

A Practical Examination of the Use of Geostatistics in the Remediation of a Site with a Complex Metal Contamination History

R. J. GLAVIN AND P. S. HOODA

Centre for Earth and Environmental Science Research, School of Earth Sciences and Geography, Kingston University, Kingston-upon-Thames, Surrey, United Kingdom

Targeted remediation strategies offer the potential to treat only those areas where contamination exceeds predefined threshold levels. We used geostatistical techniques to characterize spatial distribution of heavy metals across a contaminated site, with the aim of delineating the contaminants, which is essential for successful implementation of targeted remediation strategies.

Samples collected from three depths, 0–20 cm, 20–40 cm and 40–60 cm at 50 sample locations, were analyzed for As, Sb, Hg, Pb, Cd and Cu contents. The geostatistical analysis of this data enabled the identification of a number of contamination hotspots and trends. The visual interpretation of the data was supported by the statistical analysis in the form of Spearman's rank correlation coefficient. Additionally, classical statistics, based on the central limit theorem, showed that, in terms of obtaining the true mean for each of the contaminants within acceptable limits of precision, the site has been more than adequately sampled.

It has been demonstrated that kriging can offer the potential to map the spatial distribution of contaminants. However, the possibility of an undetected hotspot remains, even when probabilistic modelling and a secondary phase of validity sampling are employed. This together with the large number of samples required may preclude the commercial use of geostatistics in the remediation of contaminated land.

Keywords Metals, semivariogram, kriging, targeted remediation, remediation design, soil contamination.

Introduction

In the UK during the 1970s, local authorities began to experience problems in relation to the redevelopment of sites contaminated by their previous uses (Harris, 1987). The site examined by this study represents a small fraction of such an area. To date, remediation of the site has, particularly in the more recent stages of development, generally involved removing potentially contaminated material up to a depth of two meters and replacing it with uncontaminated soil. As is commonly the case with remediation schemes of this nature, the contaminated material evacuated from the site is sent to landfill (Sham, Pers. Comm., 2002). The Environment Agency estimates that there may be some 300,000 ha, approximately 1.8% of the United Kingdom's landmass affected to some extent by contamination

Address correspondence to P. S. Hooda, School of Earth Sciences and Geography, Kingston University, Kingston-upon-Thames, Surrey KT1 2EE, UK. E-mail: p.hooda@kingston.ac.uk

(DETR, 2000). Current government legislation, driven by an ever-increasing demand for housing, is aimed at putting this land back into productive use through remediation. In addition, the Landfill Directive, 1999/31/EC, aims to reduce the volume of material sent to landfill (Gregory, 2001). Targeted remediation schemes, achieved through the spatial analysis of contamination, therefore can help identify areas that require remediation, offering the potential to achieve both of these goals.

A characterization of the spatial distribution of pollutants in soils can be an important aspect in the processes of remediation, risk assessment and the identification of potential pollutant pathways (Lin, 2002). One of the prerequisites to this characterization is a phase of site investigation, which must include an adequate sampling strategy. Although site investigations generally include soil sampling to identify potential contamination linkages and locate contamination hotspots, distributions are generally not mapped. In the Environment Agency's Contaminated Land Exposure Assessment (CLEA) guidelines for assessing the risk to human health from contaminated land (EA, 2002) hotspots are not necessarily of concern, for example. The site assessments may simply consider an "appropriate averaging area" ranging from a single garden on a housing estate to the whole of an informal play area (ENDS, 2002). Accurate maps of the levels of contamination across a site, however, offer the opportunity for greater understanding of the potential environmental impacts of that site, giving a sound risk assessment combined with the possibility of targeted rather than wholesale treatment. Indeed, research by Saito and Goovaerts (2003) indicates that targeted strategies can significantly reduce the cost of remediation. Hence, understanding the distribution of any given contaminant could deliver more efficient and effective remediation practices. Assessment of the extent of soil contamination in this manner involves the extrapolation between sampling locations; geostatistics offers the potential for such extrapolation.

Geostatistics is the term applied to the distinct culture of terminology and methodology used in kriging. Kriging is a method of estimating spatially distributed values from known data points (Swan and Sandilands, 1995). Geostatistics is based upon fundamental assumptions concerning the behaviour of random variables in a spatial co-ordinate system. At its heart is the theory of regionalized variables; this postulates that the real world is a realization of a random function. By making observations it is possible to deduce some of the properties of the random function model and use these properties to constrain estimates of the realization at unmeasured locations.

Unlike traditional spatial estimation methods, geostatistics is not *ad hoc*: it is founded on a clear hypothesis and assumptions about spatial variables (Frances, 1998). In brief, the distance between known data points (lag) is plotted against their similarity (covariance) to produce a semivariogram. In theory sample points with a lag of zero should be identical; as the distance between points increases their covariance increases (i.e. they become less similar) until they reach a point at which values are no longer dependant upon each other. The lag at which this occurs is known as the range and the covariance the sill. In short the semivariogram provides information concerning the relationship between the sampled data, this information is used to estimate values at unsampled locations (kriging). The concepts and methodology of geostatistics is described in detail by Isaaks and Srivastava (1989) and Swan and Sandilands (1995). Whilst geostatistics is becoming more widely used in environmental and soil science its application in the design of remediation strategies is rarely used and could become an important process in bringing contaminated sites back into productive use.

