Estimating Exposure Point Concentrations for Surface Soils for Use in Deterministic and Probabilistic Risk Assessments

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Abstract

When estimating human or ecological risks from exposures to surface soils at terrestrial properties regulated as hazardous waste sites by federal or state agencies, risk assessors must estimate exposure point concentrations (EPCs) for compounds in the surface soils. In this manuscript, we demonstrate nonparametric methods to estimate the EPC for a single compound in surface soils for use in deterministic and/or probabilistic human or ecological risk assessments. Since regulatory agencies instruct risk assessors to consider scenarios involving long-term (chronic or lifetime) average exposures in most human risk assessments (US EPA, 1992, Exposure; Lioy, 1990; US EPA , 1989, HHEM), it is essential to distinguish (i) the number of soil samples taken in the field program, N_S, for which the field geologist has laboratory measurements from (ii) the number of exposure events, N_e ($>>N_S$) over which the exposure is properly averaged. By taking spatial information into account, we demonstrate new methods for computing the upper 95th-percentile of the uncertainty in the mean concentration that overcome the limitations in the method currently recommended by the US Environmental Protection Agency for deterministic human risk assessments. We also extend the new methods to developing EPCs for multiple compounds and to developing second-order distributions for use in probabilistic risk assessments.

1.0 Introduction

When estimating human health risks from exposures to surface soils occurring on a terrestrial property regulated as hazardous waste site by federal or state agencies, risk assessors must estimate exposure point concentrations (EPCs) for one or more compounds in soils by taking both variability and uncertainty into account.

Adopting the concepts now commonly used in risk assessments, we define variability and uncertainty as:

- Variability represents diversity or heterogeneity in a well characterized population. Fundamentally a property of the Nature, variability is usually not reducible through further measurement or study. For example, surface soil samples from different portions of a contaminated property have different concentrations of the contaminant no matter how carefully we measure the samples.

- Uncertainty represents partial ignorance or the lack of perfect information about poorly-characterized phenomena or models. Fundamentally a property of the risk analyst, uncertainty is sometimes reducible through further measurement or study. For example, a risk assessor does not know the concentration of the contaminant in each cubic centimeter of surface soil at the property, even though he or she can certainly take more samples to gain additional (but still imperfect) information about the spatial distribution.

In a fully probabilistic risk assessment, a second-order probability distribution represents the variability and the uncertainty in the exposure concentration(s) experienced by a person or animal. A second-order probability distribution is a parametric distribution for variability with parameters that are distributions for uncertainty (Burmaster & Wilson, 1996). In a deterministic risk assessment, it is necessary to reduce the second-order probability distribution of exposure concentrations to a single point value for each compound. In key guidance documents for deterministic human risk assessments (US EPA, 1989, HHEM; US EPA, 1992, EPC), the US Environmental Protection Agency (US EPA) directs risk assessors to use the one-sided 95th-percentile upper confidence limit (UCL) of the uncertainty in the arithmetic mean concentration for each compound as the exposure point concentration (EPC) for exposures to surface soils. In effect, the Agency

directs risk assessors to compute the EPC (a point value) for a deterministic human risk assessment as the 95th percentile of the uncertainty in the mean of the variability of the second-order random variable representing exposure concentrations. The Agency also recommends a parametric method for estimating this statistic from measured data (US EPA, 1992, EPC; based on: Gilbert, 1987; and Land, 1971 and 1975).

In supplemental guidance for deterministic human health risk assessments (US EPA, 1992, EPC), the Agency states that "The choice of the arithmetic mean concentration as the appropriate measure for estimating exposure derives from the need to estimate an individual's long-term average exposure. Most Agency health criteria are based on the long-term average daily dose, which is simply the sum of all daily doses divided by the total number of days in the averaging period....." (page 2, emphasis added). In this statement, the Agency focuses on the number of exposures (indexed $n_e = 1, 2, \ldots, N_e$) as a measure of variability of chemical concentration at different locations. Later in the same guidance, the Agency states "The 95 percent UCL of a mean is defined as a value that, when calculated repeatedly for randomly drawn subsets of site data, equals or exceeds the true mean 95 percent of the time. ..." (page 3). In this latter statement, the Agency focuses on the concentrations measured for the number of samples (indexed $n_S = 1, 2, \ldots, N_S$) that the field geologist collected during a field program at the property as a measure of uncertainty in the mean concentration. Overall, N_S is a measure of the (small) number of soil samples collected during a field program at the property, while N_e is a measure of the (large) number of exposures that an person has with a property over the long-term use of the property. For typical field programs and for long-term exposures, $N_S << N_e$.

In this manuscript, we accept the US EPA's policy for deterministic human risk assessments, i.e., using the one-sided 95th-percentile upper confidence limit (UCL) on the uncertainty in the mean concentration as the exposure point concentration. However, we note that the method recommended by US EPA for computing this statistic has severe limitations because it ignores all information about spatial patterns. In our experience doing human risk assessments for hundreds of properties regulated as hazardous waste sites by US EPA and/or similar state agencies, the parametric method recommended by US EPA overstates the EPC (as defined by the Agency) by failing to take spatial information into account.

In the first part of this manuscript, we contrast the limitations of the parametric methods recommended by US EPA with the strengths of alternative nonparametric methods to estimate the EPC for a single compound in surface soils through gedanken experiments at a hypothetical site where the Acme Company once manufactured widgets. Through its production and waste disposal practices, Acme Company released some hazardous substances to the surface soils on the property, but one compound -- compound X -- is by far the most prevalent and the most toxic in surface soils now that the company has ceased operation. Later in this manuscript, we discuss how to include measurements for the other compounds in the calculations.

2.0 A Hypothetical Case Study

Let us consider this hypothetical situation. Acme Company owned one property, rectangular in shape, measuring 20 u (units) in the east-west direction (along the abscissa) by 10 u in the north-south direction (along the ordinate). We have one set of measurements that shows the concentration of X (denoted [X], mg/kg) at $N_S = 17$ locations chosen by the field geologist. Using judgmental sampling common at such sites, the field geologist chose these 17 locations based on the site history and on staining observed in the surface soils. As is common at such properties, the field geologist deliberately sampled the center of the two known "hotspots" and oversampled adjacent areas without using stratified random sampling (Keeping, 1995) or adaptive sampling (Thompson & Seber, 1996).

In the first gedanken experiment (GE1) for deterministic human risk assessments, we adopt the same assumption implicit in the method recommended by US EPA -- that each person who will use the property in the future will access all subareas of the property with equal probability. In other words, each person uses the property so as to have a uniform probability of exposure over the whole property. In the second gedanken experiment (GE2) for probabilistic risk assessments, we show how to relax this assumption to let each person have unequal and nonuniform exposures to soils on the property.

Table 1 lists the coordinates and the chemical concentrations reported by the laboratory for the 17 soil samples. The arithmetic mean (AMean) of the concentration in these samples equals 64.2 mg/kg. The map in Figure 1A shows the concentrations at the 17

sampling the locations on the property. The field geologist took the first and third samples (250.2 and 294.1 mg/kg, respectively) at the two known "hotspots."

