Assessing Potential Risk of Heavy Metal Exposure from Consumption of Home-Produced Vegetables by Urban Populations

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We performed a risk assessment of metal exposure to population subgroups living on, and growing food on, urban sites. We modeled uptake of cadmium, copper, nickel, lead, and zinc for a selection of commonly grown allotment and garden vegetables. Generalized linear cross-validation showed that final predictions of Cd, Cu, Ni, and Zn content of food crops were satisfactory, whereas the Pb uptake models were less robust. We used predicted concentrations of metals in the vegetables to assess the risk of exposure to human populations from homegrown food sources. Risks from other exposure pathways (consumption of commercially produced foodstuffs, dust inhalation, and soil ingestion) were also estimated. These models were applied to a geochemical database of an urban conurbation in the West Midlands, United Kingdom. Risk, defined as a "hazard index," was mapped for three population subgroups: average person, highly exposed person, and the highly exposed infant (assumed to be a 2-year-old child). The results showed that food grown on 92% of the urban area presented minimal risk to the average person subgroup. However, more vulnerable population subgroups (highly exposed person and the highly exposed infant) were subject to hazard index values greater than unity. This study highlights the importance of site-specific risk assessment and the "suitable for use" approach to urban redevelopment. ***Key words:*** **exposure, hazard index, mapping, metals, risk, urban, vegetables.** ***Environ Health Perspect*** **112:215–221 (2004). doi:10.1289/ehp.5589 available via *http://dx.doi.org/* [Online 31 October 2003]**

The U.K. Government aims to build 60% (4.40 million) of new homes on previously developed land by the year 2016 [Alker et al. 2000; Department of Environment, Transport and the Regions (DETR) 1998]. This policy should regenerate urban and inner-city areas, thus reducing pressure on further "greenfield" (land that has had no previous building development) development. However, much of the previously developed or "brownfield" land in the United Kingdom is affected by past industrial contamination, where a brownfield may be defined as

> any land or premises which has previously been developed and is not currently fully in use, although it may be partially occupied or utilised. It may also be vacant, derelict or contaminated. (Alker et al. 2000).

Prolonged exposure to heavy metals such as cadmium, copper, lead, nickel, and zinc can cause deleterious health effects in humans (Reilly 1991). Metal contamination of garden soils may be widespread in urban areas due to past industrial activity and the use of fossil fuels (Chronopoulos et al. 1997; Sanchez-Cámazano et al. 1994; Sterrett et al. 1996; van Lune 1987; Wong 1996). Heavy metals may enter the human body through inhalation of dust, direct ingestion of soil, and consumption of food plants grown in metal-contaminated soil (Cambra et al. 1999; Dudka and Miller 1999; Hawley 1985). Potentially toxic metals are also present in commercially produced foodstuffs [Department of Environment, Food and Rural Affairs (DEFRA) 1999]. Exposure to potentially toxic metals from dust inhalation or soil ingestion is usually modeled simply as the concentration of a contaminant measured in the soil multiplied by the quantity of dust inhaled or soil ingested (Konz et al. 1989). This is a conservative approach to estimating dose, because the bioaccessibility of heavy metals adsorbed on ingested soil is not 100% (Ruby et al. 1999). However, predicting exposure to potentially toxic metals from consumption of food crops is more complicated because uptake of metals by plants depends on soil properties and plant physiologic factors. This leads to much larger uncertainties associated with estimating potential doses through food chains compared to the uncertainties associated with other exposure pathways such as soil ingestion and dust inhalation (McKone 1994).

Under Part IIA of the Environmental Protection Act 1990, the U.K. Government favors a "suitable for use" approach to redevelopment (DETR 2000): Land is contaminated only if the current or intended use of a site has the potential to cause an unacceptable health risk to human occupants or to the environment. Under the U.K. Town and Country Planning Act 1990 (DETR 2000), this approach requires that land be assessed for redevelopment on a site-specific basis. At present, concentrations of metals in the soil are compared to metal-specific "trigger values" (termed "maximum contaminant levels" or "maximum contaminant concentrations" in North America). In the past these trigger values were based on total contaminant concentration in the soil [e.g., Inter-departmental Committee on the Redevelopment of Contaminated Land (ICRCL) 1987]. More recently, the introduction of Contaminated Land Exposure Assessment [CLEA; DEFRA and Environment Agency 2002a] in April 2002 has replaced these trigger values with generic soil guidance values (SGVs; DEFRA and Environment Agency 2002c). The SGVs are considered a significant improvement on the previous ICRCL values and for Cd at least, soil pH categories are employed where food plants are to be grown. Where a soil exceeds the SGV, it is recommended that a risk assessment or remediation measure is performed for the site in question (DEFRA and Environment Agency 2002c). Additionally, exceedance of an SGV indicates that some further risk management action should be undertaken. However, the use of single trigger values or SGVs for most scenarios may represent a poor indication of the risk associated with a specific site. There is therefore a requirement for site-specific risk assessment based on commonly measured geochemical and population parameters.

Current risk assessment models typically predict uptake of metals by vegetables using a concentration ratio (CR) relating the concentration of a metal in the soil to its concentration in a crop plant. For example, CLEA uses expressions combining CR values with metal distribution coefficients (k_d) published in Baes et al. (1984). These relations were partly parameterized using pH-dependent k_d algorithms from metal solubility studies in the literature (Anderson and Christensen 1988; Jopony and Young 1994). This results in a prediction of plant metal uptake from soil pH and total soil metal content, $[M_{soil}]$. Similarly, heavy metal uptake by plants has been modeled by

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incorporating bioavailability into transfer models to improve final predictions of metal concentrations in plant tissues. Free metal ion activity (M^{2+}) is often considered to be the most bioavailable species (Sauvé et al. 1998), so CR expressions relating the free ion activity of Cd, Cu, Pb, Ni, and Zn in the soil solution to the metal concentration in crop plants have been developed (Hough et al. 2003).

