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Spatially Resolved Hazard and Exposure Assessments: An Example of Lead in Soil at Lavrion, Greece

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Spatially resolved hazard assessment (SRHA) and spatially resolved exposure assessment (SREA) are methodologies that have been devised for assessing child exposure to soil containing environmental pollutants. These are based on either a quantitative or a semiquantitative approach. The feasibility of the methodologies has been demonstrated in a study assessing child exposure to Pb accessible in soil at the town of Lavrion in Greece. Using a quantitative approach, both measured and kriged concentrations of Pb in soil are compared with an "established" statutory threshold value. The probabilistic approach gives a refined classification of the contaminated land, since it takes into consideration the uncertainty in both the actual measurement and estimated kriged values. Two exposure assessment models (i.e., IEUBK and HESP) are used as the basis of the quantitative SREA methodologies. The significant correlation between the blood-Pb predictions, using the IEUBK model, and measured concentrations provides a partial validation of the method, because it allows for the uncertainty in the measurements and the lack of some site-specific measurements. The semiquantitative applications of SRHA and SREA incorporate both qualitative information (e.g., land use and dustiness of waste) and quantitative information (e.g., distance from wastes and distance from industry). The significant correlation between the results of these assessments and the measured blood-Pb levels confirms the robust nature of this approach. Successful application of these methodologies could reduce the cost of the assessment and allow areas to be prioritized for further investigation, remediation, or risk management.

Key Words: **exposure assessment; hazard; exposure; lead; metals.**

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INTRODUCTION

Hazard assessment (HA) and exposure assessment (EA) for environmental contaminants are two fundamental components of environmental risk assessment (ERA) (DoE, 1995; Calow, 1998). ERA is the process by which hazards are identified, exposure quantified, and dose–response relationships determined for risk characterization (NAS, 1983). ERA has become an accepted basis for the assessment and management of contaminated sites in many countries (Ferguson and Denner, 1994; NEHF, 1996; Ferguson *et al.*, 1998), particularly for environmental relationships between soil, water, and air. However, on a site-specific basis, conventional (i.e., traditional) ERA and especially human health risk assessment (HHRA) approaches have several spatial limitations that could be overcome by using a geographical information system (GIS).

First, the ERA often provides only a single assessment in a context that can be highly variable, both temporally and spatially. The average measured concentration in the medium of concern (e.g., air, water, or soil) is used to quantify the hazard by comparing this value with some regulatory threshold (e.g., NEHF, 1996; ICRCL, 1987; MH, 1994). However, within any study area, sectors of high, medium, and low hazards are identifiable by looking at the spatial distribution of the contaminant levels. Similarly, ERAs have both spatial and time variability. The exposure or dose that each individual receives depends on the hazard present at the specific locations where most time is spent. Probabilistic exposure and risk assessments can allow for uncertainty in the input data and the predictions, but input measurements are still population based rather than specific to

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individual people. To the knowledge of the authors, there appear to be no published studies that have assessed exposure from environmental contaminants on a person by person basis in the general population.

Second, ERA should not be based, from the beginning of the process, on an exhaustive and often expensive sampling campaign. ERA can be considered as a process consisting of several tiers. To be efficient, data from each tier are used to evaluate whether there is sufficient information to make a qualified management decision. If so, the assessment is concluded at that point (DoE, 1995). Therefore, existing qualitative ("soft") information (e.g., from geological, geographical, geomorphological, or sociological sources) can sometimes be very valuable in providing a sound and less expensive assessment than direct observation on specific parameters alone and considered in isolation. Quantitative assessments do not necessarily produce a result of better quality, especially if they are carried out in a very simplistic way (e.g., by comparing the environmental measurement or prediction against a regulatory threshold). Statutory thresholds do not incorporate uncertainty by definition; measurements normally carry an appreciable amount of uncertainty, which is not necessarily stated (Burmaster and Harris, 1993; Finley and Paustenbach, 1993; Ferguson *et al.*, 1998).

Third, exposure estimates from exposure assessment models would depend on the circumstances in which the chemical occurs. An assessment based on total metal levels in the environment will generally be misleading, because different species of a given metal can have substantially different bioavailabilities (ICME, 1997). Similarly, the spatial variability of the bioavailability factor should be taken into account even within a small area, because the metal could have been derived from different sources. In this paper, the term bioavailability is used to describe the amount of a heavy metal that is absorbed into the blood stream and thus available for transfer to target organs such as the brain, liver, kidney, or bone. It follows the definition of bioavailability given by Mushak (1991) for environmental Pb.

The final problem with ERA is concerned with "risk perception" (Durant, 1997). Experts and nonexperts should be able to understand the results of the assessment, but usually the terminology and numerical outputs tend to confuse and management guidelines cannot be drawn. The appropriate spatial visualization of problems and assessment can facilitate improved communication of risk between parties.

GIS FOR HAZARD AND EXPOSURE ASSESSMENT

GIS is a tool that has the potential to overcome some of these limitations by combining spatial data from diverse sources. It allows description and analysis interactions, develops predictive models, and provides support for decisionmaking. GIS is widely applied for environmental modeling (Bonham-Carter, 1994; Goodchild *et al.*, 1996), but very few applications in human health-related studies have been published, most of which concern applications in epidemiology (Guthe *et al.*, 1992; Kitron and Kasmierczak, 1997; Vine *et al.*, 1997) rather than risk or exposure assessments (Valjus *et al.*, 1995; Siniscalchi *et al.*, 1996).

The aim of this study is to describe and evaluate different methodologies for assessing personal hazard and exposure from environmental contaminants incorporating spatial parameters. These methodologies have been named either "spatially resolved hazard assessment" (SRHA) or "spatially resolved exposure assessment" (SREA), with both quantitative and semiquantitative elements.

The quantitative SRHA and SREA are more traditional in concept and are based on a threshold approach. To account for measurement uncertainty in the SRHA (Ramsey and Argyraki, 1997), both a deterministic and a probabilistic approach are used. The semiquantitative SRHA and SREA are based on modeling using "weighted factors in linear combination" (WLC), described in detail below, a method allowing for the impact of more influential parameters.