In Figures 1A and 1B, we use Shepard's functions (Schwab, 1996) to interpolate and visualize the general shape of the contours and concentrations of the compound X in the surface soils. [EndNote 1]. Shepard's functions are a well studied and powerful nonparametric way to analyze, interpolate, and plot data with two or three spatial dimensions (Gordon & Wixon, 1978; Franke, 1982; Renka, 1988a, b, c). Figure 1A shows the estimated contours for 25, 100, 150, and 200 mg/kg, while Figure 1B shows the shape of the spatial distribution in perspective. By dividing the numerically integrated volume under the Shepard's function by the area of the property (200 u^2), we estimate the spatially-averaged mean concentration of X equals 29.6 mg/kg.

3.0 The First Gedanken Experiment for Deterministic Risk Assessments

In this section, we estimate the EPC using the method recommended by US EPA and by two alternative methods that take the spatial pattern of contamination into account.

3.1 Using Land's Method to Estimate the EPC for [X]

Figures 2A and 2B show Normal and the LogNormal probability plots (Burmaster & Hull, 1996), respectively, for the 17 data points. Given that the 17 points more closely follow a straight line in the lower plot (Ott, 1990), we use the parametric method recommended by US EPA (US EPA, 1992, EPC; Gilbert, 1987; Land, 1971 and 1975) for LogNormal distributions to estimate the EPC for deterministic human health risk assessments as the one-sided upper 95th percentile of the uncertainty in the mean concentration:

$$UCL = \exp\left[\overline{y} + 0.5 \bullet s^2 + \frac{s_y \bullet H}{\sqrt{N_S - 1}} \right] \qquad \text{Eqn 1}$$

where exp[•] is the exponential function, $\overline{y}$ is the arithmetic mean of the natural logarithms of the data, s_y is the standard deviation of the natural logarithms of the data, N_S is the number of soil samples, and H is interpolated from tables (see Gilbert, 1987). This method excludes all information about the spatial pattern of contamination and it implicitly assumes that each person has uniform exposure to all areas of the property. For the data in Table 1, the EPC for a deterministic human risk assessment (taken as

the upper 95th percentile UCL of the uncertainty in the mean concentration) = 2,465. mg/kg, a value approximately an order of magnitude higher than the highest measurement made at a known "hotspot" on the property. [EndNote 2].

3.2 Using the Bootstrap Method to Estimate the EPC for [X]

Statisticians now use the "Bootstrap Method" as a robust, nonparametric method for analyses based on a small number of assumptions (Efron & Tibshirani, 1993, 1991; Efron, 1988). Using the Bootstrap Method, an analyst re-samples (with replacement) the original data to make inferences about the underlying distribution. Using the standard technique, we drew N_b = 1,000 Bootstrap samples (each of size N_S, with replacement) to estimate the full distribution of the uncertainty in the arithmetic mean concentration of the data in Table 1. This Bootstrap method also excludes all information about the spatial pattern of contamination and it implicitly assumes that each person has uniform exposure to all areas of the property. When applied to zero-dimensional data (i.e., with zero spatial coordinates), the ordinary Bootstrap method changes the relative weights of the individual measurements by integers as follows: for each measurement with its weight decremented to zero, another has its weight increased by one. In effect, the basic Bootstrap re-samples the data (with replacement) to create N_b samples of size N_S each.

Figure 3A shows the estimated PDF for the uncertainty in the mean concentration. [EndNote 3]. Table 2 summarizes the distribution of the uncertainty in the mean from these 1,000 Bootstrap samples. As estimated by this method, the EPC for a deterministic human risk assessment (taken as the upper 95th percentile UCL of the uncertainty in the mean concentration) = 99.2 mg/kg.

3.3 Using Voronoi Diagrams to Estimate the EPC for [X]

Over the last 300 years, many mathematicians, scientists, and engineers have independently invented Voronoi diagrams (often called Thiessen polygons) as the nonparametric method of spatial analysis based on one simple assumption. As paraphrased from a definitive treatise, a planar ordinary Voronoi diagram associates each point in a (bounded) plane with the closest neighbor for which a measurement is available (Okabe et al, 1992, p. 66). Applied to the data in Table 1, this procedure results in a tesselation of the plane into the set of 17 polygons (Figure 4), each one

surrounding one of the 17 sampling locations (Martin, 1996). In effect, we assume (conservatively) that the concentration at any point at coordinates {x,y} has the same concentration as the closest of the N_S = 17 points where samples were measured.

With this minimalist assumption about the two-dimensional spatial distribution of the contamination, and with the continuing assumption that each person has his or her exposures distributed uniformly across the who property, we see that the spatial average concentration is a weighted average of the 17 measurements, with each weight equal to the area of its associated polygon as a fraction of the area of the bounding rectangle. For the data in Table 1, this weighted average concentration = 36.5 mg/kg. With the Voronoi diagram, the weight of the i^{th} observation is A_i / A_T, where A_i is the area of the area of the i^{th} polygon and A_T is the total area (= sum of the A_i).

Next, we drew N_b = 1,000 Bootstrap samples (each of size N_S, with replacement) to estimate the full distribution of the uncertainty in the area-weighted arithmetic mean concentration. When applied to two-dimensional data (i.e., with two spatial coordinates), the Bootstrap method changes the relative weights of the individual measurements by nonintegers as follows: for each sample with its weight decreased to zero, each of one or more measurements has its weight increased through an expansion of the subareas associated with it. In this manuscript, the algorithm passes only the unique measurements in the Bootstrap sample to the Voronoi diagram.

Figure 3B shows the estimated PDF for the uncertainty in this area-weighted mean concentration, and Table 2 summarizes the distribution of the uncertainty in the area-weighted mean of these 1,000 Bootstrap samples. As estimated by this method, the EPC for a deterministic human risk assessment (taken as the upper 95th percentile UCL of the uncertainty in the mean concentration) = 70.4 mg/kg.

3.4 Using "Extruded" Voronoi Diagrams to Estimate the EPC for [X]

To see a three-dimensional plot of the data in Table 1, we "extrude" each polygon in Figure 4 to a height corresponding to its associated concentration as shown in Figure 5. This surface in Figure 5 is represented by the formula in Eqn 2 (Eberhard Lange, 1995).

$$C[x, y] = \frac{\sum_{n_s=1}^{N_s} C[x_i, y_i] \bullet W_i[x, y]}{\sum_{n_s=1}^{N_s} W_i[x, y]} \qquad \text{Eqn 2}$$

Eqn 2 is an "interpolation radial basis function" which estimates the concentration of compound X at any point {x, y} using a spatially-weighted sum (with normalized weights that sum to one everywhere). In Figure 5, each of the extruded areas has a cross section identical in size and shape to its corresponding polygon in Figure 4.

Throughout this manuscript, we use the weighting function, $W_i[x, y] = \exp[- d^2 \bullet ((x - x_i)^2 + (y - y_i)^2)]$, a function based on Euclidean distance. Lange (1995) also suggests using other weighting functions commonly used in geostatistics (Isaaks & Srivastava, 1989). The parameter d adjusts the "stiffness" of the interpolant (Cressie, 1993).

