation of the variance associated with interpolation is a major benefit of kriging, and one that other interpolators lack. The estimated standard deviations for soil Hg were high relative to the estimates themselves, emphasizing the uncertainty inherent in interpolating across large areas from variable and widely scattered data. The standard deviation map will be useful in guiding additional sampling or data acquisition to improve map accuracy and precision. Digital maps facilitate such improvements, allow analysis and manipulation in a GIS, and permit electronic data transfer.

Readers are cautioned to consider the nature of the original data, their extent and spatial variability, the kriging resolution, and the estimated standard deviations when using and interpreting these maps. There are biases inherent in the data set used. The EPA study sampled agricultural soils where Hg content might have increased by the addition of Hg-containing sludge. Some soils exhibit considerable variation in the total Hg content in different horizons within the profile [10]. Further, we emphasize that the maps delineate gross trends in total soil Hg on the scale of conterminous Taiwan. Small-scale interpolated maps show broad, average trends, and may misrepresent them as continuous or as having greater precision than the data allow. In reality, local situations may be much more complex, e. g., where parent materials change abruptly within relatively small distances. A global variogram determined across a large and diverse land area may be inadequate to represent regional or localized spatial variability. Therefore, regional analyses of spatial variability, with regions based perhaps on soil parent material, may provide more accurate and precise estimates of the total soil Hg. The magnitude of the standard deviations emphasizes the uncertainty inherent in interpolating from variable, widely spaced data. The map of estimated standard deviations indicates clearly where an investment in obtaining more data would reduce estimation variances and improve the map.

CONCLUSIONS

This study has shown the ordinary kriging following careful exploratory statistical analysis and proper variography can yield reasonable results. The kriged map of total soil Hg indicated that there were three hot spots in Taiwan around Toufen, Changhua and Pingtung areas. The mercury emission from industry like chlor-alkali plants might be the predominant factor. Geostatistics and GIS are essential tools for analyzing georeferenced information and advancing our understanding of spatial variability at various scales. Geostatistics and GIS will be indispensable in characterizing and summarizing this information to provide quantitative support for decision and policy making for agriculture, health and natural resource management.

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以地理統計與地理資訊系統方法建立土壤汞分佈圖

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摘　　要

本文利用環保署針對全台灣主要農業地帶大樣區土壤重金屬含量調查結果，其中之表土（0～15公分）汞含量資料，以地理統計方法及地理資訊系統工具建立全台灣土壤汞分佈圖。表土汞含量之範圍由0.01至2.61 mg/kg，算術平均值爲0.16 mg/kg。經半變異圖分析，並以最小平方法摸擬建立之指數半變異模式推估克利金網格值涵蓋全台灣，並以此建立台灣表土汞含量分佈圖。本研究結果雖然無法顯示局部高濃度地區之細微變化，但對概況趨勢仍具參考價值。圖中台灣有幾處表土汞含量較高地區，分別是苗栗縣頭份附近，彰化市附近及屏東市附近，主要原因可能分別是由鹼氯工廠、金屬處理加工廠及工業區排放含汞污泥處置不當所造成，汞分佈圖有利於對將來汞來源之追蹤及地區性魚體、土壤母質中汞含量分佈相關性之研究。