CASE HISTORY: LAVRION, GREECE

To assess the viability of the SRHA and SREA in practice, an area has been selected known to contain very high levels of potential exposure from metals in soil. This is the town of Lavrion in Greece, 54 km southeast of Athens. Lavrion has an extensive mining and smelting history which probably began before 3000 BC and continued intermittently until 1977 AD (Demetriades *et al.*, 1996). The concentration of Pb in soil in the studied area is generally very high (median, 7410 μg Pb/g soil, n = 213) and well above a typical background crustal abundance of 1.3 μg Pb/g (Krauskopf and Bird, 1995). People in the town live on or close to smelter wastes and some of them eat homegrown vegetables and fruits. Furthermore, health studies on children have indicated that there is some evidence for substantial exposure to Pb, with documented medical evidence for mental retardation, a slower response rate, and increase in sickliness in school-age children (Eikmann *et al.*, 1991; Makropoulos *et al.*, 1991; Kafourou *et al.*, 1997).

Previous studies include soil geochemical mapping, both regional (Demetriades *et al.*, 1994a,b, 1996) and local (Hadjigeorgiou-Stavrakis and Vergou-Vichou, 1992; Demetriades *et al.*, 1997), soil geochemical mapping, lithological mapping (Demetriades *et al.*, 1998), characterization of smelter wastes (Kontopoulos *et al.*, 1995; Xenidis *et al.*, 1997), sequential extraction soil analysis (Demetriades *et al.*, 1994a,b; Li *et al.*, 1995), and a number of human health studies (Eikmann *et al.*, 1991; Makropoulos *et al.*, 1991; Kafourou, 1997). The feasibility of the SERA approach was assessed for selected pathways of Pb exposure. The existing person-specific information included blood-Pb concentration of selected children (Table 1), but it did not include details of their activity patterns (e.g., time spent outside or inside the house, consumption of home-grown vegetables and fruits). Nevertheless, such an approach could easily be expanded to include these aspects in the future.

The exposure situation at Lavrion appears to be multiple-source and multiple-pathway. However, there are several gaps and inconsistencies in the biomedical data, obtained from previous studies, which have thus limited the scope of the current assessment concerning Pb in soil.

The three site-specific objectives selected for this study have been (a) to define the hazards and exposure to be assessed, (b) to produce both hazard and exposure maps using GIS techniques, and (c) to test the validity of the assessment by comparing predicted exposure with biomedical measurements.

Sources of Information

The site information was provided in digital form by the Institute of Geology and Mineral Exploration (IGME), Greece (Demetriades *et al.*, 1998). A total of three distributions for Pb were used as the basis for the quantitative maps: (1) total Pb concentration at each sampling site, (2) kriged total soil-Pb concentration, and (3) kriged extractable soil-Pb concentration. The semiquantitative maps are based on a larger number of factors derived from various images, i.e., land use, stacks, rivers, lithology, town plan, and smelter wastes.

Total soil-Pb concentrations were analyzed by ICP–AES after a digestion with nitric and perchloric acids (Thompson and Walsh, 1988). A method of sequential chemical extraction adapted from the procedure of Tessier *et al.* (1979) was used to measure the concentration of Pb partitioned in the various geochemical fractions in the soil. This adaptation was made in order to make the operation easy, efficient, and suitable for ICP–AES analysis (Li *et al.*, 1995).

The kriged total Pb concentration and kriged extractable concentration were plotted after an exhaustive geostatistical structural analysis on the two data sets (Demetriades *et al.*, 1998). The spatial relationships were examined by constructing variograms in different directions of raw and log (base 10) total and extractable Pb contents (Pennatier, 1996). Since both data sets are positively skewed, the log-transformed Pb concentrations gave more realistic variograms. The different geostatistical models were examined against the actual Pb values with cross-validation kriging techniques (Englund and Sparks, 1988). The model with the lowest mean kriging standard deviation and estimation error was selected for total and extractable Pb concentrations. The log-transformed concentration estimates were then transformed back to their original units (μg Pb/g soil) in order to create the different maps.

METHODS FOR QUANTITATIVE HAZARD MAPPING

To assess the hazard as "the total Pb concentration in soil," quantitative hazard mapping has been employed. The quantitative hazard assessment maps (both probabilistic and deterministic) are based on the threshold concept, whereby the land is classified on the basis of an established regulatory or statutory threshold value. In this case the ICRCL (ICRCL, 1987) trigger concentration for parks, playing fields, and open space (2000 μg Pb/g soil) has been selected to demonstrate the approach, although more appropriate lower, site-specific, statutory threshold values could be considered, if required. The image used for the elaboration of these maps is the total soil-Pb concentration at each sample site. The total soil-Pb concentration is classified on the basis of its position relative to the statutory threshold in order to create the categories for the different hazard maps.

The hazard identification map displays the distribution of concentration of the substance across the area (Fig. 1a). The deterministic hazard map (Fig. 1b) defines two categories for the level of contamination in soil: (1) contaminated or (2) uncontaminated. Such a classification does not, however, consider the uncertainty in the measurement of the concentration of the substance in soil, which can have profound effects on a realistic assessment of the degree and extent of contamination.