This function in Eqn 2 (with weights based on Euclidean distance) has many useful properties. When d equals zero, all spatial information is lost, i.e., Eqn 2 equals the simple arithmetic mean of the N_S measurements. As $| d |$ tends to ∞ , $C [x, y]$ takes the value of the nearest measured sample, just as a Voronoi diagram does. When $0 < | d | < \infty$, $C [x, y]$ smoothly interpolates the concentrations between the measured locations. Figure 5A shows the interpolation radial basis function in Eqn 2 (with d = 2) for the data in Table 1. We used numerical integration to find the area-weighted arithmetic mean of this function for the 17 samples; it equals 36.5 mg/kg, the same value found using the Voronoi diagram above.

Next, we drew N_b = 1,000 Bootstrap samples (each of size N_S, with replacement) to estimate the full distribution of the uncertainty in the arithmetic mean concentration. Figure 3C shows the estimated PDF for the uncertainty in this statistic, and Table 2 summarizes the distribution of the uncertainty in the mean of these 1,000 Bootstrap samples. Overall, this method using "extruded" Voronoi diagrams is mathematically equivalent to the previous method using ordinary Voronoi diagrams. Thus, as expected, the EPC using this method for a deterministic human risk assessment (taken as the

upper 95th percentile UCL of the uncertainty in the mean concentration) = 69.8 mg/kg (a value within the random sampling error of the previous result, 70.4 mg/kg).

4.0 The Second Gedanken Experiment for Probabilistic Risk Assessments

Here we use the data set in Table 1, but we change the assumption on how the property will be used in the future by considering an ecological example with a species of small mammals, perhaps rabbits or groundhogs. (While we use an ecological example in this section, the methods are equally applicable to human health risk assessments.) We now assume (i) that each animal on the property in the future will have chronic exposures to different subareas of the property and (ii) that the chronic exposures are both uneven (in space) and unequal (between animals). In effect, we develop a second-order random variable containing both variability (representing the different animals in the population using the property) and uncertainty (representing our lack of perfect information on the spatial pattern of contamination) (Burmaster & Wilson, 1996).

Since there are an infinite number of ways that animals can have unequal and uneven exposures to subareas of a property, we demonstrate the type of calculation with a specific example. To give a specific example to illustrate the method, we assume that each or animal has chronic exposure to the soils in a particular subarea, say a square, that is smaller than and wholly contained within the rectangular property. Figure 6A shows a set of 40 random squares placed randomly inside the rectangular property. In this example, (i) the x- and y-coordinates for the center of each square are randomly drawn from independent Uniform distributions, Uniform [min, max] = Uniform [2, 18] and Uniform [2, 8], respectively, and (ii) the half-length of the side of each square is independently drawn from a Triangular distribution, $\underline{b}$ ~ Triangular [min, mode, max] = Triangular [0.5, 1.6, 2] (Evans et al, 1993). (Of course, it is possible to specify other assumptions, including ones for nonsquare subareas and/or for nonuniform and/or nonindependent distributions.) We further assume that each animal behaves in such a way that it is exposed to soils nonuniformly inside a square, i.e., that each animal has a weighted average chronic exposure. In particular, we use the normalized bivariate density shown in Figure 6B to represent the spatial weighting function inside each square.

With these assumptions, we start the calculations. First, we estimate a nonparametric first-order random variable representing the variability in the chronic exposures

experienced by different animals conditional on a known spatial distribution of concentration. In the language of second-order random variables, this is the "inner loop" for variability (Burmaster & Wilson, 1996). For a particular (random) animal in the population, the computer simulates a random square, computes the weighted average concentration inside that square conditional on the known spatial distribution of contamination, and reports a value.

$$ACE_1 = \int_{x=0}^{20} \int_{y=0}^{10} C[x, y] \bullet K[x, y] \, dx \, dy \qquad \text{Eqn 3}$$

where ACE_1 denotes one animal's average chronic exposure concentration, C [x, y] comes from Eqn 2, and K [x, y] denotes kernel, i.e., a bivariate probability density function representing the person's intensity of exposure over its square. In this example, we use the kernel shown in Figure 6B and Eqn 4: [EndNote 4]

$$K[x, y] = \frac{\pi^2}{16 \bullet b^2} \bullet \text{Cos}\left[\left(\frac{\pi}{2 \bullet b}\right) \bullet (x - x_{cen})\right] \bullet \text{Cos}\left[\left(\frac{\pi}{2 \bullet b}\right) \bullet (y - y_{cen})\right] ;$$

$$x_{cen} - b \le x \le x_{cen} + b$$
$$y_{cen} - b \le y \le y_{cen} + b$$

$$= 0 ; \quad \text{elsewhere} \qquad \text{Eqn 4}$$

where the square subarea has the point (x_{cen}, y_{cen}) as its center and the length b as its half-width and Cos [•] is the cosine function.

This calculation is repeated hundreds of times to simulate the nonparametric distribution of the variability in different animals' average chronic exposures conditional on the known spatial distribution of contamination. The solid line in Figure 7 shows the estimated PDF for this first-order random variable using C [x, y] from Eqn 2 and the 17 data points in Table 1. Here, $N_{innerloop}$ = 1,000.

Next, we include the nonparametric uncertainty in the spatial distribution of contamination by adding an "outer loop" to the algorithm. In particular, we use the Bootstrap method to pass random subsets of the 17 data points to the "extruded" Voronoi surface (Eqn 2 with weights based on Euclidean distance). For each iteration of

this outer loop, the computer again makes $N_{innerloop}$ calculations for variability among people (or animals). The dashed line in Figure 7 shows the estimated PDF for variability among people for one Bootstrap sample from the data. The solid and dashed lines in Figure 7 are the first three realizations of a nonparametric second-order random variable for the variability and the uncertainty in exposures, given the set of $N_S = 17$ measurements and the characteristics of the exposed population.

With more computation, we can add more realizations of this second-order random variable to Figure 7. We see this nested-loop algorithm as a second-order random number generator for variable and uncertain exposures by animals using the property. Using methods from computer science to speed the method, its computational burden is manageable for either a human or ecological risk assessment.

5.0 Computations for Multiple Compounds

When multiple chemicals are present in each of the soil samples from a property, a risk assessor must preserve the (spatial) correlation when computing the EPCs (point values) for deterministic risk assessments or estimating second-order probability distributions for probabilistic risk assessments. None of the methods discussed so far preserve this essential information.

Here, we offer a solution to this problem based on an extension of the methods developed to find the "toxic equivalents" for some groups of compounds, i.e., polycyclic aromatic hydrocarbons (PAHs) and chlorinated dioxins and furans (see also, Ginevan & Splitstone, 1995). Let $n_S = 1, 2, ..., N_S$ index the different locations where the field geologist took a soil sample for chemical analysis, and let $n_C = 1, 2, ..., N_C$ index the different compounds. We form the matrix **X** with N_S rows and N_C columns. Each element in the matrix **X** is the chemical concentration measured at the n_S-th location for the n_C-th compound. Next, we form the column vector **T** with N_C rows. Each element in column vector **T** is the toxicity constant for the n_C-th compound (as discussed in the next paragraph). With these definitions, we use matrix algebra (Strang, 1988) to compute the column vector **Ξ** with N_S rows.

$$\mathbf{\Xi} = \mathbf{X} \bullet \mathbf{T} \qquad \text{Eqn 5}$$

Each element of the column vector **Ξ** is the toxicity-weighted sum of the concentrations the n_S-th location.