Risk assessment strategies are often aimed at population subgroups. It is common practice to identify vulnerable people in society, such as young children or the elderly, and assess potential risks to the health of these population subgroups [Dudka and Miller 1999; Government/Research Councils Initiative on Risk Assessment and Toxicology (GRCIRAT) 1999]. Ryan and Chaney (1995) considered young children to be highly exposed individuals (HEIs). Thus risk assessment can usefully focus on highly exposed subpopulations on the basis that if the risk to the HEI is acceptable then most of the population is protected.

In this study we assessed the potential risk to population subgroups living on and consuming vegetables grown on a large urban site located in the West Midlands of England, United Kingdom. Since the 19th century, when rich coal deposits enabled the development of a thriving steel industry, the area has been one of the United Kingdom's premier industrial regions. During the 20th century the area became a center for the manufacturing and chemical industries. This study was primarily concerned with dietary exposure to heavy metals of subpopulations residing on this site, and has focused on those metals for which full information was available (Cd, Cu, Pb, Ni, and Zn).

The potential cancer risk from Cd and Ni has not been addressed directly, because these risks are largely associated with occupational inhalation exposure (Goyer and Clarkson 2001). Also, cancer slope factors for these metals remain controversial (Waalkes and Rehm 1994) and have not been adopted in the United Kingdom at present (DEFRA and Environment Agency 2002b). Instead, index doses (IDs) are used in the United Kingdom which represent the level at which risk to cancer is minimal (DEFRA and Environment Agency 2002b). These IDs are derived from occupational studies and relate to exposures rarely experienced in a nonoccupational setting.

Uptake of Cd, Cu, Ni, Pb, and Zn by a variety of vegetable crops was modeled using the soil characteristics most likely to control bioavailability and commonly reported in geochemical surveys and experimental data sets ($[M_{soil}]$, pH, and organic carbon). This provided a model that could be used to predict heavy metal concentrations in home-grown vegetables, $[M_{plant}]$, from typical site-specific data sets. The contribution made to dietary metal inputs from commercially produced foodstuffs was derived from dietary survey data (DEFRA 1999). Dietary data were adjusted according to the quantities consumed by the population subgroups of interest. Soil ingestion and dust inhalation data were derived for each population subgroup using a simple activity budget based on respiratory rates. The risk assessment was applied to data from a geochemical survey of the urban conurbation used in this study. The potential risks of living within, and consuming vegetables from, the urban area were calculated for three population subgroups: the average person, the highly exposed person, and the highly exposed infant. These data were used to construct "risk maps" for the urban area.

Materials and Methods

Literature database of metal uptake studies. A large database of heavy metal uptake by plants was collated from literature studies. The minimum requirements for inclusion in the database were "reliable" values of $[M_{soil}]$, pH, soil organic carbon and corresponding $[M_{plant}]$ measurements. Data were considered reliable if they were derived from a survey of cultivated plants or from trials in which plants were grown under field conditions, where the growth media were contaminated and/or urban soils. This condition was important because growing conditions more closely represented those used in the production of homegrown vegetables. Data from field-based trials were derived from a large number of studies, the details of which are described by Hough (2002). A significant proportion of this data (45%) was derived from a large study conducted by Moir (1992), which compiled metal concentrations in vegetables (broccoli, cabbage, carrot, lettuce, parsnip, potato, and spinach) grown on U.K. urban allotments. Ten study sites were chosen in Birmingham, Brighton, Darlington, Guildford, Hammersmith and Fulham (London), Leeds, Nottingham, Richmond, Shrewsbury, and York. Vegetable crops were harvested between July 1987 and January 1988, and corresponding soil samples taken for analysis. Soil pH ranged from 6.08 to 7.90 (1:2.5 soil:water ratio) and "loss on ignition" (%LOI) from 5.47% to 15.5%. Vegetable samples were digested in boiling nitric acid (HNO_3) and assayed for Cd, Cu, Ni, Pb, and Zn.

The final database provided 3,621 data points, with over 650 data points for each metal studied. The full data set covered 19 different fruit, vegetable, and arable crops. However, for this study, data for the main vegetables grown domestically in the United Kingdom were examined: broccoli, cabbage, carrot, lettuce, parsnip, potato, radish, spinach, and tomato. A summary of the complete data set showing the ranges of the key variables is presented by Hough (2002).

Model development. Models based on a pH-dependent Freundlich relation can be used to describe metal solubility in soils (Hough et al. 2003; Jopony and Young 1994). This approach can be used to predict free metal ion activity in the soil pore water (M^{2+}) from total soil metal content which is assumed to be adsorbed on humus, $[M_C]$, (milligrams of a specific metal per kilogram of soil organic carbon) and soil pH:

$$p\left(M^{2+}\right) = \frac{p\left[M_c\right] + k_1 + k_2 pH}{n_F}, \qquad [1]$$

where k_1 and k_2 are empiric, metal-specific constants and n_F is the power term from the Freundlich equation.

Metal uptake by vegetables is often characterized by a soil-to-plant concentration ratio, *CR*. This concept may be adapted to describe the quotient of metal concentration in the plant $[M_{plant}]$ (milligrams per kilogram) to metal ion activity in soil pore water (M^{2+}) (moles per liter) derived from Equation 1:

$$CR = \log \frac{\left[M_{plant}\right]}{\left(M^{2+}\right)} \qquad [2]$$

Equations 1 and 2 were combined into a single expression relating $[M_{plant}]$ to pH and $[M_C]$ (Equation 3):

$$\log\left[M_{plant}\right] = C + \beta_1\left[pH\right] + \beta_2 \log\left[M_c\right], \qquad [3]$$

where C, β_1, and β_2 are empiric metal- and vegetable-specific coefficients.

The use of $[M_C]$ in Equation 3 requires values for organic carbon content (% C). Of the 38 studies used in the database, 15 (66.2% of the data) report only measured values for loss on ignition (%LOI). Therefore, values of % LOI were converted to %C by assuming %C = 0.58 LOI (Rowell 1997). This assumption may lead to overestimation of soil carbon content at small values of %LOI due to losses of hygroscopic water in clay during the assay of %LOI. However, studies of the relation between %C and %LOI (e.g., Howard and Howard 1990; Wang et al. 1996) have provided only soil-specific conversion equations that cannot be applied generically.