Effects of Uncertainty

To address the uncertainty of the measurements, a probabilistic classification of contaminated land (Ramsey and Argyraki, 1997) is used to produce the

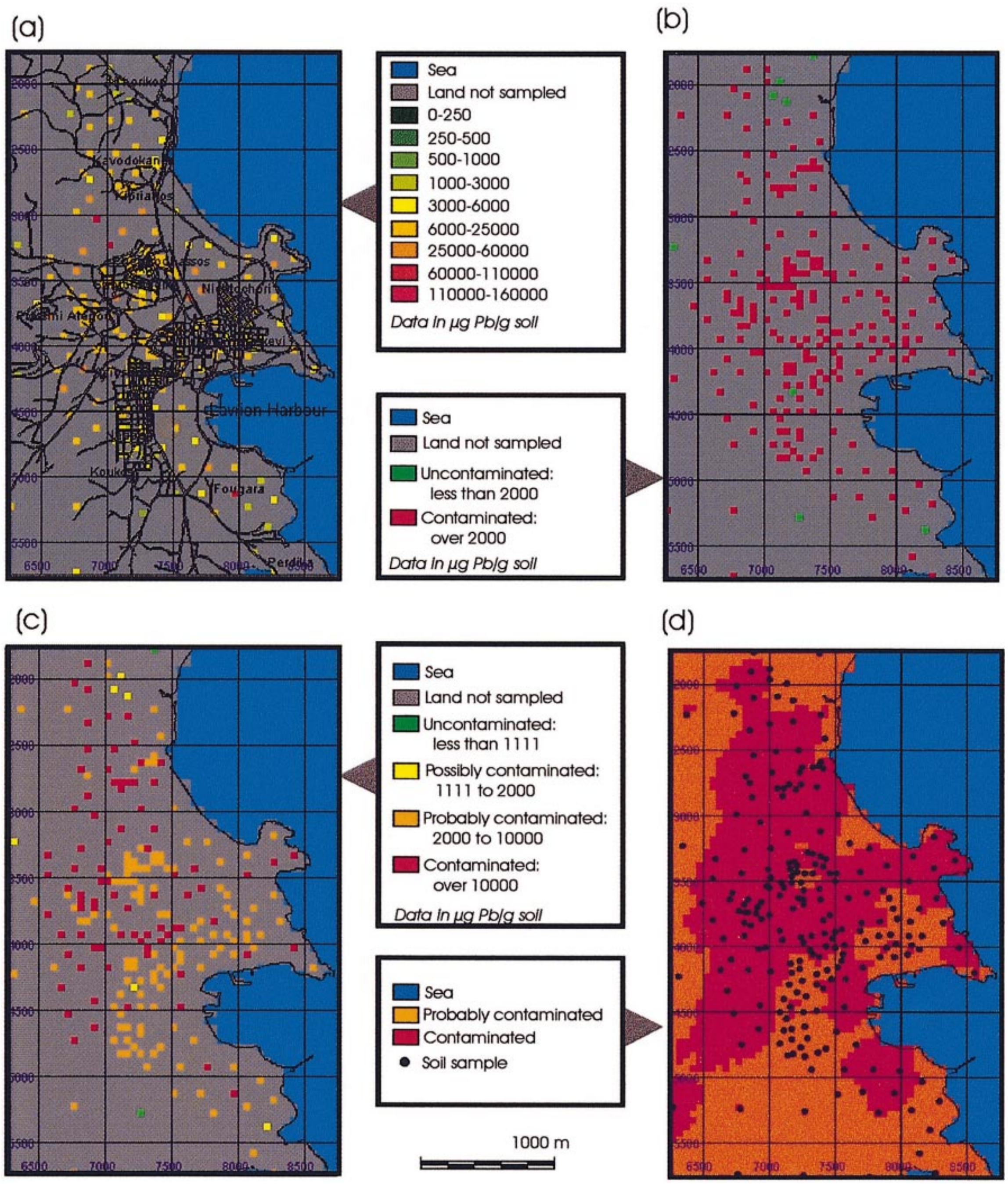

FIG. 1. Quantitative hazard maps. (a) Hazard identification based on total soil-Pb concentrations at each sample site; (b) deterministic hazard based on total soil-Pb concentrations at each sample site; (c) probabilistic hazard based on total soil-Pb concentrations at each sample site and measurement uncertainty (80% of the concentration of Pb); (d) probabilistic hazard based on kriged soil-Pb concentrations and kriging uncertainty (using kriging standard error of estimation).

probabilistic hazard map (Fig. 1c). The probabilistic hazard map addresses decision-rule uncertainty by considering the uncertainty as being on the statutory threshold used to determine the decision rule. The classification defines four categories based on the extent of overlap of the uncertainty with a single threshold value: (1) contaminated, (2) probably contaminated, (3) possibly contaminated, and (4) uncontaminated. For the uncontaminated category, for example, the entire range of uncertainty is lower than the regulatory threshold value (T). The probability of this site being contaminated, because the element concentration lies over the statutory threshold, is, therefore, <0.025 (i.e., 2.5%). This computational device can be used for classifying soil samples directly, without the need to calculate uncertainty values for each measurement.

The use of duplicate samples is one method that has been proposed for the estimation of sampling uncertainty (Ramsey and Argyraki, 1997). Unfortunately, such duplicates were not available at the time of this study. However, in order to illustrate the technique, an uncertainty of 80% has been chosen as a worst-case scenario.

An alternative approach to the probabilistic hazard map was applied using the kriged estimates of concentration, described above. Two times the logarithm to the base 10 of the kriging standard error of estimation (lg-ksd) is used as an estimate of uncertainty (U); this is different for each kriged cell in the region. The images of log base 10 for the kriged concentration of Pb and of the log base 10 for the kriging standard error of estimation are used.

METHODS OF QUANTITATIVE EXPOSURE MAPPING

Two exposure assessment models have been used to derive the exposure assessment maps, either directly (USEPA, 1994) or indirectly using the model algorithms (Shell, 1994). In total, three different methodologies were developed, one based on the IEUBK model and two based on the HESP model. The IEUBK (USEPA, 1994) and the HESP (Shell, 1994) models have been chosen to predict exposure using the bioavailable fraction of Pb in soil. The bioavailable fraction is obtained by multiplying the total Pb concentration measured at the site by a bioavailability factor that is assumed to be constant throughout the surveyed area. The bioavailable fraction can either be specified by the user or taken as a default value provided by the model.