In practice, the risk assessor may need to form two (or more) different column vectors for **T** and then compute two (or more) different column vectors for **Ξ**. For example, if the risk assessor desires to compute the total incremental lifetime cancer risk and assumes the additivity of risk, a first column vector, $\mathbf{T}_{carc}$, might hold the Cancer Slope Factor (CSF) for each compound regulated as a carcinogen. This column vector would have a zero in the position for each compound without a CSF. If the risk assessor desires to compute the total hazard index and assumes additivity over noncancer endpoints , a second column vector, $\mathbf{T}_{noncarc}$, might hold the inverse Reference Dose (RfD^{-1}) for each compound regulated as a noncarcinogen. This column vector would have a zero in the position for each compound without an RfD. As appropriate, the risk assessor can modify these ideas to compute separate sums over the neurotoxicants, the reproductive toxicants, or other classes or subsets of compounds.

With these definitions, the risk assessor may now use nonparametric (or parametric) methods to compute the EPC for deterministic risk assessments or the second-order probability distribution for probabilistic risk assessments. As long as the risk assessor makes corresponding changes in the calculations in other portions of the risk assessment, the use of one or two toxicity-weighted sums preserves the crucial spatial correlations and also speeds the overall computations.

6.0 Discussion and Conclusions

Based on the work of Chen (1995), the US EPA has recently published a new guidance manual (US EPA, 1996, SSG) that discusses some of the limitations of the Land method, i.e., the parametric method that the Agency currently recommends for calculating EPCs for soils for deterministic risk assessments (US EPA, 1992, EPC). However, the problems with the Land method are profound. First, the distributions are rarely LogNormal in practice, a prerequisite for the Land method. Second, Schmoyer et al (1996) discuss certain fundamental statistical difficulties associated with estimating and testing the mean of a LogNormal distribution, the distribution used in the Land method. Third, as Ginevan and co-authors (Ginevan & Putzrath, 1994; Ginevan & Splitstone, 1995) have emphasized, it is crucial not to destroy the spatial information inherent soil measurements taken at known locations. Finally, when several

contaminants co-occur in soil samples with different spatial patterns, it is essential to preserve the spatial patterns and correlations among the concentrations of the contaminants.

Based on the results in this manuscript, we recommend several methods that overcome the limitations of the Land method (and any other methods, including the Chen method and the methods in Armstrong (1992), that destroy the spatial information). For deterministic human risk assessments involving exposures to soils, i.e. with spatial coordinates, we recommend the use of the Bootstrap method with Voronoi diagrams to estimate the EPC as the one-sided 95th-percentile upper confidence limit (UCL) of the uncertainty in the arithmetic mean concentration. [For deterministic risk assessments involving zero-dimensional (nonspatial) data, we recommend the ordinary Bootstrap method to estimate, when necessary, the one-sided 95th-percentile upper confidence limit (UCL) of the uncertainty in the arithmetic mean concentration.] For probabilistic human or ecological risk assessments, we recommend the use of Boostrap method, random kernels, and an interpolation function like Eqn 2 (perhaps with different distance metrics) to develop second-order random variables for exposure concentrations. Third, when multiple compounds are present in either deterministic or probabilistic human or ecological risk assessments, we recommend the method in Section 5 as a way to preserve the essential spatial patterns and spatial correlations inherent in the soil data.

Many of the methods in this manuscript rest fundamentally on the Voronoi diagram, a method used in different branches of science and engineering for over 300 years. In particular, we rely on surfaces interpolated using "extruded" Voronoi diagrams (as represented by Eqn 2 with weights based on Euclidean distances). At the same time that we recommend this method for general use, we realize that there are many other functions for interpolating surfaces (for example, the Shepard's functions demonstrated in Figure 1B and the many methods commonly used in geostatistics (Isaaks & Srivastava, 1989; Cressie, 1993)). Thus, even though Voronoi methods require minimal assumptions and have stood the test of time in other disciplines, we make no claim that Voronoi methods are either unique or optimal for this or other situations. In fact, we see the choice of the interpolant as an area ripe for continued research.

The methods recommended here have higher computational burdens than the Land or Chen methods. In an age when desktop computers clocked faster than 200-MHz sell for a few thousand dollars, the computational burdens are acceptable as compared to other

steps in either a deterministic or probabilistic risk assessment, and the computational burdens are less than those for even more powerful methods based on random walk models (Gaylord & Nishidate, 1996), fractal Browian motion (Maeder, 1996), or Lévy flights (Viswanathan et al, 1996). At the cost of the extra computational burden, the methods recommended here overcome the severe limitations of the Land and Chen methods (and other nonspatial methods). First, they are inherently robust methods since they are based on the nonparametric Bootstrap method. Second, they are inherently spatial methods because they are based on the simplest yet conservative assumption from geostatistics. Third, when used with the Method in Section 5, they inherently preserve the spatial correlations in the data.

While none of the methods is easily inverted, a risk assessor can use any of the methods to develop a cleanup target for the remediation of soils in iterative calculations. If the "baseline" risk assessment yields an unacceptable risk, then the risk assessor and remediation engineer can hypothesize the treatment or removal of soils in certain portions of the property -- and then re-run the risk assessment. Paul Anderson (1996) calls this approach the "pick-up" method for determining the extent of site remediation. The risk assessor and the remediation engineer can work together to find the combination of treatment and removal of soils that achieves the stated health goal at optimal cost. When used in this fashion, the methods recommended here extend work by Bowers et al (1996).

This approach should provide further insights for risk managers that may question the value of additional sampling (e.g., Sedman et al., 1992). First, by defining the average exposure concentration spatially, the method considers the uncertainty that arises from small sample sizes in terms of its effects on the estimated risk for an exposed individual, and not simply the uncertainty that arises from the inherent spatial variability of contaminants at the site. Second, by showing the portion of the site area that each measurement is assumed to represent, this method provides a graphical means for identifying those sampling locations that may be over or under represented. For example, if there is a very long distance between a "hot spot" sampling location and one at the site boundary, then the area assumed to be represented by the concentration of the "hot spot" may be much larger than the actual area of the "hot spot" identified by site experts, and consequently sampling between these two points could substantially effect the estimated average. [EndNote 5]. Third, the method provides an opportunity for analysts to speculate about how much the average and uncertainty about it might

change if additional samples are collected. Given our assumption that most sites are sampled so as to ascertain the magnitude of the contamination in suspicious areas, we expect that in most cases collecting additional samples will reduce the value of the 95th-percentile UCL of the average both by reducing the average and by increasing the sample size, although the amount of reduction may or may not be significant. Finally, by providing a means to generate probabilistic distributions for EPCs, this method facilitates probabilistic risk assessment, quantitative uncertainty analysis, and value of information (VOI) analysis (see Thompson and Graham [1996] for an description of these). Using a formal VOI approach, the risk manager can determine whether the benefits of having the information and the reduction in uncertainty gained by additional sampling justify the costs of sampling (although the stakes of the decision should be large enough to justify the additional analytical burden of performing the analysis). Informally, the risk manager could begin by considering whether a change in the estimated EPC would be likely to change the remediation decision, how much the decision might change, and how much it costs to obtain the information. If the expected change is significant, then additional sampling may be worthwhile if it is of reasonably high quality and low cost.