To use all the available data, other approximations had to be made. Where data were derived from studies of sludge application to soils with metal loadings expressed in kilograms per hectare (e.g., Keefer et al. 1986), the assumption of a 0.20-m depth of root zone (in accord with the soil sampling depth used to derive the geochemical survey data used in this study) and a soil dry bulk density of 1.25 g/cm^3 were applied to convert metal concentrations in the sludge to milligrams per kilogram. Also, where $[M_{plant}]$ values were reported on a fresh weight basis (e.g., Jinadasa et al. 1997), these were converted to dry weight using mean vegetable water contents published by Duckworth (1966).

Equations predicting metal uptake were parameterized for nine vegetables (broccoli, carrot, cabbage, lettuce, parsnip, potato, radish, spinach, and tomato) for each of the five metals (Equation 3). The equations were validated using a generalized cross-validation approach (Shao 1993) to assess whether sufficient data had been used. This procedure involves leaving *nv* observations out of the full parameterization data set (*n*), parameterizing the model using the reduced data set and estimating the model prediction error [cross-validated residual standard deviation (RSD_{cv})] using the omitted data.

There are $\binom{n}{nv}$ combinations for leaving *nv* data points out of the parameterization data set *n*, so Monte Carlo cross-validation was undertaken with $nv = 2, 4, 8, 16, 32, 64, \ldots, n$. For each value of *nv* the random selection of the omitted data values was repeated 10^5 times. Checks were undertaken to ensure that this adequately sampled the $\binom{n}{nv}$ possible data selections. The RSD_{cv} was calculated using the omitted data generated by each iteration. Values of RSD_{cv} were compared to the prediction error returned from the full parameterization data set (RSD). A low value of RSD suggests that the model is a good fit. The value of RSD_{cv} is inherently greater than RSD because in this case a theoretically independent data set has been used to parameterize the model. Thus the closer the value of RSD_{cv} is to RSD, the more robust the model is. As *nv* increases, the difference between RSD_{cv} and RSD should reduce until an optimal *nv* is reached. At this point it could be concluded that sufficient data have been used to parameterize the model.

Dose–response assessment and risk characterization. Risk may be characterized using a hazard quotient (HQ). This is the ratio of the average daily dose (ADD; milligrams per kilogram per day) of a chemical to a reference dose (RfD, milligrams per kilogram per day) defined as the maximum tolerable daily intake of a specific metal that does not result in any deleterious health effects:

$$HQ = \frac{ADD}{RfD}. \qquad [4]$$

If HQ > 1.00, then the ADD of a particular metal exceeds the RfD, indicating that there is a potential risk associated with that metal. In the United Kingdom, DEFRA and the Environment Agency have published reference doses for Cd and Ni (DEFRA and Environment Agency 2002d, 2002e). Reference doses for Cu, Pb, and Zn were derived using the framework recommended for U.K. risk assessments (DEFRA and Environment Agency 2002b). Index doses (to assess cancer risk) for Cd and Ni were not employed because they are set at a level similar to background concentrations and because they were derived for compounds of Cd and Ni, which are rarely present in a nonoccupational environment and are difficult to assess from measures of total metal concentration. The RfDs used in this study are given in Table 1. Although U.K. policy is moving away from this, most RfDs still use experimental data using laboratory animals in their derivation. For Cd, the RfD is derived from epidemiologic data using human subjects. Different levels of uncertainty are therefore associated with the different RfD values. The RfDs may be derived from a no observed adverse effect level (NOAEL) or a lowest observed adverse effects level (LOAEL) which could lead to inconsistencies in the final RfD estimation. Essential elements (Cu and Zn in this study) have greater RfD values because of their lower toxicity.

Home-produced vegetables. The uptake models were used to estimate concentrations of Cu, Cd, Ni, Pb, and Zn in home-produced vegetables using data from the British Geological Survey's (BGS) baseline geochemical database (GBASE) for an urban area in the West Midlands, United Kingdom. Data for the consumption of home-produced vegetables (broccoli, carrot, cabbage, lettuce, parsnip, potato, radish, spinach, and tomato) distributed for the population subgroups were determined from dietary surveys (Gregory et al. 1990, 1995; Konz et al. 1989). The average person population subgroup was considered to consume the mean quantity of home-grown vegetables. The highly exposed person and the highly exposed infant population subgroups were considered to be the 95th percentile consumers of home-grown vegetables for their age class. The contribution to ADD from home-grown vegetables was calculated from the model predictions combined with the dietary consumption data for each population subgroup. The HQ was then calculated across the entire urban conurbation.

Commercially produced foodstuffs. The contributions to metals in the diet from commercially produced foodstuffs were calculated using data from the U.K. Total Diet Survey (DEFRA 1999). Food and beverage consumption was distributed according to age and body weight (Table 2). The ADDs for all five metals were determined, and the HQ calculated.

Dust inhalation. For dust inhalation, the contribution to ADD was calculated as shown in Equation 5 (Konz et al. 1989):

$$ADD = \left(\frac{M_i R_i}{B}\right) F_{ex,} \qquad [5]$$

Table 1. Metal reference doses (RfD) and index doses (ID).

Metal	Critical effect (reference)	Experimental doses[a]	RfD (mg/kg/day)
Cd	Significant proteinuria human studies involving chronic exposures (WHO 2001)	NOAEL (food): 0.01 mg/kg/day	0.01×10^{-2}
Cu	Increased protein droplets in epithelial cells of the proximal convoluted tubules of rats (Hérbert 1993)	LOAEL: 10 mg/kg/day	0.04×10
Ni	Decreased body and organ weights (Ambrose et al. 1976)	NOAEL: 5 mg/kg/day	0.05×10^{-1}
Pb`	Inhibition of ferrochelatase resulting in accumulation of eryhrocyte protoporphyrin (Mushak et al. 1989)	NOAEL: 25 µg/dL	0.35×10^{-3}
Zn	47% decrease in erythrocyte superoxide dismutase concentration in adult females (Yadrick et al. 1989)	LOAEL: 59.3 mg/kg/day	1.00×10

WHO, World Health Organization.
[a]RfD values may be based on a NOAEL or a LOAEL.