Exposure assessment mapping based on the HESP model has here been based first on the total Pb concentration and uses the HESP model algorithm directly. The alternative is predicting exposure by using the experimentally measured bioavailable fraction in each soil sample directly and modifying the HESP algorithm slightly to allow for this difference (Appendix 1). By using this second approach, the personal exposure can be predicted by knowing the bioavailable Pb fraction at an individual's place of residence. For the development of this methodology, the available data from the sequential extraction of Lavrion soils for chemical fractionation of trace elements were used as an approximation for the bioavailable fraction of Pb in soil (Demetriades *et al.*, 1998). The sequential chemical extraction aims to examine the partition of the elements among the various geochemical phases of the soils on their chemical reactivity (Tessier, 1979). The first and second extractions (exchangeable and bound onto carbonates, respectively) represent the most readily dissolved fraction of Pb in soil (Li *et al.*, 1995). These concentrations have been added together to represent a bioavailable fraction to children. There are several *in vitro* methods that have been developed to mimic bioavailability from ingested Pb in soil and dust that could be used in the future to derive a more refined assessment (e.g., Ruby *et al.*, 1995).

Exposure Assessment Maps Using the HESP Model as a Basis

The methodology to create these maps includes two steps, the first being to use a spreadsheet, such as Excel, to determine the uptake from direct ingestion of soil or dust ($DU_{n,x}$), which is calculated for a child living in a house with a garden. Then, using a GIS, such as Idrisi (Eastman, 1997), the calculated $DU_{n,x}$ is assigned to every house of interest (i.e., those with children who have permitted blood-Pb measurements) in order to create two different quantitative exposure maps. Both the quantitative exposure map based on the HESP model and the extractable soil-Pb concentration (Fig. 2c) and the quantitative exposure map based on the HESP model and the total soil-Pb concentration (Fig. 2d) use the value of either kriged extractable or total soil-Pb concentration corresponding to each house where the assessed children live.

Exposure Assessment Map Using the IEUBK Model as a Basis

The integrated exposure uptake–biokinetic model (IEUBK) (USEPA, 1994) is a mathematical model that derives a deterministic estimate for blood-Pb concentration in young children based on environmental exposure to Pb from various sources including soil (Tsuhi and Searl, 1996). The user is able to specify uptake, but not biokinetic assumptions, which are included in the model by default.

The quantitative exposure map using the IEUBK model is based directly on the "batch-mode" run, i.e., the soil concentration for a house is used to estimate an individual child's mean blood-Pb. All default values of the model are used. These results are imported into the GIS for creating the quantitative exposure map based on the IEUBK model and the total soil-Pb concentration (Fig. 2b).

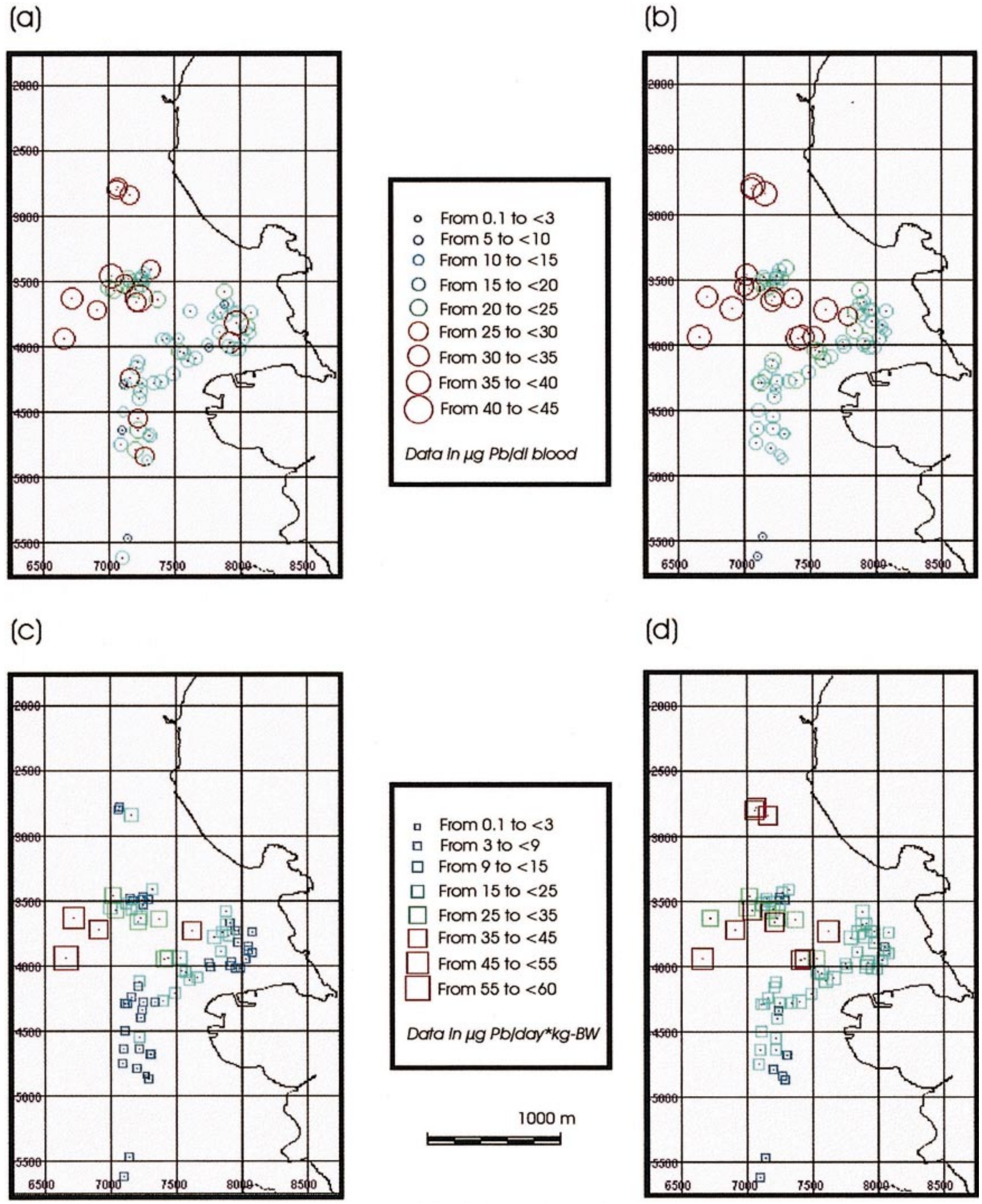

FIG. 2. Quantitative exposure maps. (a) Measured blood-Pb levels of 7-year-old children in µg Pb/dl blood; (b) predicted blood-Pb levels of 7-year-old children in µg Pb/dl blood using IEUBK model; (c) predicted average daily exposure (ADE) to 7-year-old children using extractable soil-Pb concentration in mg Pb/day ∗ kg body wt using HESP model; (d) predicted average daily exposure (ADE) to 7-year-old children using total soil-Pb concentration in mg Pb/day ∗ kg body wt using HESP model.