The aims of this work are to illustrate the potential value of geostatistics in the design and implementation of targeted remediation strategies. Maps produced for a series of depth

intervals will be used to delineate both metal contamination hotspots and trends and hence illustrate the potential value of geostatistics in the design and implementation of remediation strategies.

Materials and Methods

Study Site

For reasons of confidentiality the name and exact location of the site cannot be given. This also means that the details given in this section concerning the site history must also be limited. The site itself forms part of a much larger development in South London. Redevelopment of the area began in the 1960s, prior to which the land had been owned by the Ministry of Defence and was used for a variety of industrial processes. In the 1970s, as redevelopment began to gather pace, severe contamination from the manufacture of munitions, gas generation and dumping of waste material was encountered (Harris, 1987). Historically, the entire site was marshland and formed part of a natural drainage basin. As the site developed this marshland was filled using the wastes from the various industrial processes in operation. The industrial origin of the material used in this reclamation has resulted in many of the present problems of contamination associated with the site. In addition, it was common practice for buildings, roads and tracks to be built on foundations constructed from material of an industrial origin (Lowe, 1984). The complex pattern of contamination resulting from these practices is compounded by limited information on both the evolution of the site and the processes in operation throughout its development. Nevertheless, a site investigation commissioned by the Greater London Council in 1980 identified a range of industrial activities, including metal forging, non-ferrous metal foundries, metal plating, development of paints, manufacture and testing of weapons and explosives, destruction of explosives and incendiary devices and coal gas production (Lowe, 1984). The investigation report also identified a number of contaminants commonly produced by these processes, including Pb, Cu, Cd, Hg, Sb and As (Lowe, 1984).

Site Sampling and Soil Analysis

The study area comprises approximately 21,759 square meters. No remediation work has been carried out on this part of the site. Fieldwork was conducted in June 2002. Based on hand augered sections taken from the site prior to the main sampling, and accounting for resource limitations and time restrictions, the soil profile was split into three layers, 0–20 cm, 20–40 cm and 40–60 cm. These layers broadly reflect the evolution of the site. In brief, the upper twenty centimeters of the fill profile generally consisted of organic rich sandy soil; this was light brown in color and contained some tailings material, ranging in size from a few mm to 2 cm. This layer gradually graded into loose dark brown/grey sandy gravel consisting of industrial tailings. Beneath this was pale brown/grey silt rich clay. This layer also contained some tailings material, particularly in the upper few cm. The tailings material described is a waste product, likely to have been derived from the variety of industrial processes previously in operation on the site. The gradational nature of the three layers made their exact boundaries difficult to discern. However, the geometry of the site is broadly flat and the three layers appear sub-parallel to this surface. Sample depths were therefore kept constant.

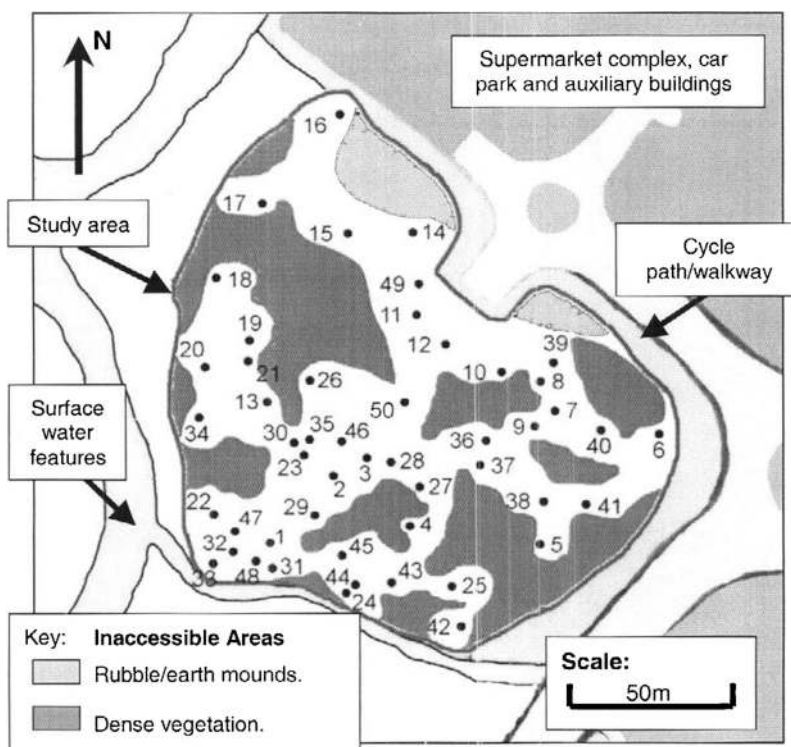


Figure 1. The map illustrating sample locations.

For all three layers, hand augered samples were taken at 50 sample points, giving a total of 150 samples. In general, it is considered that a minimum of three samples per sampling point is required to provide adequate information on the vertical spread of contamination (Lord, 1987). For kriging, variograms computed on fewer than 50 data are of little value (Webster and Oliver, 1992). Sample locations are illustrated by Figure 1. In some instances the stony nature of the soil combined with the roots of trees meant that it was not possible to obtain a full sample for the 40–60 cm interval. Figure 1 also illustrates those parts of the site made inaccessible, either through the growth of thick vegetation, mainly nettles and brambles, or through the deposition of rubble arising from the recent construction of a walkway/cycle path around the site. The location of these areas was noted and subsequently mapped during fieldwork.