Table 2. Activity budgets used in this study for the three population subgroups: average person, 95th percentile person, and the highly exposed infant.

Activity budget		Average person	95th Percentile person	Highly exposed infant
Age (years)		39.5	38.0	2.00
Weight (kg)		74.7	74.7	11.0
Activity (min/day)				
Location [ventilation (m^3/day)]				
Indoors	10.8	1,020	1,020	1,080
	32.3	180	180	180
Outdoors	10.8	60	120	120
	32.3	90	120	120
Soil ingestion (mg/kg/day)	0.80	0.94	9.11	

where M_i is the inhaled metal concentration (micrograms per cubic meter), R_i is the inhalation rate (cubic meters per day), B is the body weight of the exposed subject (kilograms), and F_{ex} is the fractional exposure (defined as the ratio of the exposure duration to an averaging time).

The inhaled contaminant concentration, M_i, was calculated using the method employed in *Risk Assistant* v1.1 (Hampshire Research Institute 1995):

$$M_i = DM_{PM}F_{PM}F_{cont}, \qquad [6]$$

where D is the concentration of dust in the air (assumed 70.0 μg/m^3 which is the annual mean dust concentration for the United States; Hampshire Research Institute 1995), M_{PM} is the contaminated concentration on airborne particulate matter (assumed equal to [M_{soil}] where dust is derived from the soil), F_{PM} is the proportion of particulate matter that is respirable [assumed PM_{10} ~ 73% as 27% of airborne particles are generally found to be > 10 μm in equivalent diameter (Wark et al. (1998)] and F_{cont} is the fraction of dust that is derived from the contaminated source. For time spent outdoors, F_{cont} was assumed to be equal to [M_{soil}]; when indoors, F_{cont} = 0.445[M_{soil}] because 44.5% of indoor dust is considered to be derived from outdoor soil (Trowbridge and Burmaster 1997).

Dust inhalation was distributed according to a simple activity budget (DEFRA and Environment Agency 2002a). This combined the level of activity and the location of the activity, i.e., indoors or outdoors. The amount of activity a person does, and how strenuous the activity determines inhalation rate, R_i (Equation 5). Activity budgets combined with specific inhalation rates are displayed in Table 2 for average person, highly exposed person, and highly exposed infant. Body weights (B; Equation 5) for these individuals are also shown (Table 2). An exposure duration of 365.25 days was used; as in all noncarcinogenic studies, this was equal to the averaging time.

Soil ingestion. For soil ingestion, the ADD was calculated as shown in Equation 7 (Konz et al. 1989):

$$ADD = \left(\frac{M_{soil}R_{in}}{B}\right)F_{ex}, \qquad [7]$$

where R_{in} is the soil ingestion rate (kilograms per day). Ingestion rates (R_{in}) used in Equation 7 were those employed in the CLEA model (DEFRA and Environment Agency 2002a). These were derived from literature studies where tracers are used to estimate soil ingestion (Calabrese et al. 1997; Stanek et al. 2001) and are displayed in Table 2. Due to the nature of tracer studies, the soil ingestion estimates were assumed to incorporate all soil, including that adhered to vegetables. A small percentage of young children will consume much larger quantities than the mean values presented in Table 2. However, in this study the "pica" child (Calabrese et al. 1999) has not been included. Values for B and F_{ex} were the same as for Equation 5. Average daily doses from soil ingestion were calculated for the three population subgroups and for all five metals. Values of HQ were subsequently calculated.

For each metal, the HQ from dietary sources (both commercial and home produced), dust inhalation and soil ingestion were aggregated (Equation 8). Standard risk assessment procedure is to evaluate the sum of HQ for the individual metals to provide a hazard index (HI; Hampshire Research Institute 1995). This is a controversial methodology, and is not considered in the United Kingdom unless the chemicals of interest act on the same organ in the body. In this study however, the HI method has been used to provide an aggregated estimate of risk (Equations 8 and 9):

$$HQ_M = HQ'_{veg} + HQ'_{soil} + HQ'_{inhale} + HQ'_{diet}, \qquad [8]$$

Table 3. Parameter values for Cd (Equation 3).

Vegetable	*n*	*C*	β1	β2	RSD	RSD$_{CV}$
Broccoli	84	$-2.31 \times 10^{-2} \pm 0.32$	$-1.53 \times 10^{-1} \pm 0.04$	$2.94 \times 10^{-1} \pm 0.05$	0.21	0.21
Cabbage	107	$-6.11 \times 10^{-1} \pm 0.22$	$-6.23 \times 10^{-12} \pm 0.03$	$1.20 \times 10^{-1} \pm 0.05$	0.26	0.27
Carrot	126	$1.24 \times 10^{0} \pm 0.26$	$-3.11 \times 10^{-1} \pm 0.04$	$3.55 \times 10^{-1} \pm 0.07$	0.32	0.33
Lettuce	88	$2.02 \times 10^{0} \pm 0.27$	$-3.59 \times 10^{-1} \pm 0.04$	$3.18 \times 10^{-1} \pm 0.06$	0.17	0.18
Parsnip	84	$6.47 \times 10^{-1} \pm 0.57$	$-2.73 \times 10^{-1} \pm 0.08$	$1.91 \times 10^{-1} \pm 0.09$	0.35	0.37
Potato	59	$-9.76 \times 10^{-1} \pm 0.25$	$-4.76 \times 10^{-2} \pm 0.04$	$4.87 \times 10^{-1} \pm 0.09$	0.35	0.36
Radish	38	$-1.37 \times 10^{0} \pm 0.45$	$6.56 \times 10^{-2} \pm 0.07$	$5.04 \times 10^{-1} \pm 0.09$	0.35	0.38
Spinach	102	$1.66 \times 10^{0} \pm 0.21$	$-2.85 \times 10^{-1} \pm 0.03$	$5.09 \times 10^{-1} \pm 0.05$	0.21	0.22
Tomato	30	$-2.07 \times 10^{0} \pm 0.32$	$2.67 \times 10^{-1} \pm 0.06$	$2.34 \times 10^{-2} \pm 0.10$	0.18	0.20

n, number of observations. *C*, β1, and β2 are the constants (± SE) from Equation 3.