METHOD OF SEMIQUANTITATIVE HAZARD MAPPING

According to the definition of hazard (Calow, 1998), a toxic chemical is not necessarily hazardous if the circumstances under which it occurs prevent the potential for harm. The quantitative hazard mapping described above, however, provides a simplistic approach, since the hazard is only related to the Pb concentration in soil and not to the circumstances under which it occurs. It is based on a single-objective/single-criterion decision rule (Bonham Carter, 1994). The objective is to establish whether the concentration of Pb in soil is a hazard. The criterion is to classify the concentration as hazardous or not with respect to an established regulatory threshold. To characterize the particular circumstances under which Pb occurs in soil to refine the concept of hazard, a different approach should be taken. To emphasize the conceptual difference in concepts of hazard, the second hazard is defined as "Pb accessible to children from soil and dust." This recognizes that only a fraction of total Pb in soil is likely to be hazardous to children, i.e., the fraction accessible to children because of the circumstances under which it occurs. In this case, a single-objective/multicriteria evaluation needs to be applied. This can be achieved using the a decision support approach within the GIS called multicriteria evaluation (MCE) (Eastman, 1997), whereby the criteria are combined according to the objective of the decision. MCE is run using the WLC procedure, for which continuous criteria (factors) and Boolean constraints are standardized to a common scale (0 for lowest hazard rising to 1 for highest hazard) to be combined in proportions to their degree of influence on the "availability of Pb in soil and dust to children." A somewhat similar procedure has been applied to address mineral exploration issues (Bonham-Carter, 1994).

The methodology has been called semiquantitative hazard mapping because it is based on maps containing both qualitative information (e.g., lithology and land use) and quantitative information (e.g., soil-Pb concentration). The analysis relies on a numerical categorization of hazard, which is created partly using fuzzy logic. Fuzzy sets enable the classification of parameter groups that are not delimited by sharp boundaries, i.e., the transition between membership and nonmembership of a fuzzy set is gradational (Eastman, 1997).

The methodology developed for computing the semiquantitative hazard map based on MCE and WLC involves five basic steps: (1) setting the objective; (2) selecting, (3) weighting, and (4) standardizing the criteria; and (5) creating the maps.

METHOD OF SEMIQUANTITATIVE EXPOSURE MAPPING

Similar logic has been applied to the exposure assessment, as described above, for the semiquantitative hazard assessment mapping. The quantitative exposure assessment provides an average daily exposure (ADE) that is compared with a tolerable daily intake (TDI) in order to assess risk. It consists again of a single-criterion decision rule. Exposure, however, is a function of both hazard and time of exposure. Therefore, the same multicriteria used to assess a hazard may also be employed to assess exposure, adding a further criterion that is related to time of exposure. This factor concerning time of exposure depends on the social characteristics of the population, the distribution of their time, and their activity patterns.

RESULTS AND DISCUSSION

Quantitative Hazard Mapping (Figs. 1a–1d)

The hazard identification map (Fig. 1a) displays the high level of total Pb concentration in the soil across the area; these vary from 810 to 152,000 μg Pb/g soil.

The regulatory threshold used for the deterministic hazard map (Fig. 1b) is the ICRCL (1987) trigger concentration for parks, playing fields, and open space, 2000 μg Pb/g soil. This statutory threshold has been selected to demonstrate the methodology at the highly contaminated urban area of Lavrion, although a lower threshold may be more appropriate for certain types of land use. Most of the samples are contaminated according to this classification, but this approach ignores uncertainty in the measurements.

The probabilistic hazard map based on total soil-Pb concentration at each sampling site (Fig. 1c) is based on an assumed measurement uncertainty of 80%, taken as the worst-case scenario.

A comparison of the deterministic and probabilistic hazard maps shows, in both cases, that the major part of the soil cover is contaminated. The large measurement uncertainty has, nevertheless, a small effect in expressing a reduced probability of contamination, except at a small proportion of localities (mainly north, west, and south of the area). This contrasts with the large effect of the probabilistic approach when areas have concentrations close to the statutory threshold value (Ramsey and Argyraki, 1997). The "probably contaminated land" displayed in the probabilistic hazard map based on kriged soil-Pb concentrations (Fig. 1d) is more frequent in those areas where the sampling density is lower (i.e., kriging standard error of estimation is high), north, west, and south of the study area.

Quantitative Exposure Map Based on the IEUBK Model (Fig. 2b)

From the 234 children that participated in the biomedical survey, only those below age 84 months (7 years old) were selected, since it is the last category considered in the IEUBK model. The total number of children in this age group is 103 with an average age (arithmetic mean) of 6.5 years. The number of children used in the analysis is further reduced to 75, as some of the children live in the same house, and only the first is recorded in the GIS database. Another drawback is that it is not possible to predict, by using the IEUBK model in batch mode, the exposure from soil and dust only. Therefore, the predicted blood-Pb level is estimated from all other possible pathways (i.e., water, air, food). Nevertheless, by checking the results in normal mode, the magnitude of the intake from soil and dust is much higher than that from all the other pathways of exposure.

The measured blood-Pb has an arithmetic mean (AM) and standard deviation (SD) of 19.1 and 8.6 μg Pb/dl blood, respectively; whereas the predicted blood-Pb has, correspondingly, values of 21.1 and 8.6 μg Pb/dl blood. The Pearson's correlation coefficient between predicted (dependent variable) and measured (independent variable) blood-Pb levels is 0.41, which is statistically significant ($P = 0.05$). The regression equation for the relationship is

$$\begin{aligned}&\text{predicted blood-Pb level}\\&= 12.8 + (0.43 * \text{measured blood-Pb level});\\&[n = 75].\end{aligned}$$

The model predictions are, therefore, higher than the measured blood-Pb levels by up to 22.4 μg/dl. From this value upward, the situation is the inverse; the predictions are lower than the measured values. The degree of scatter could be related to all the other factors not accounted for in the analysis, i.e., specific time activity patterns, nutritional factors, and personal variability.