The sample strategy employed was random, based on predefined random bearings and distances. However, the inaccessibility of parts of the study area made this difficult to implement in the field. Random sampling is most commonly used at sites such as this, where the spatial distribution of contamination is anticipated to be highly variable (Byrnes, 1994). Random sampling reduces the probability of missing linear features running across the site, but tends to produce clustering (Lord, 1987), as seen in Figure 1. The occurrence of clustering, however, was increased by the presence of large areas from which samples could not be taken. The disadvantage of this clustering is that parts of the study area remain under-sampled.

Soil samples were oven-dried (40°C), disaggregated and passed through a 2-mm nylon sieve, and this <2 mm fraction was used in the soil analysis. Total metal contents in the

samples were estimated by digestion in concentrated HNO_3 (15.5 M) following standard laboratory procedure (Hooda et al., 1999). The metal concentrations in the digest solutions were then analysed by inductively coupled plasma emission spectrometry (ICP-AES).

Geostatistical and Statistical Analysis

Geostatistical analysis was employed to analyze the spatial distribution of contamination. The production of maps through kriging and the associated variogram modelling was performed using Krig-Tree, a geostatistical package developed by Eni Spa. Also, based on the central limit theorem, classical statistical analysis was used to calculate the optimum number of soil samples. As such it was assumed that the real mean (μ_0) is equal to the sample mean (μ) with additional sample means of $\mu \pm 10\%$, $\mu \pm 20\%$ and $\mu \pm 30\%$. The optimum number of samples (N) required to give a meaningful average within the required limits was calculated using standard score (Z) against the desired degree of probability using the expression (where σ is the standard deviation):

$$N = \left(\frac{Z\sigma}{\mu - \mu_0} \right)^2 \quad (\text{From Hooda } et al., 1986) \quad \text{Equation 1}$$

In addition to the use of classical statistics, Spearman's rank correlation coefficient was used to assess the relationship between the different contaminants and layers. The Spearman's rank correlation coefficient provides a reliable measure of the strength of the relationship between two variables (Wilson, 1984).

Results and Discussion

Example results of the geostatistical analysis of the data can be seen from Figures 2 to 7. These are typical of the results obtained and include the semi-variograms and resultant maps for As (layer 1), Pb (layer 2) and Hg (layer 3). The somewhat "spiky" nature of the experimental variogram for As is typical of those for layer 1 (Figure 2). This spiky nature combined with the short range of the model variogram is indicative of data with little or no spatial dependence, probably a reflection of the chaotic way in which waste has been deposited across the site. Additionally, anthropogenic influences such as fly tipping may have affected contaminant distributions in this layer. The model variograms for layers 2 and 3 (Figures 4 and 6) are, however, generally much smoother, indicating a much greater degree of spatial dependence. All three of the maps (Figures 3, 5, and 7) contain a number of contamination hotspots. Table 1 gives the values used to denote contamination hotspots for each of the elements mapped. The map of the distribution of contamination for As (Layer 1) (Figure 3) shows two hotspots, both of which are located in the southeastern portion of the study area. These are delineated by tightly packed contours illustrating rapid changes in contaminant concentrations. Similar hotspots can be found on the maps for Cd and Sb for this layer. In addition, the more central of the two hotspots is also found on the map of As for layer 2. These similarities are also evident from the statistical analysis of the data, for example the Spearman's rank correlation coefficient between As and Sb is 0.6, whilst the correlation coefficient for As and Cd is 0.5.

The hotspots on the map for Pb (layer 2) (Figure 5) are again marked by tightly packed contours, further illustrating the rapid changes in contaminant concentrations. This map is most similar to those for Hg and Cu for layer 2; furthermore, both hotspots have similar locations on the maps of Sb and Cd (layer 2). When comparing the three layers it can be seen

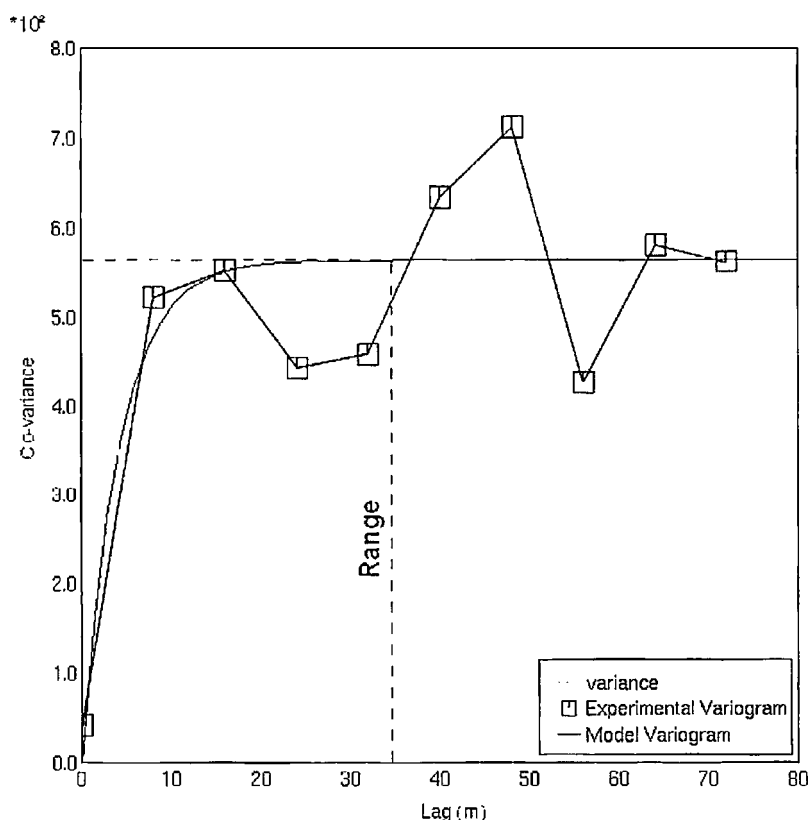


Figure 2. The variogram for arsenic, layer 1. (Lag in m)