Table 4. Parameter values for Cu (Equation 3).

Vegetable	*n*	*C*	β1	β2	RSD	RSD$_{CV}$
Broccoli	82	$9.21 \times 10^{-1} \pm 0.14$	$-3.15 \times 10^{-2} \pm 0.02$	$6.52 \times 10^{-2} \pm 0.02$	0.08	0.09
Cabbage	90	$8.35 \times 10^{-1} \pm 0.14$	$-1.60 \times 10^{-2} \pm 0.02$	$-2.76 \times 10^{-2} \pm 0.01$	0.09	0.10
Carrot	94	$8.54 \times 10^{-1} \pm 0.17$	$-4.04 \times 10^{-2} \pm 0.02$	$9.35 \times 10^{-2} \pm 0.01$	0.11	0.11
Lettuce	82	$1.26 \times 10^{0} \pm 0.14$	$-2.28 \times 10^{-2} \pm 0.02$	$-1.53 \times 10^{-2} \pm 0.02$	0.10	0.10
Parsnip	84	$7.92 \times 10^{-1} \pm 0.15$	$-2.21 \times 10^{-2} \pm 0.02$	$8.65 \times 10^{-2} \pm 0.02$	0.09	0.09
Potato	31	$1.34 \times 10^{0} \pm 0.38$	$-9.32 \times 10^{-2} \pm 0.05$	$7.72 \times 10^{-2} \pm 0.06$	0.15	0.16
Radish	16	$-2.83 \times 10^{0} \pm 0.57$	$4.16 \times 10^{-1} \pm 0.08$	$3.10 \times 10^{-1} \pm 0.07$	0.18	0.20
Spinach	80	$1.10 \times 10^{0} \pm 0.13$	$1.17 \times 10^{-2} \pm 0.02$	$-2.01 \times 10^{-2} \pm 0.02$	0.08	0.08
Tomato	24	$8.92 \times 10^{-1} \pm 0.37$	$2.36 \times 10^{-2} \pm 0.04$	$8.60 \times 10^{-3} \pm 0.03$	0.05	0.05

n, number of observations. *C*, β1, and β2 are the constants (± SE) from Equation 3.

Table 5. Parameter values for Ni (Equation 3).

Vegetable	*n*	*C*	β1	β2	RSD	RSD$_{CV}$
Broccoli	82	$1.60 \times 10^{0} \pm 0.34$	$-2.60 \times 10^{-1} \pm 0.05$	$1.97 \times 10^{-1} \pm 0.05$	0.21	0.22
Cabbage	90	$1.24 \times 10^{0} \pm 0.37$	$-2.28 \times 10^{-1} \pm 0.05$	$2.52 \times 10^{-1} \pm 0.04$	0.25	0.27
Carrot	94	$1.23 \times 10^{0} \pm 0.50$	$-2.36 \times 10^{-1} \pm 0.07$	$1.59 \times 10^{-1} \pm 0.04$	0.33	0.34
Lettuce	78	$1.31 \times 10^{0} \pm 0.33$	$-2.01 \times 10^{-1} \pm 0.05$	$6.51 \times 10^{-2} \pm 0.05$	0.20	0.21
Parsnip	84	$3.19 \times 10^{0} \pm 0.73$	$-5.61 \times 10^{-1} \pm 0.10$	$4.66 \times 10^{-1} \pm 0.11$	0.45	0.47
Potato	31	$9.08 \times 10^{-1} \pm 0.61$	$-2.23 \times 10^{-1} \pm 0.09$	$1.50 \times 10^{-1} \pm 0.09$	0.24	0.26
Radish	16	$-3.62 \times 10^{0} \pm 1.08$	$5.14 \times 10^{-1} \pm 0.15$	$3.27 \times 10^{-1} \pm 0.06$	0.21	0.26
Spinach	80	$8.14 \times 10^{-1} \pm 0.35$	$-1.48 \times 10^{-1} \pm 0.05$	$6.58 \times 10^{-2} \pm 0.05$	0.21	0.22
Tomato	24	$-2.30 \times 10^{0} \pm 0.69$	$3.41 \times 10^{-1} \pm 0.10$	$1.41 \times 10^{-1} \pm 0.04$	0.17	0.20

n, number of observations. *C*, β1, and β2 are the constants (± SE) from Equation 3.

Table 6. Parameter values for Pb (Equation 3).

Vegetable	*n*	*C*	β1	β2	RSD	RSD$_{CV}$
Broccoli	82	$-3.22 \times 10^{-1} \pm 0.35$	$7.32 \times 10^{-2} \pm 0.05$	$-2.22 \times 10^{-2} \pm 0.05$	0.22	0.23
Cabbage	100	$-2.09 \times 10^{0} \pm 0.33$	$3.31 \times 10^{-1} \pm 0.04$	$-1.67 \times 10^{-1} \pm 0.04$	0.27	0.30
Carrot	106	$-1.38 \times 10^{0} \pm 0.36$	$1.93 \times 10^{-1} \pm 0.05$	$-7.66 \times 10^{-3} \pm 0.04$	0.32	0.33
Lettuce	80	$-9.84 \times 10^{-1} \pm 0.45$	$1.48 \times 10^{-1} \pm 0.06$	$-4.62 \times 10^{-2} \pm 0.08$	0.34	0.36
Parsnip	84	$7.01 \times 10^{-1} \pm 0.45$	$-1.11 \times 10^{-1} \pm 0.06$	$-4.36 \times 10^{-2} \pm 0.07$	0.29	0.29
Potato	42	$-3.18 \times 10^{0} \pm 0.51$	$3.67 \times 10^{-1} \pm 0.07$	$1.40 \times 10^{-1} \pm 0.05$	0.40	0.45
Radish	16	$-1.08 \times 10^{0} \pm 2.03$	$5.33 \times 10^{-2} \pm 0.27$	$3.25 \times 10^{-1} \pm 0.60$	0.39	0.50
Spinach	82	$-3.94 \times 10^{-1} \pm 0.53$	$8.84 \times 10^{-2} \pm 0.07$	$-1.75 \times 10^{-2} \pm 0.06$	0.33	0.35
Tomato	24	$-4.37 \times 10^{-1} \pm 1.15$	$-9.62 \times 10^{-2} \pm 0.15$	$3.73 \times 10^{-1} \pm 0.34$	0.29	0.32

n, number of observations. *C*, β1, and β2 are the constants (± SE) from Equation 3.