The highest positive residuals from the regression are located toward the central part of the Lavrion urban area, where there are very high levels of Pb in soil, which do not result in very high blood-Pb levels in children. Similarly, relatively high measured blood-Pb levels (from 25 to 30 μg Pb/dl blood) were not predicted by the model, in parts of Lavrion with much lower soil-Pb levels, perhaps indicating contributions to blood-Pb from some other exposure route, or biokinetic differences among the child population.

Quantitative Exposure Assessment Maps Based on the HESP Model (Figs. 2c and 2d)

The equations and values used are given in Appendix 1. All values are those given by the HESP model, except those related to the time activity pattern. For this analysis, the factor N_y (fraction of time spent annually indoors, outdoors, sleeping, or away from the location) is calculated using a child distribution of time estimated for Lavrion children (Table 1).

The DU (total direct ingestion of soil or dust) calculated for a house with a garden is 231.9 mg soil/day. Most children sampled for blood-Pb were between the ages of 7- and 8 years. The average body weight for a male child of 7.5 years, used in the CLEA model (Ferguson, 1995), was employed instead of the value provided by HESP, which is for a 4-year-old child. The weight is assumed constant and has a value of 24.2 kg.

One method of checking the broad validity of the predicted exposure of children to Pb by this method is to quantify the relationship with measured internal exposure as blood-Pb. The Pearson's regression coefficient (r) for the relationship between predicted ADE using (i) extractable soil-Pb concentrations (Fig. 2c) and (ii) total soil-Pb contents (Fig. 2d) and the measured blood-Pb levels were found to be 0.34 and 0.40, respectively ($P < 0.001$). Predictions with respect to extractable soil-Pb concentrations (Fig. 2c) are low compared to the measured blood-Pb contents especially in the northern part of the Lavrion urban area. Pearson's r for the relationship between the predictions by IEUBK (Fig. 2b) and HESP (Fig. 2d) is 0.95 ($P < 0.001$).

Semiquantitative Hazard and Exposure Mapping

Setting the objective. The primary objective of the semiquantitative hazard and exposure maps is to assess hazard and exposure from "Pb accessible to children." A secondary objective is to test the relevance of including the Pb concentration in soil as a factor in the hazard and exposure analysis. Four different maps

TABLE 1
Measured and Predicted Blood-Pb Levels in Children (μg Pb/dl blood) and Pb Concentrations in Soil Samples (μg Pb/g soil) at lavrion, Greece

	Measured blood-Pb level (μg Pb/dl)	Predicted blood-Pb level (μg Pb/dl)	Pb concentration in soil samples (μg Pb/g soil)
Percentiles			
10th	11.0	13.5	2670
20th	12.6	15.2	3730
30th	14.3	16.2	4650
40th	16.3	17.4	6050
50th	18.1	18.7	7410
60th	19.1	19.7	9700
70th	21.0	20.9	12,270
80th	25.2	28.4	15,130
90th	28.3	34.3	21,620
99th	38.4	38.4	66,350
99.9th	40.5	38.6	144,120
Range	6.7–40.7	8.8–38.6	810–151,580
n	103	103	2210

are created: two hazard maps and two exposure maps. The first map of each type (i.e., hazard and exposure) includes the Pb concentration in soil as a factor in the analysis, whereas the second does not.

Criteria selected. Ten criteria were selected for this analysis from among a larger number that could influence the hazard and exposure (e.g., wind direction, position of the hill with respect to wind direction). These criteria are divided in two categories related to whether they are likely to increase Pb concentration in soil (e.g., when the area is situated over the site of a former metal-related industry) or to make Pb more accessible to children (e.g., degree of dustiness of the waste) (Table 2). The proximity to roads factor is included in the analysis because at the time of the biomedical survey (N. Vlachoyiannis, personal communication) most of the roads in Lavrion were not tarred, and significant amounts of dust could have been derived from them.

The exposure factor is exclusive to the exposure maps (Figs. 3c and 3d). The image used as a basis for estimating the "time of exposure" is the land-use map. Four main categories of exposure can thus be established based on the areas where children spend most of their time. The categories are (a) residential area (with a garden), (b) agricultural area, (c) recreational area, and (d) school. Exposure in the rest of the area is taken as minimal.

Weighting the criteria. The technique used in this paper for assigning appropriate weights is that developed by Saaty (1977; Eastman, 1997) in the context of a decision-making procedure known as the analytical hierarchy process (AHP). The weights are developed by providing a series of pairwise comparisons of the relative importance of factors to the suitability of pixels for the hazard being evaluated. These pairwise comparisons are then analyzed to produce a set of weights that sum to 1 (Table 2).

Standardization of criteria. All areas in the different criteria maps are allocated a value of hazard ranging from 0 to 255. However, the hazard values are not necessarily assigned in a similar manner. Some criteria have discrete categories (e.g., degree of dustiness of the waste or whether or not a site overlies quaternary deposits), and others have continuous categories (e.g., proximity to current or previous stacks or proximity to roads).

A value is given to the discrete categories of a hazard. Continuous categories of hazards are based on the application of fuzzy-set membership functions.

Creating the maps. Once the criteria maps (images) have been developed, the MCE module of the Idrisi GIS is used following the logic of WLC. The module requires the specification of the number of factors, their names, and the weights to be applied to the factors. The exposure maps should be interpreted as the potential child exposure expected at a specific location (e.g., if a child lives there, studies there, plays there); it is not the total exposure expected.

All of the regression coefficients (r) between Figs. 3a and 3d and the measured blood-Pb concentrations are statistically significant, having values of 0.47, 0.43, 0.36, and 0.34, respectively ($P < 0.001$). This confirms that the MCE–WLC approach shows effectively the areas with highest exposure, even when soil-Pb concentrations are not included in the model.