that this map is very different to that of Pb for layer 1; however, a number of similarities, including the location of the two hotspots, can be made with the map of Pb for layer 3. Again a number of the observed similarities are also evident from the statistical analysis of the data. For example the correlation coefficient between Pb and Cu is 0.6 as is that between As and Sb.

Examining the distributions produced for Hg (layer 3) (Figure 7) reveals two hotspots, both of which are located in the southern portion of the study area. Like the hotspots for the preceding figures these are marked by tightly packed contours. In addition to the two

Table 1

Values used to denote contamination hotspots for each of the elements. Since one of the prerequisites of the confidentiality agreement reached with the site owners is that legislative or guideline values may not be used, these values are based on the data for each of the elements

	As	Hg	Sb	Pb	Cd	Mg	Cu
Level of contamination indicative of a hotspot (mg/kg)	35	12	16	1500	12	4000	3000

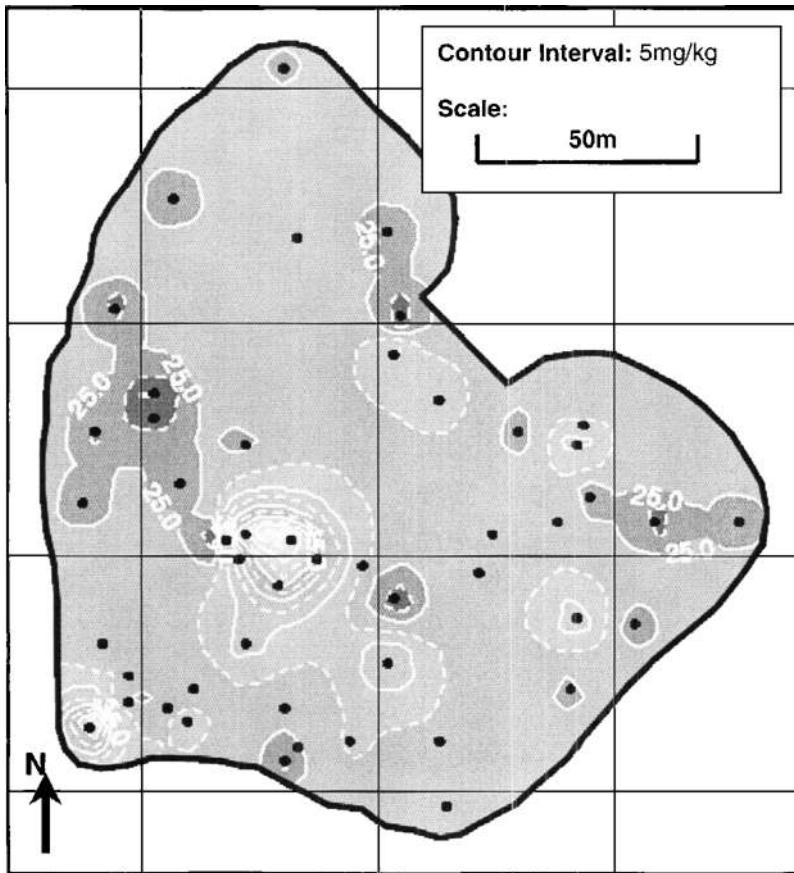


Figure 3. The map for arsenic, layer 1. Two hotspots can be seen from this map, in the center and south west of the study area; both are denoted by tightly packed contours. Table 1 defines the values used to denote a hotspot for each of the elements.

hotspots, an east-west trend can be seen running across the center of the map. This map shows little similarity to those for layers 1 and 2 for Hg; similarities can, however, be drawn with the maps for Pb, Cd and Cu for layer 3. All four elements have similarly positioned hotspots whilst those for Hg, Pb and Cu have similar east-west trending features running across the center of the study area. As for layers 1 and 2, visual evidence of the relationships between elements is supported by the statistical analysis of the data. For example, the Spearman's rank correlation coefficient for Cu and Pb is 0.6 and the correlation confident between Cu and Cd is 0.5

From the geostatistical analysis of the data it has been possible to discern a number of contaminant hotspots and trends. As illustrated by the discussion of Figures 3, 5 and 7, many of the hotspots identified can be traced between both layers and elements; the correlation of which is supported by the statistical analysis of the data. Since one of the aims was to delineate contamination hotspots and thus promote the design of a targeted remediation strategy, the hotspots for all the contaminants analyzed have been plotted on a single map (Figure 8). The map shows that geostatistical analysis can provide a valuable tool to describe the distribution of contamination and for the delineation of hotspots. However, it