The methods recommended here can also be extended to handle data sets that include concentrations reported as "nondetects." For example, the methods recommended here work well when the risk assessor assigns a concentration of one-half the detection limit for samples reported by the laboratory as "nondetect."

Finally, the methods recommended here are conservative in the sense they protect public health and ecological integrity. The random process (Bras & Rodriguez-Iturbe, 1993) used to create the data set has a mean concentration of 25 mg/kg over the whole rectangle. [EndNote 6]. The results here are protective of public health precisely because (i) they use the 95th percentile of the uncertainty in the mean concentration for deterministic human risk assessments, (ii) they include both the variability and the uncertainty in the second-order random variable for probabilistic risk assessments, (iii) they preserve essential information on the spatial correlations among concentrations, and (iv) the field geologist used judgmental sampling based on the site history to sample preferentially the "hotspots" and "warmspots" on the property.

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Dedication

We dedicate this manuscript to George B. Thomas, Jr.

Trademarks

EndNotes

1. Risk assessors rarely encounter situations that have enough data to support the use of kriging or other more advanced geostatistical methods (Cressie, 1993; Isaaks & Srivastava, 1989). In particular, kriging requires much more data and it requires the data to meet stringent assumptions for the variogram(s) (Isaaks & Srivastava, 1989; Cressie, 1993).

2. In such a circumstance, the US EPA's policy for human risk assessments directs the analyst to use the maximum concentration on the property as the EPC for all locations on the property.

3. We use a nonparametric method from Silverman (1986) with a Gaussian kernel.

4. It is easy to implement other kernels K[x, y] in Eqn 2 once the exposure patterns are measured or modeled (e.g., Freshman & Menzie, 1996).

5. In contrast, a strict arithmetic average of n samples implicitly attributes one nth of the site area to the "hot spot" and this amount decreases as additional samples are collected.

6. Let $g(\mu_x, \sigma_x, \mu_y, \sigma_y, \rho) = \frac{1}{2 \cdot \pi \cdot \sigma_x \cdot \sigma_y \cdot \sqrt{1-\rho^2}} \cdot$

$$\exp\left[-\frac{1}{2 \cdot (1-\rho^2)} \cdot \left\{ \left(\frac{x-\mu_x}{\sigma_x}\right)^2 - 2 \cdot \rho \cdot \left(\frac{x-\mu_x}{\sigma_x}\right)\left(\frac{y-\mu_y}{\sigma_y}\right) + \left(\frac{y-\mu_y}{\sigma_y}\right)^2 \right\} \right]$$

then, $h(x, y) =$

```
  400 g( 5, 0.8, 5, 0.8, +0.6) +
  600 g( 5, 1.0, 6, 1.0, -0.6) +
 2000 g( 9, 1.8, 4, 0.9, -0.7) +
 1000 g(11, 2.5, 5, 0.6, -0.1) +
 1000 g(13, 2.1, 6, 0.6, -0.2)
```


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Table 1
Concentrations of Compound X in Soils

index	X-Coordinate (u)	Y-Coordinate (u)	Soil Concentration (mg/kg)
•••••	•••••	•••••	•••••
1	5.4	5.4	250.2
2	8.6	5.8	51.3
3	8.8	4.2	294.1
4	17.5	7.5	0.1
5	8.8	2.9	52.3
6	2.7	2.7	0.7
7	2.5	7.5	5.3
8	6.4	4.7	150.9
9	5.1	6.6	95.9
10	12.8	6.6	79.6
11	15.7	4.9	33.7
12	3.9	3.7	29.5
13	7.5	6.6	7.5
14	10.9	1.8	9.1
15	14.6	1.4	1.3
16	12.6	3.9	25.4
Ns = 17	6.2	3.2	3.9

Table 2
Empirical Cumulative Distribution Functions (CDF)

Method	Number of Chemical Samples (Ns)	Number of Boostrap Samples (Nb)	AMean (mg/kg)	Minimum (mg/kg)	Perc 01 (mg/kg)	Perc 05 (mg/kg)	Perc 10 (mg/kg)	Perc 25 (mg/kg)	Perc 50 (mg/kg)	Perc 75 (mg/kg)	Perc 90 (mg/kg)	Perc 95 (mg/kg)	Perc 99 (mg/kg)	Maximum (mg/kg)
GE1, for Deterministic Risk Assessments														
Land/US EPA	17	0										2465.1		
Simple Mean	17	1,000	63.9	14.4	22.5	33.5	37.8	49.7	62.5	76.6	91.7	99.2	113.1	129.0
Voronoi	17	1,000	46.3	14.9	19.3	26.2	30.7	36.6	44.5	54.1	64.8	70.4	86.5	109.4
Interpolant	17	1,000	45.5	12.8	20.8	25.7	29.6	36.0	43.7	53.3	63.8	69.8	88.4	108.3
GE2, for Probabilistic Risk Assessments														
First Run	17	1,000	59.5	0.1	0.1	5.3	9.6	20.6	40.2	79.4	139.7	177.0	235.4	282.5
Second Run	17	1,000	64.1	0.1	0.1	2.0	6.9	22.3	37.4	79.6	163.3	209.5	278.7	294.1
Third Run	17	1,000	59.7	0.1	0.0	1.3	4.7	17.9	35.9	79.6	148.9	201.5	272.8	294.1