$$HI = HQ_{Cd} + HQ_{Cu} + HQ_{Ni} + HQ_{Pb} + HQ_{Zn}, \quad [9]$$

where HQ_M refers to the HQ for a specific metal, subscripts *veg*, *soil*, *inhale*, and *diet* refer to home-grown vegetables, soil ingestion, dust inhalation, and non–home-grown food, respectively. Values of HI were used to produce maps of the urban area thus showing where the greatest potential for risk was located.

Map generation. The geochemical and locational data were loaded, compiled, and merged in the BGS Geochemistry Database, held in the ORACLE relational database management system (version 8; ORACLE, Thames Valley Park, Berkshire, UK). The data can be retrieved by means of a user-friendly "front end" interrogation program.

For generating, processing, and editing the geochemical images for single elements used in this study, the concentration data were interpolated by a proprietary gridding software module developed at BGS as an add-on to the widely available public-domain NIH-Image version 1.62 program (modified by BGS; National Institutes of Health, Bethesda, MD, USA) running on a Macintosh G3 computer. The gridding algorithms used are broadly similar to, and produce a nearly identical output to, commercial gridding packages such as Northwood's Vertical Mapper for MapInfo (Northwood Geoscience Ltd. 1996). This software produced grids for each chemical element by interpolation of the data using the method of inverse distance weighting (Webster and Oliver 2001), where each grid cell (pixel) in this case represents 25 × 25 m on the ground. In this technique, each grid cell is given a calculated value derived from data for nearby sample sites (eight "neighbor" sites are used in the maps shown here). The calculation uses data from all sample sites within 1,500 m, weighted in accordance with distance (r) such that the weighting (W) is proportional to r^{-2}:

$$W = \frac{k}{r^2} \quad [10]$$

Table 7. Parameter values for Zn (Equation 3).

Vegetable	*n*	*C*	β_1	β_2	RSD	RSD_{CV}
Broccoli	82	$1.87 \times 10^0 \pm 0.23$	$-1.50 \times 10^{-1} \pm 0.02$	$2.87 \times 10^{-1} \pm 0.05$	0.11	0.11
Cabbage	106	$2.28 \times 10^0 \pm 0.38$	$-2.17 \times 10^{-1} \pm 0.03$	$2.63 \times 10^{-1} \pm 0.07$	0.23	0.24
Carrot	92	$2.13 \times 10^0 \pm 0.32$	$-2.82 \times 10^{-1} \pm 0.04$	$3.63 \times 10^{-1} \pm 0.07$	0.17	0.17
Lettuce	82	$2.21 \times 10^0 \pm 0.21$	$-1.68 \times 10^{-1} \pm 0.02$	$2.43 \times 10^{-1} \pm 0.05$	0.10	0.10
Parsnip	84	$1.44 \times 10^0 \pm 0.29$	$-1.67 \times 10^{-1} \pm 0.03$	$3.06 \times 10^{-1} \pm 0.06$	0.14	0.15
Potato	31	$1.23 \times 10^0 \pm 0.39$	$-1.14 \times 10^{-1} \pm 0.05$	$2.24 \times 10^{-1} \pm 0.09$	0.13	0.15
Radish	36	$6.97 \times 10^{-1} \pm 0.48$	$4.75 \times 10^{-2} \pm 0.08$	$2.11 \times 10^{-1} \pm 0.08$	0.26	0.28
Spinach	80	$2.00 \times 10^0 \pm 0.35$	$-1.69 \times 10^{-1} \pm 0.04$	$4.14 \times 10^{-1} \pm 0.08$	0.16	0.17
Tomato	24	$1.95 \times 10^0 \pm 0.84$	$-4.43 \times 10^{-2} \pm 0.20$	$-1.43 \times 10^{-2} \pm 0.22$	0.16	0.18

n, number of observations. *C*, β_1, and β_2 are the constants (± SE) from Equation 3.

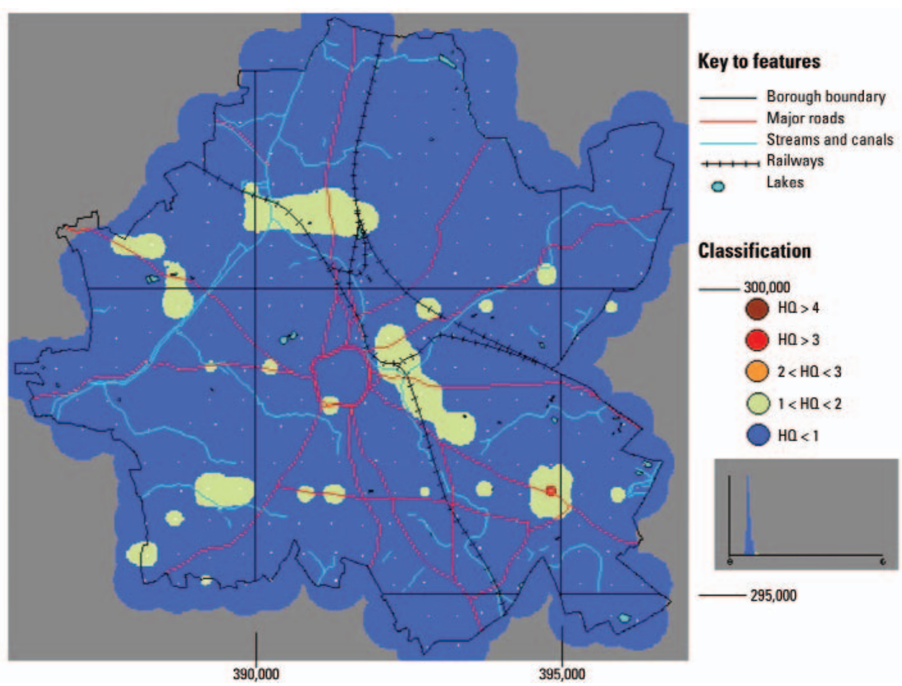

Figure 1. Maps of HI values for the average person population subgroup for an urban conurbation in the West Midlands, United Kingdom. Units are British National Grid eastings and northings in meters.