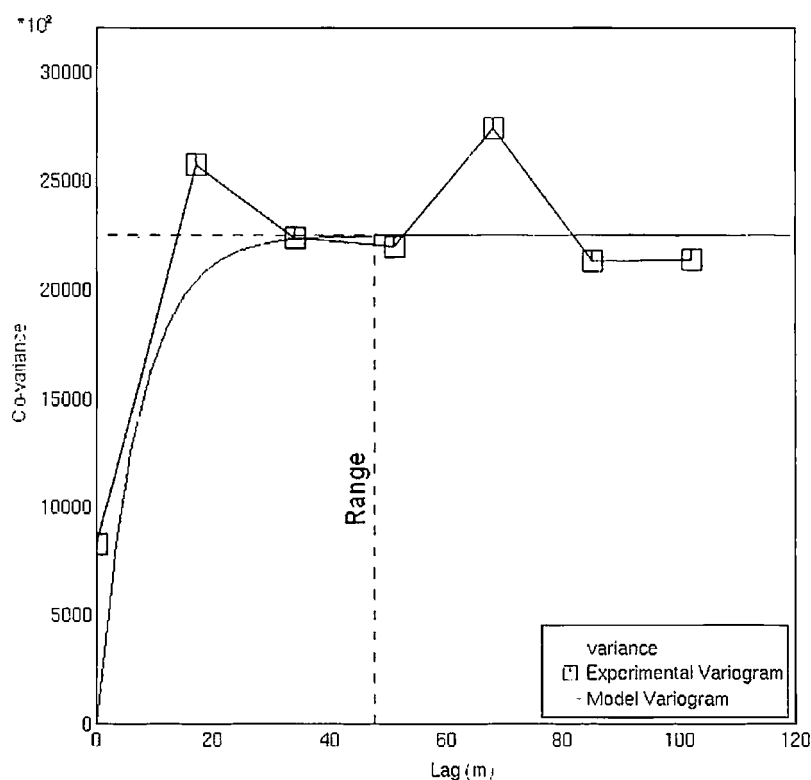


Figure 4. The variogram for lead, layer 2. (Lag in m)

also illustrates that, for sites such as this, where a complicated industrial past has produced a high density of hotspots covering a large portion of the site, targeted remediation may not always be practical. Furthermore, this map emphasizes the weakness of the sample strategy employed by this study. Whilst an important element of geostatistical analysis, clustering of data points, primarily the result of the inaccessibility of large portions of the study area, has meant that the areas that appear least contaminated occur where the sample density is also least. When combined with the complex evolution of the site and frugal information available on the probable distribution of contamination, the inadequate distribution of sample points means that there remains the possibility of hotspots being undetected.

Additionally, some of the maps produced through geostatistical analysis show evidence of bull's eyes. Bull's eyes result from the failure to extrapolate beyond the range of the control point values and are common to maps where data is sparse and points are spatially independent (Swan and Sandilands, 1995). Examples of such features can be seen from Figures 5 and 7. In general, these artefacts are most common to layer 3. Here the failure to penetrate beneath 40 cm at a number of sample locations means that this layer has 7 fewer data points than the overlying layers. As a result the initial data points become increasingly isolated, making them more prone to bull's eyes. Whilst in many instances a circular contour pattern may be considered indicative of a hotspot, in the case of isolated control points, assumptions concerning the distribution of contamination should not be made without further, more detailed, or targeted investigation.

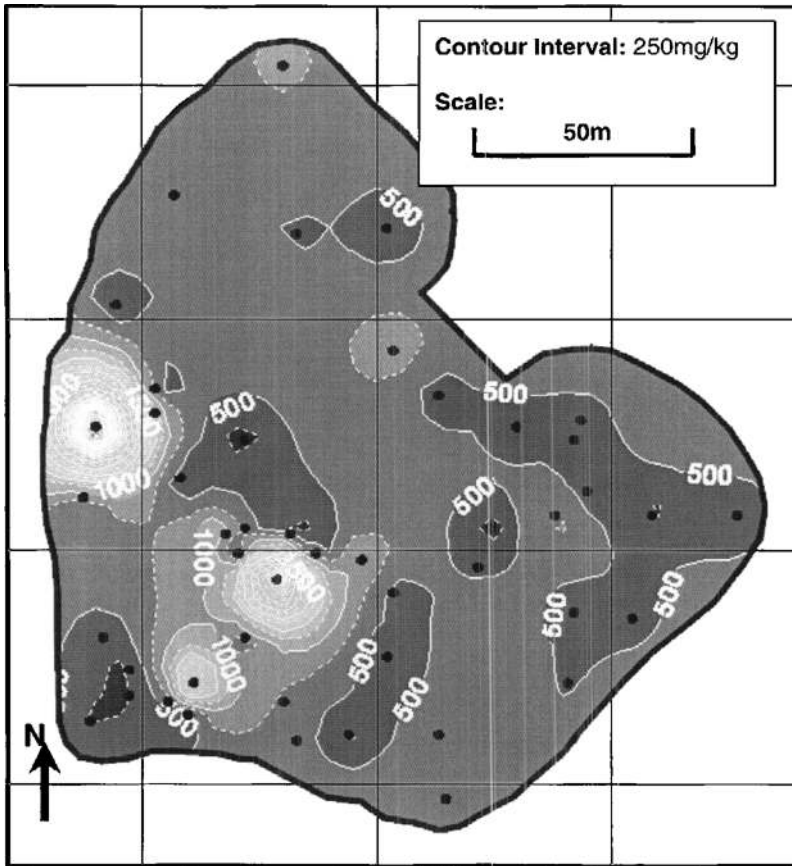


Figure 5. The map for lead, layer 2. Hotspots are again denoted by tightly packed contours and are a lighter shade than areas where concentrations are lower.