TABLE 2
Criteria Used in the Semiquantitative Hazard and Exposure Maps (Figs. 3a–3d)

Factor	Weights for hazard map with soil-Pb factor	Weights for hazard map without siol-Pb factor	Weights for exposure map with soil-Pb factor	Weights for exposure map without soil-Pb factor
Likely to increase the Pb concentration in soil				
Soil Pb concentration	0.2881	N/A[a]	0.2157	N/A[a]
Over metal-related industry	0.0520	0.2121	0.0404	0.1511
Proximity to industry	0.0850	0.1337	0.0612	0.0997
Proximity to previous stacks	0.0787	0.0816	0.0582	0.0603
Proximity to rivers	0.0151	0.0182	0.0128	0.0151
Over Quanternary deposits	0.0323	0.0320	0.0248	0.0246
Likely to make Pb more accessible to children				
Proximity to roads	0.0757	0.0577	0.0560	0.0481
Dustiness of wastes	0.2881	0.3308	0.1998	0.2468
Proximity to wastes	0.0850	0.1337	0.0612	0.0997
Time of exposure	N/A[a]	N/A[a]	0.2699	0.2544

[a] N/A, not applicable.

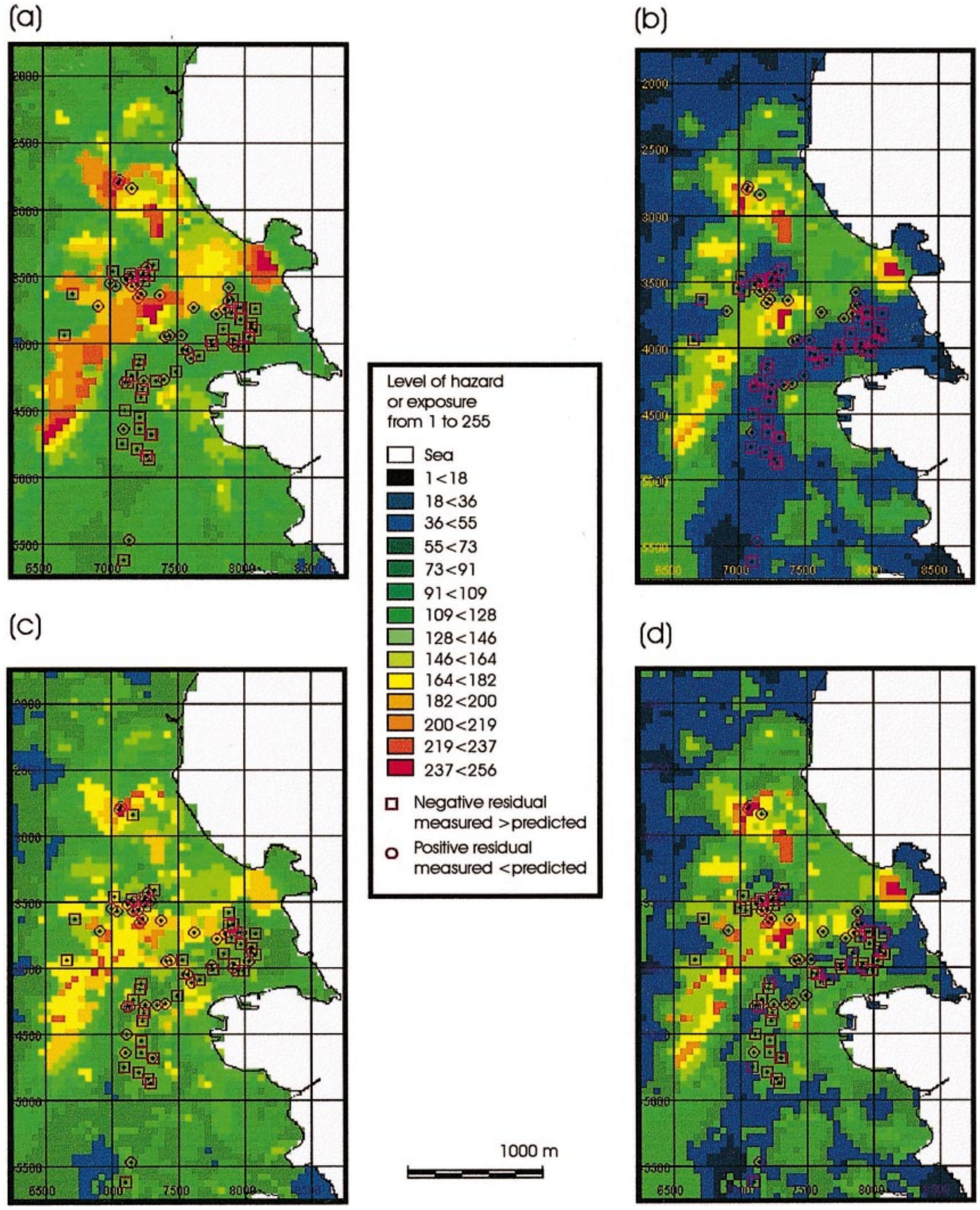

FIG. 3. Semiquantitative hazard and exposure maps. (a) Hazard map including the factor of total soil-Pb concentration; (b) hazard map excluding the factor of total soil-Pb concentration; (c) exposure map including the factor of total soil-Pb concentration; (d) exposure map excluding the factor of total soil-Pb concentration.

CONCLUSIONS

This study has illustrated the relevance of different spatially resolved hazard and exposure assessment techniques in assessing contaminated land. The following five conclusions summarize the most significant aspects of this study:

1. Different methodologies have been devised using GIS for spatially resolved hazard and exposure assessment, permitting spatial differences in the

distribution of both hazard and target to be evaluated. The advantages of such maps are that they help to prioritize areas for action, such as remediation or identification of areas unsuitable for housing.

2. A partial validation of the methods has been achieved from significant correlations between the Pb exposures predicted by the models and the measured values of internal exposure from blood-Pb.