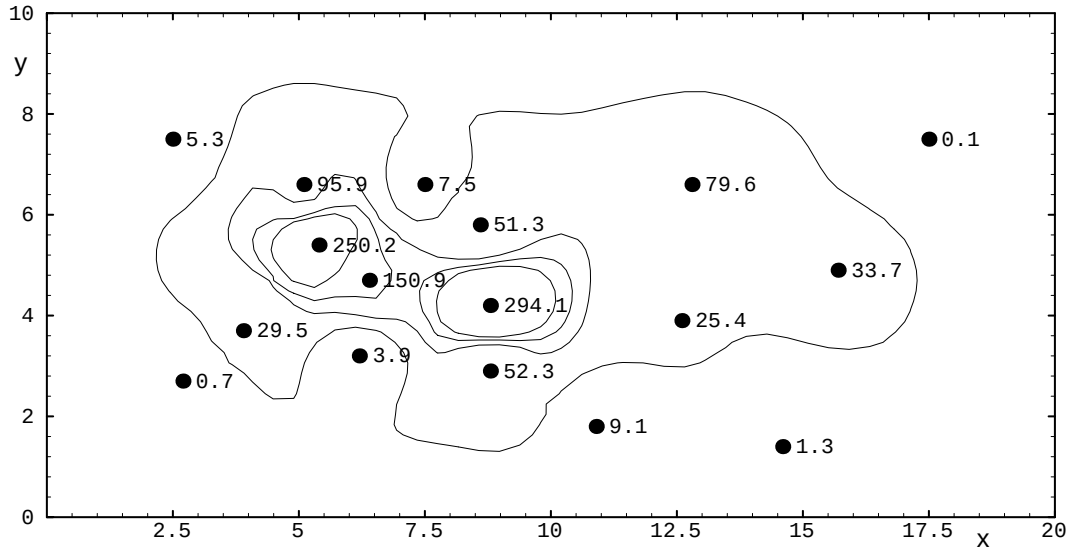

Figure 1A
Sample Locations and
Estimated Concentration Contours

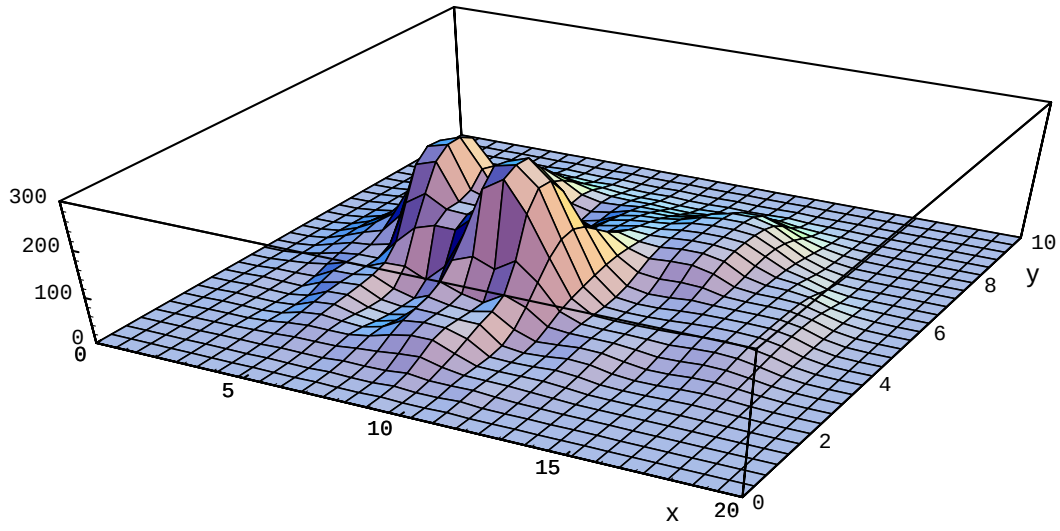

Figure 1B
Concentrations Estimated by
Shepard's Functions

Mma = 500, 80 perc

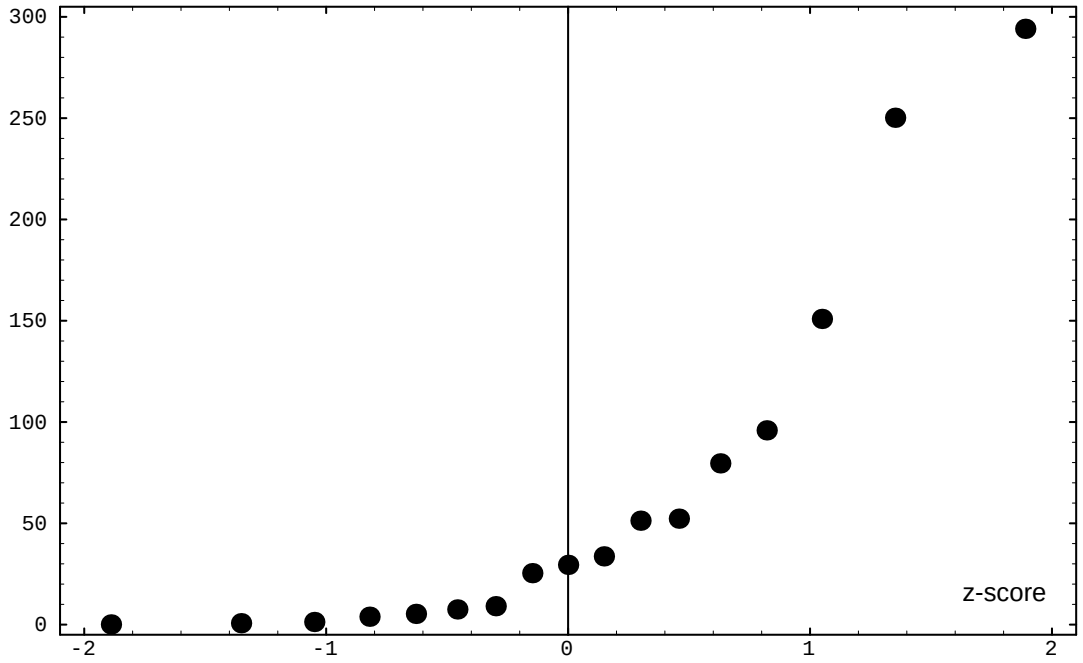

Figure 2A
Normal Probability Plot, N_S = 17

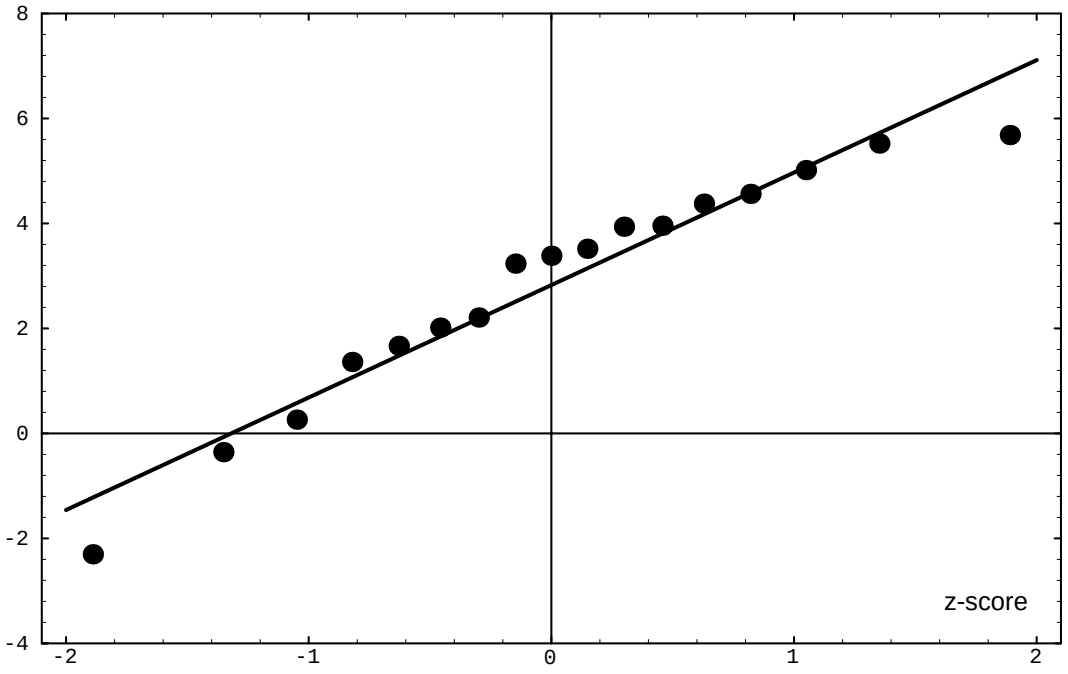

Figure 2B
LogNormal Probability Plot, N_S = 17

Mma = 500, 80 perc

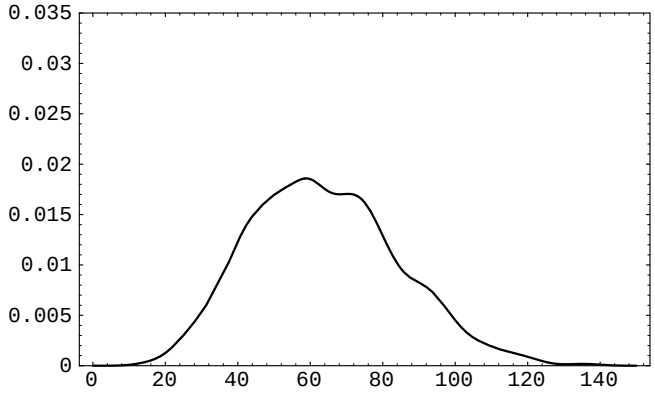

Figure 3A
PDF for Uncertainty
in the
Mean Concentration
Estimated using
Ordinary Bootstrap,
N_S = 17 and N_b = 1,000