Had the study area been more accessible, a more balanced coverage of the site could have been achieved and the problems encountered reduced. Whilst the use of a systematic sample strategy could have ensured that the entire study area was equally sampled, the inaccessible nature of portions of the site rendered its implementation in the field impractical. Additionally, in the case of a regular grid there are no lags at an offset less than the grid interval and very little is known about the shorter lags, since the sample variogram does not provide information for distances shorter than the minimum spacing between the sample data (h). With a scattered data set separations are more evenly distributed across many values of h , although the variogram produced may be more difficult to interpret; there is likely to be significantly more data points with very short offsets (Isaaks and Srivastava, 1989). It is these short offsets, close to the origin, that are often the most crucial part of the variogram analysis (Frances, 1998). In addition, to achieve the same sample density across the study area a grid spacing of approximately 20 m would have been required. This is larger than some of the hotspots illustrated in Figure 6. Furthermore, in the case of a regular grid linear features, such as the numerous tracks and roads that previously dissected the site, running parallel or sub-parallel to sample points may also be missed. In such instances the probability of areas of elevated contamination remaining undetected can

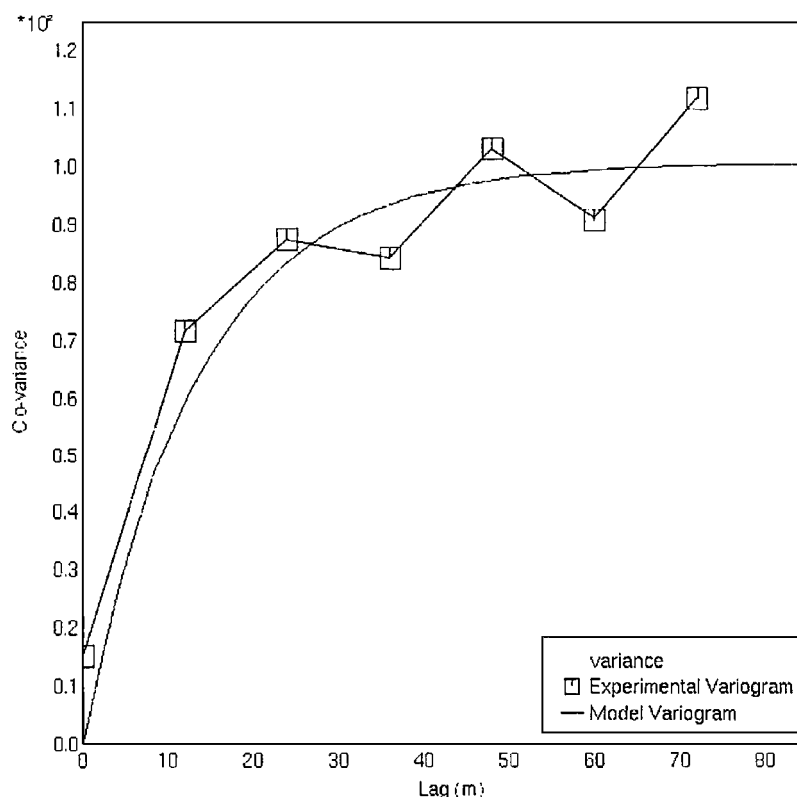


Figure 6. The variogram for mercury, layer 3. (Lag in m)

be reduced through the use of a smaller grid increment. However, this produces increased sample numbers, which has important implications in terms of both time and costs.

A more viable solution to the problems encountered could therefore be the use of a stratified random sample strategy. Not only do such strategies give a good general coverage of a site, but produce samples at a variety of offsets. In terms of the geostatistical analysis of contaminated land this type of sample strategy probably represents the most appropriate practice and could prove a valuable tool in the planning of a suitable remediation scheme. However, once again the inaccessibility of parts of the study area would have made a stratified random sample strategy difficult to implement in the field.

In many instances site investigations may involve more than one phase of sampling. Once the distribution of contamination has been analysed, areas considered worthy of further examination are revisited. This may help fully assess the extent of a contamination hotspot or to investigate areas considered under-sampled. In terms of the geostatistical analysis of contaminated land, this practice can be used to check the validity of estimations at unsampled locations and assign a level of confidence to the maps produced. Even a small number of additional samples can achieve these aims. The results of this study are worthy of such a second stage of investigation. Further information, even as few as ten samples, possibly taken during the winter months when the thick vegetation making the site inaccessible at the time of sampling had died back, could help to complete the picture of the distribution of contamination.