3. The semiquantitative method, using the MCE–WLC technique, has been shown to integrate semiquantitative information, such as dustiness of waste and presence of metal-related industry. This can improve upon traditional EA/RA procedures that rely solely on soil or dust-Pb concentrations. Moreover, the robust nature of this approach was demonstrated by its correlation with measured blood-Pb levels, even when data on soil-Pb were excluded from the method. The semiquantitative hazard maps had a higher regression coefficient than the quantitative exposure maps, proving the strength of the semiquantitative method.

4. The significant correlation between the blood-Pb predictions using the IEUBK model and the blood-Pb measurements provides a partial validation of the method, allowing for uncertainty in measurements (from both sampling and analysis) and the lack of some site-specific measurements.

5. The successful application of these methodologies to the Lavrion urban area, although preliminary in nature, does nevertheless demonstrate the potential of the spatially resolved technique. Further refinement will include consideration of other exposure pathways to permit more accurate exposure assessments for Pb and more rigorous validation of the methodology. The principles of this approach should be generally applicable to other potentially contaminated areas and other contaminants in the Lavrion urban area. Selection of suitable exposure routes and validation methods will clearly require site-specific adaptation of the methods.

APPENDIX 1: ALGORITHMS AND VALUES USED TO DEVELOP THE QUANTITATIVE EXPOSURE MAPS BASED ON THE HESP MODEL

The HESP model is a computer model that derives a deterministic value of exposure to humans from contaminants in soil. It estimates exposure for two types of residents: an adult and a young child. The calculated exposure represents the ADE in μg Pb/day/kg body wt. The average indicates the year's average, independent of seasonal changes. The algorithm used in HESP to predict exposure through direct ingestion of soil and dust (DU) is

$$\mathrm{DU} = \left(\sum \mathrm{DU}_{n,x}\right) \times c_{\mathrm{s}}/w, \tag{1}$$

where $\mathrm{DU}_{n,x}$ is uptake from direct ingestion of soil or dust per unit body weight per season (mg soil/day), n is index indicating soil or dust, x is index indicating summer or winter, c_{s} is the concentration in the soil's top layer (0–0.25 m) (mg/kg dry matter), and w is receptor weight (kg).

The concentration of Pb in dust is not required, since it is approximated by allocating a fraction of the outdoor soil to indoor dust. The model provides default values for the rest of the input parameters, e.g., child's weight, time activity patterns, bioavailability fraction.

$\mathrm{DU}_{n,x}$, the uptake from direct ingestion of soil or dust per unit body weight per season (mg soil/day), is calculated by adding the contribution of both soil and dust ingestion during the summer and winter seasons:

$$\mathrm{DU}_{x,\mathrm{dust}} = \mathrm{AID} \times f_{\mathrm{a,ing}} \times f_{\mathrm{rs,i}} \times f_{\mathrm{i}} \times N_{\mathrm{i}} \tag{2}$$

$$\mathrm{DU}_{x,\mathrm{soil}} = \mathrm{AID} \times f_{\mathrm{a,ing}} \times f_{\mathrm{o}} \times N_{\mathrm{o}}, \tag{3}$$

where AID is the amount of soil ingested daily (mg/day) based on the average amount on a yearly basis, $f_{\mathrm{a,ing}}$ is fraction uptake/intake for ingestion (bioavailability) [dimensionless], $f_{\mathrm{rs,i}}$ is fraction of soil in indoor dust [dimensionless], f_{y} is fraction of soil or dust covering the skin indoors or outdoors [dimensionless], f_{y} is [$\mathrm{DAE_y}/(\mathrm{DAE_i} + \mathrm{DAE_o})$], DAE is the amount of soil or dust on the skin [$\mathrm{kg/m^2}$], N_{y} is fraction of time spent annually indoors, outdoors, sleeping, or away from the location ($N_{\mathrm{y}} = \sum -N_x$) (always ≤ 1) [dimensionless], and index y indicates indoors (i), outdoors (o), sleeping, or away from the location,

$$N_x = \sum t_x 1_y/24 \times (t_x 2_y/7) \times f_x, \tag{4}$$

where $t_x 1_y$ is time spent indoors, outdoors, sleeping, or away from the location in a season per day (h/day), $t_x 2_y$ is time spent indoors, outdoors, sleeping, or away from the location in a season per week (days/week), and f_x is fraction of the season.

Equation (1) is slightly modified in order to use c_{e} instead of c_{s} as follows:

$$\mathrm{DU} = \left(\sum \mathrm{DU}_{n,x}\right) \times c_{\mathrm{e}}/(w \times f_{\mathrm{a,ing}}), \tag{5}$$

where c_{e} is the extractable soil-Pb concentration, which in theory should correspond to "$c_{\mathrm{s}} \times \mathrm{f_{a,ing}}$" (the total soil-Pb concentration multiplied by the bioavailable fraction for soil and dust ingestion) in Eq. (1).

A value of extractable soil-Pb concentration is given to each house using a kriged extractable soil-Pb concentration image. For both maps, the child's weight (w) is taken as constant (i.e., 24.2 kg).

TABLE A1
Values used for Eqs. (1) to (4)

Parameter[a]	Value	Unit
AID	1100	mg/day
$f_{\mathrm{a,ing}}$ (for Pb)	0.3	Dimensionless
$f_{\mathrm{rs,i}}$ (for Pb)	0.5	Dimensionless
f_{I}	0.09894	Dimensionless
f_0	0.90106	Dimensionless
DAE	0.00056	kg/m^2
DAE	0.0051	kg/m^2

[a] The terms have been defined in the text.

TABLE A2
Time Activity Patterns for a Child Living in a House with a Garden at Lavrion

Time category (h/day, unless indicated otherwise)	Child (Lavrion)	
	Summer	Winter
Active days(AD)	5	5
Passive days(PD)	2	2
Time sleeping, AD – PD	10	10
Time away, AD	6	6
Time outside, AD	4	1
Time inside, AD	4	7
Time away, PD	0	0
Time outside, PD	6	2
Time inside, PD	8	12
Time swimming	0	0

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