In areas where it had not been possible to collect samples at a density at or close to the normal sample density of the survey, a gridded image can give misleading results. Accordingly gridded areas which fell more than 2,000 m from a sample point were zone-blanked—set to null (no value) after interpolation and before classification. This has the additional effect of removing overextrapolated data from the percentile classification, allowing a more accurate fit with the data distribution of the real data. In this case study, zone-blanked areas account for < 2% of the area, and some overshoot beyond the borough boundary has been allowed. Clearly in the absence of data, contaminant risk in these areas cannot be assessed.

The gridded data were then used to produce percentile-based color-classified images. Most of the images used in the BGS geochemical atlases use standard class interval boundaries set at the 5th, 10th, 15th, 25th, 50th, 75th, 90th, 95th, and 99th percentile levels. However, because of the special requirements of the maps shown here, which need particular data-level boundaries to be emphasized (HI = 1, 2, 3), each data distribution was statistically analyzed to determine the required percentile transitions, which were then fed back into the classification section of the program. An appropriate color scheme was then applied to the resolved classes.

The raster maps produced by these methods were imported into a geographic information system (GIS) environment using MapInfo (version 4; MapInfo Corp., Troy, NY, USA) and georegistered so that other spatial information such as geology and topographic features could be mapped accurately with the geochemical data.

Results and Discussion

Plant metal concentration [M_{plant}]. Regression analysis of [M_{plant}] against pH and [M_C] (Equation 3) provided good estimates of uptake for Cd, Cu, Ni, and Zn by all vegetables, with less satisfactory results for Pb. Tables 3–7 present the regression estimates of C, β_1, and β_2 (Equation 3) together with the residual standard deviation (RSD; $\log_{10}$ milligrams per kilogram). For Cd, Cu, Ni, and Zn the prediction RSDs were generally low. For example, for Cu the RSD ranged from 0.05 (tomato, n = 24) to 0.21 (radish, n = 21), with a mean RSD of 0.11 on a $\log_{10}$ scale. However, for Pb (Table 6) the RSD ranged from 0.22 (broccoli, n = 82) to 0.56 (cabbage, n = 116), with a mean RSD of 0.35 on a $\log_{10}$ scale. The reason for this may be that uptake of Pb by vegetables is relatively small compared with Pb concentrations in the local soil and dust; the most significant source of lead contamination of vegetables is atmospheric deposition (Dalenberg and Van Driel 1990). Thus, predicting concentrations of Pb in plants from soil characteristics is difficult, especially if plant samples are not thoroughly washed before analysis. In general, the RSDs achieved for root vegetables and protected vegetables, such as tomatoes, were smaller than the RSDs achieved for larger leafy vegetables such as cabbage.

The RSD_{cv} was calculated for all 45 transfer models (Tables 4–8). This is an estimate of the model uncertainty when it is applied to a new data set. In all cases, values of RSD_{cv} were close to but greater than the regression RSD. This suggests that the uptake models are robust and can be applied generically.

Risk assessment. Figures 1, 2, and 3 show the HI maps for the average person, the highly exposed person, and the highly exposed infant population subgroups for the urban conurbation used in this study. For the average person population subgroup (Figure 1), most (89%) of the urban area provides an HI < 1.0; only 11% provides an HI > 1.0 and 2.0. The average HI for the entire population subgroup was 0.83. These results suggest that most of this population subgroup will not experience any form of deleterious health effects from living within and consuming vegetables from this urban location.

For the highly exposed person population subgroup (Figure 2), 44% of the urban area provides an HI between 1.0 and 2.0, 52% of the urban area provides an HI between 2.0 and 3.0, and 4.2% have an HI > 3.0. The average HI for the entire population subgroup was 2.13.

For the highly exposed infant population subgroup (Figure 3), most (52%) of the urban area provides an HI between 2.0 and 3.0. Only 3.9% of the urban area provides an HI < 2.0, with 30% providing an HI between 3.0 and 4.0. Fourteen percent of the urban area provided an HI > 4.0. The average HI for the entire population subgroup was 3.22.

Caution is required when interpreting the results of this form of risk assessment. Although an HI > 1.0 suggests that a person may experience adverse health effects during his or her lifetime, the HI is a highly conservative index and relates to very minor biologic responses (Teuschler et al. 1999). The RfD values (Equation 4) used to determine HQ are based almost exclusively on toxicologic tests of the metals on animals (GRCIRAT 1999). The final RfDs are determined using uncertainty factors for the extrapolation of the results to humans (GRCIRAT 1999). The HI is a relative index and although it can be used to identify population subgroups that potentially are at higher risk, it indicates only the relative severity of those risks. However, an HI > 1.0 is still considered undesirable when looking at the overall health of a population or population subgroup.

The risk assessment applied to the study site did not take into account "suitable for use" policy. In this study, we assumed that the land use would all be the same—i.e., housing with gardens. Obviously there may be areas within the urban conurbation which are not suitable for housing because of other land use pressures. In practice, if the more contaminated areas were converted to residential use, then uncontaminated soil may be imported. Also where low soil pH contributes to high metal uptake rates, then, in practice, liming would substantially correct this problem if the land use were converted to domestic gardens.