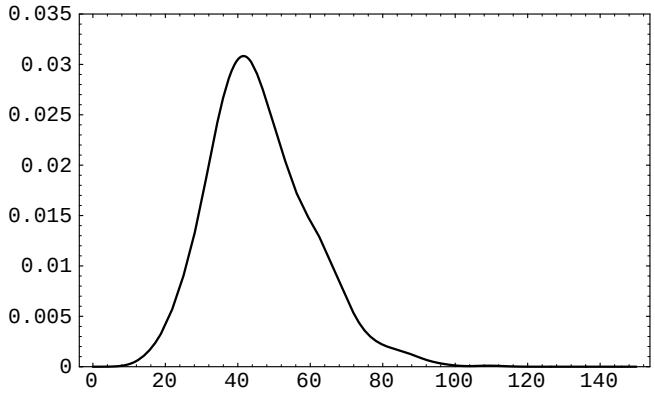

Figure 3B
PDF for Uncertainty
in the Area-Weighted
Mean Concentration
Estimated using
Voronoi Bootstrap,
N_S = 17 and N_b = 1,000

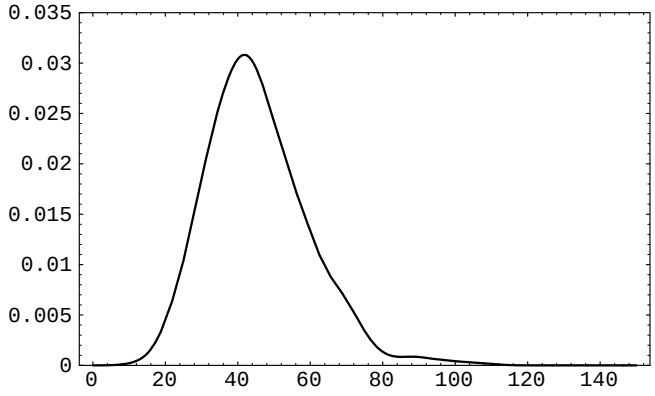

Figure 3C
PDF for Uncertainty
in the Area-Weighted
Mean Concentration
Estimated using
RBF Bootstrap,
N_S = 17 and N_b = 1,000

Mma = 300, 80 perc

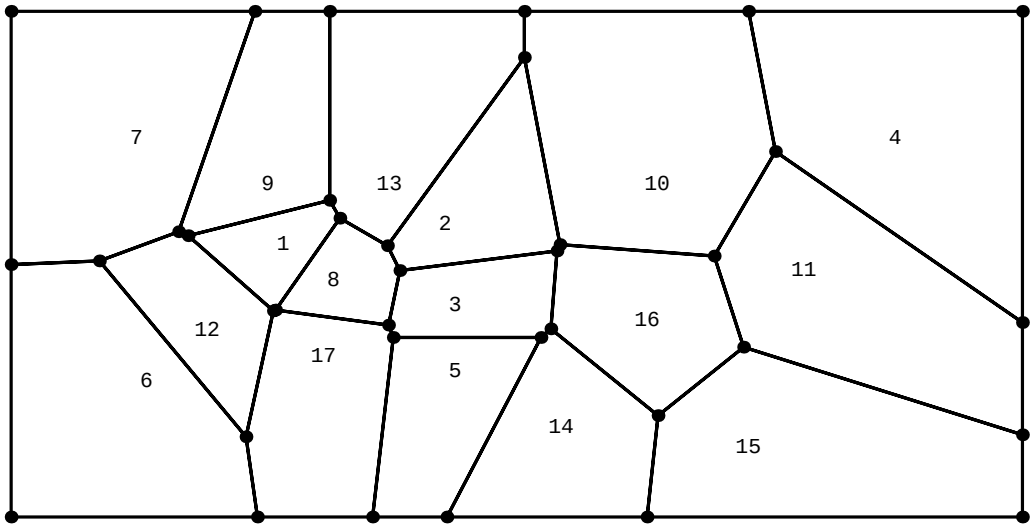

Figure 4
Voronoi Diagram for Samples, N_S = 17

Mma = 500, 80 perc

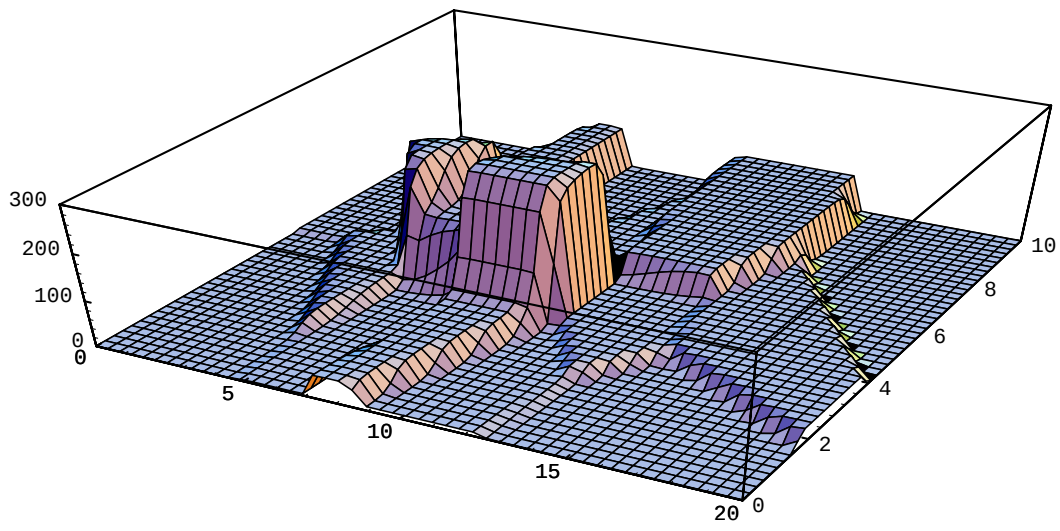

Figure 5
"Extruded" Voronoi Diagram
Using Interpolation Radial Basis Function, N_S = 17