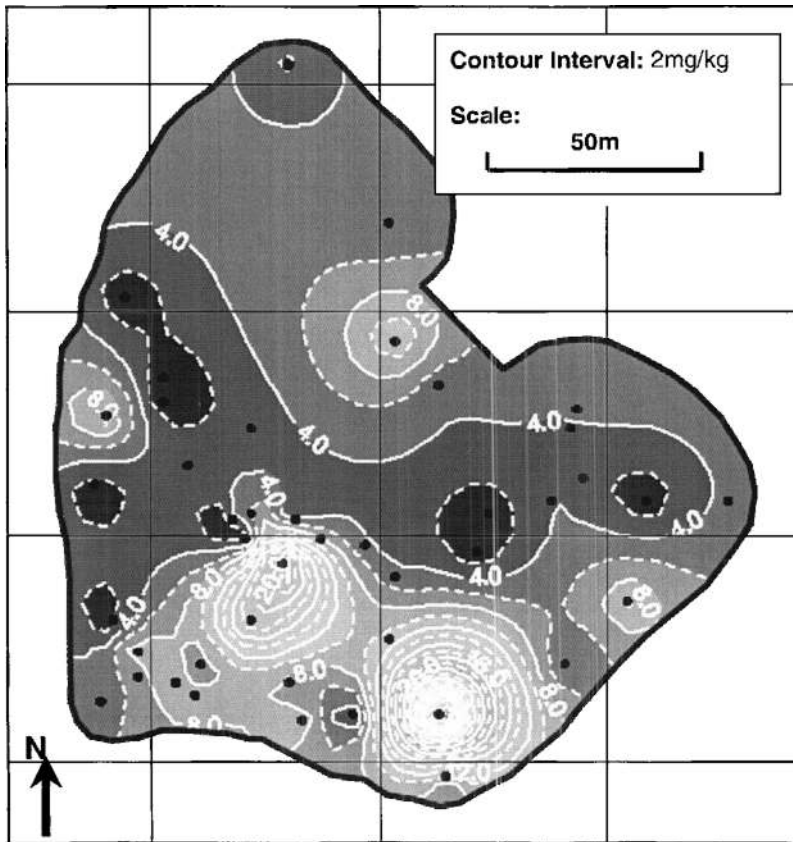


Figure 7. The map for mercury, layer 3. A clear trend can be seen running across the center of this map, whilst hotspots are delineated by tightly packed contours. Hotspots are a lighter shade than areas where concentrations are lower.

In addition, one of the functions of geostatistics is probabilistic modelling. In such instances a range of maps is produced, resulting in a series of contaminant distributions each with a different probability. For example, Barabas *et al.* (2001) and Saito and Goovaertis (2003) use indicator kriging to model the probability distributions of contamination combined with accuracy plots to assess the validity of the models produced. Probabilistic modelling has not been used as part of this study; however, it provides the opportunity to qualitatively assess the validity of any maps generated by kriging and hence assign a measure of uncertainty. Additional sample phases may be used to validate any maps and hence reduce these uncertainties.

Sample Numbers (Classical Statistics)

The number of soil samples necessary to sufficiently characterise an average for each of the contaminants was calculated from Eq. 1, using the means and standard deviations from the results of the laboratory analysis. For this calculation it was assumed that the true mean, μ_0 , was equal to the sample mean, μ , $\pm 10\%$, $\pm 20\%$ and $\pm 30\%$. Some of the values from this analysis are summarised in Table 2. From Table 2 it can be seen that, for a probability of

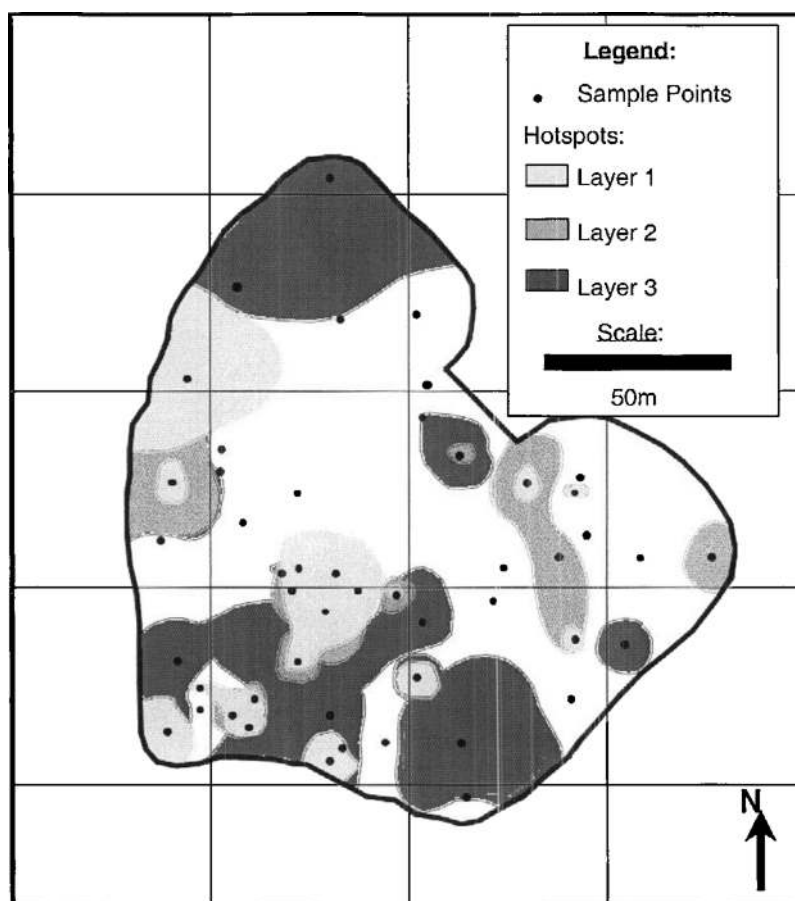


Figure 8. The contamination hotspots located through geostatistical analysis. The information for all three layers has been combined on this map. This is because remediation strategies are likely to require excavation to at least 50 cm or to an uncontaminated natural stratum. Table 1 defines the values used to denote a hotspot for each of the elements. Colour coordinated lines have been used to delineate the extent of overlapping hotspots from different layers.