The proportion of the HI that was attributable to the different metals used in this study did not vary among population subgroups. The largest contribution to HI was from Pb (about 40% of HI) and Cd (about 30% of HI). Ni and Cu provided the lowest contribution to HI at about 10 and 14%, respectively. The proportion of the HI attributable to different exposure

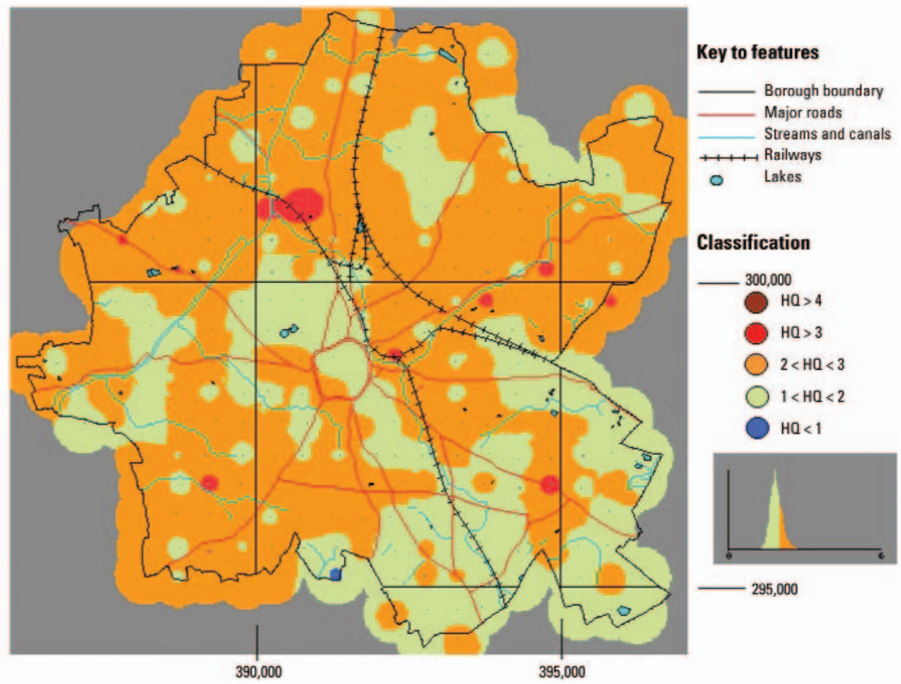

Figure 2. Maps of HI values for the highly exposed person population subgroup for an urban conurbation in the West Midlands, United Kingdom. Units are British National Grid eastings and northings in meters.

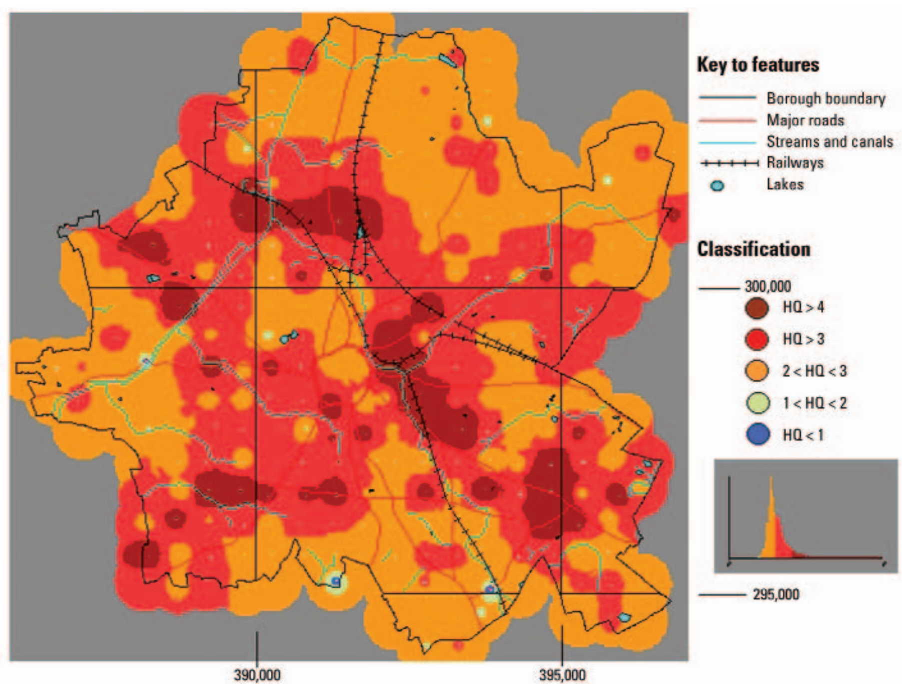

Figure 3. Maps of HI values for the highly exposed infant population subgroup for an urban conurbation in the West Midlands, United Kingdom. Units are British National Grid eastings and northings in meters.

pathways varied between the population subgroups. In all cases most HI was attributable to dietary exposure (average person 94%, highly exposed person 86%, highly exposed infant 73% of the HI). The contribution to the HI from soil ingestion was largest for the highly exposed infant (22% of the HI). The contribution from dust inhalation exposure was greatest for the highly exposed person (8% of the HI) because this population subgroup is more likely to experience occupational exposure.

Conclusions

Risk assessment models must be fairly generic to satisfy a wide range of applications. The simple uptake models described in this article provide species-specific prediction at a similar generic level to existing models such as CLEA. Final predictions of Cd, Cu, Ni, and Zn content of food crops were reasonable, although the models for Pb uptake were less useful. Cross-validation has shown that sufficient data has been used to parameterize the models and give prediction errors close to the regression RSD.

The HI maps (Figures 1, 2, and 3) of the urban study area show that HI estimates can vary considerably with geochemical parameters. As a result, not all sites within the study area may be suitable for housing or allotments without remediation. Most of the population have an HI < 1.0 on the majority of sites within the urban area, although certain "hot spots" still pose some potential health threat. Most high HIs are located around junctions of major roads, railways, and canals, all of which are considered major sources of potentially toxic metals in the urban environment. These maps highlight the importance of a site-specific approach to urban redevelopment and also the benefit of the "suitable for use" approach to development. Regardless of the HI maps, more detailed site surveys would always be required when assessing a specific site within this urban area, because the spatial data used to construct the HI maps are of fairly low resolution.

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