Figure 5B

Mma = 500, 80 perc

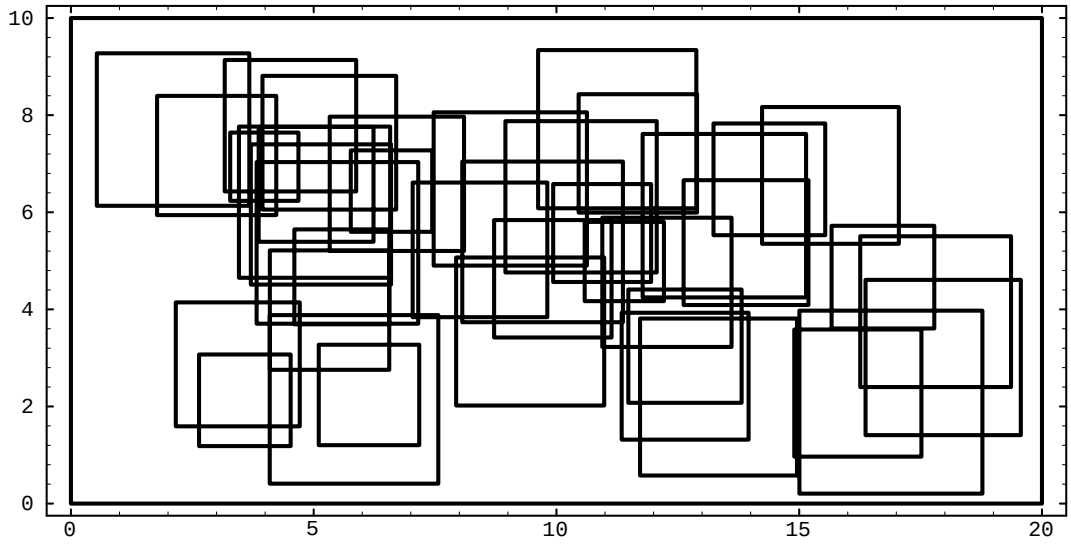

Figure 6A
40 Random Squares (See Text)

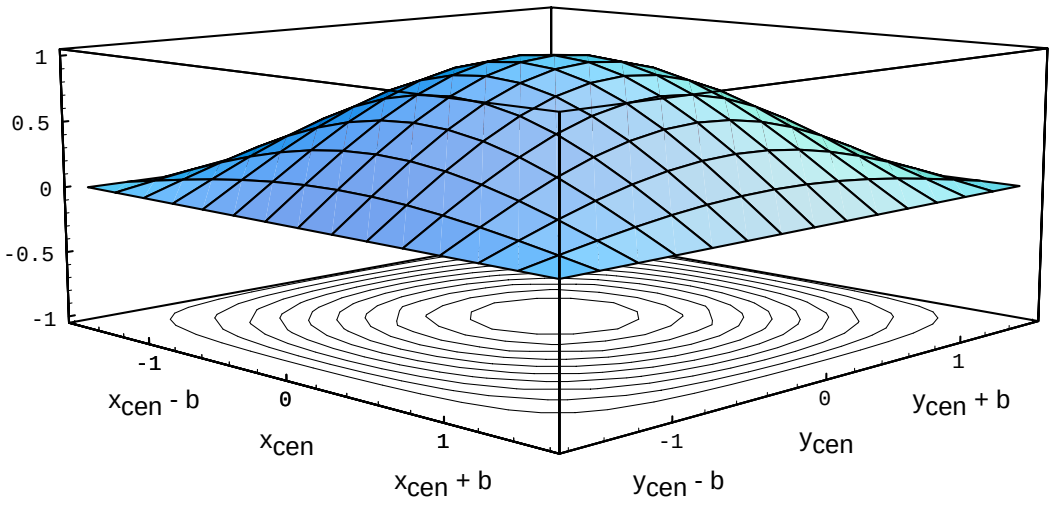

Figure 6B
The Kernel in Eqn 4

Mma = 500, 80 perc

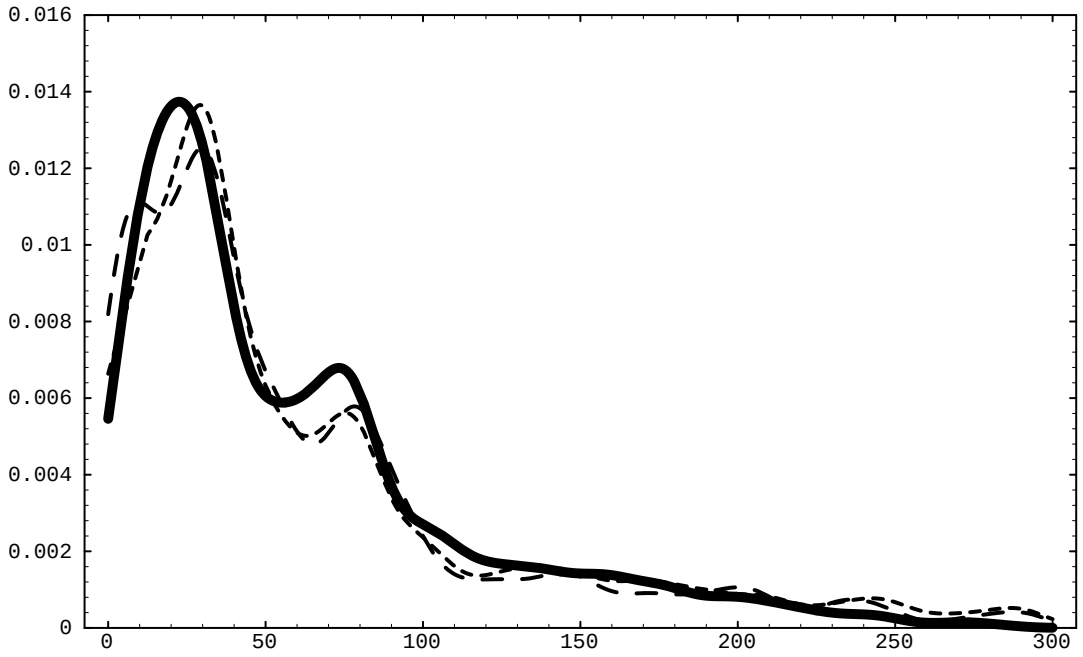

Figure 7
Three Realizations of a
Second-Order Random Variable

Mma = 500, 80 perc