80% and precision limits of $\pm 20\%$ around the true mean, the site is more than adequately sampled for the majority of elements. This illustrates that fewer samples than were taken would have been required to adequately describe the distribution of contamination and therefore partially negates the impacts of the clustering of sample points. However, given the random and highly variable nature of contamination across the site, maximizing the sample rate appears appropriate.

In terms of classical statistics, the degree of precision between the true mean and the sample mean is likely to be determined by nature of the study in question, combined with the perceived tolerable limits. In terms of the site investigations associated with the remediation of contaminated land it would appear desirable to narrow the precision limits around the true mean value, therefore increasing the sample size. However, using the data from Table 2, to narrow down the difference between the sample mean and the true mean from $\pm 30\%$ to $\pm 10\%$ using a probability of 80%, for layer 3, would require increasing the

Table 2

Number of samples required for each layer to give a sample mean within $\pm 10\%$, $\pm 20\%$ and $\pm 30\%$ of the true mean for a probability of 80%. Av. = an average of all the properties and elements measured

Probability = 80%								
Layer	μ_0	Number of samples						
		As	Hg	Sb	Pb	Cd	Cu	Av.
1	$(1 \pm 0.1)\mu$	37	73	35	32	182	658	113
	$(1 \pm 0.2)\mu$	9	18	9	8	46	165	28
	$(1 \pm 0.3)\mu$	4	8	4	4	20	73	13
2	$(1 \pm 0.1)\mu$	20	98	25	283	117	405	105
	$(1 \pm 0.2)\mu$	5	24	6	71	29	101	26
	$(1 \pm 0.3)\mu$	2	11	3	31	13	45	12
3	$(1 \pm 0.1)\mu$	24	137	41	342	100	306	106
	$(1 \pm 0.2)\mu$	6	34	10	86	25	77	26
	$(1 \pm 0.3)\mu$	3	15	5	38	11	34	12

sample size from 12 to 106 samples. In terms of this study, for an area of 21,759 square meters, this is far higher than the number of samples that would generally be taken during a site investigation. In practice the number of samples taken are likely to be governed by factors such as legislative guidelines, prior knowledge of the site, the likely distribution of contamination and financial considerations.

Conclusions

The most important factor influencing the distribution of trace elements across the study area appears to be its complex industrial evolution. The variety of industrial processes and the chaotic manner in which the waste arising from these processes was disposed of have contributed to the current problems of contamination associated with the site. The random nature of this distribution is reflected in the spiky nature of many of the experimental variograms generated during geostatistical analysis, illustrated by Figures 2 and 4. Nevertheless, the maps produced provide a good visual indication of the levels of contamination across the study area (Figures 3, 5 and 7), thus offering a substantial tool to aid the design and implementation of a suitable and cost effective remediation strategy, or the identification of areas requiring further investigation. Furthermore, this visual interpretation of the maps was supported by the statistical analysis of the data.

Classical statistics showed that for the majority of elements, the area was more than adequately sampled, however many of the maps, particularly those for layer 3 show evidence of bull's eyes (Figure 7). These artefacts can effect the visual interpretation of the data. Furthermore, due to the inaccessibility of parts of the study area, the sample strategy used during site investigation has left a large proportion of the site under-sampled. Even in instances where a more even sample distribution can be achieved, either through the use of systematic or stratified random sampling, sections of the site in-between samples will always remain unsampled. Whilst kriging provides a valuable tool in estimating the levels

of contamination at these unsampled locations it does not provide an answer without some degree of uncertainty and there is always the possibility of a hotspot remaining undetected. In many instances this uncertainty can be at least partially negated through detailed examination of the site history and identification of areas more likely to be contaminated. Sample strategies can then be adopted to map out and trace any potential contamination plumes. Uncertainties can also be reduced through a second stage of site investigation, even using a small number of samples, which can be used to validate any maps produced. In addition, probabilistic modelling can be used to assign degree of uncertainty to the distributions produced.

In cases such as the site examined in this study, the complexity of the distribution of contamination hinders geostatistical analysis meaning that targeted sample strategies are not always possible. Even with a high sample density the rapid changes in contaminant concentrations, illustrated by the tightly packed contours surrounding hotspots on many of the maps (Figures 3, 5 and 7), means that the potential for contaminated areas remaining undetected may be unacceptably high for many potential developers. As a result, remediation strategies such as site wide excavation and evacuation or treatment of all potentially contaminated material may provide the only feasible solution. Additionally, developers are likely to be unwilling to meet the high costs of extensive sampling strategies.

Whilst it has been demonstrated that kriging can offer the potential to map the spatial distribution of contaminants, there are a number of limitations associated with the methodology. Even in instances where probabilistic modelling and a secondary phase of validity sampling are employed, the possibility of an undetected hotspot remains. This potential risk to human health combined with the large number of samples required may preclude the commercial use of geostatistics in the remediation of contaminated land.

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