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Risk Analysis of a Farm Area Near a Lead- and Cadmium-Contaminated Industrial Site

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The Asua Valley is an area on the outskirts of Bilbao where industry and small farms still coexist despite decades of serious environmental pollution. The present study was carried out to estimate the risk to which the residents of the area are exposed as a result of soil/dust ingestion and consumption of locally grown fresh produce, and, on the basis of this data, to delimit the areas that might require environmental clean-up. The relation between lead and cadmium content in soil and plant samples was assessed by multiple linear regression. The level of soil lead content for proposing intervention was determined by assessing the exposure of young children due to soil ingestion, assuming a "central-estimate" ingestion rate of 110 mg/day. Vegetable sampling was stratified according to the level of cadmium in the soil. The intervention content of cadmium was established as the midpoint of the soil sampling stratum previous to the one registering a vegetable consumption hazard quotient of 1; in this interval the intake reached 57% of the TDI.

KEY WORDS: *lead, cadmium, soil pollution, risk analysis, environmental clean-up.*

Introduction

THE Asua Valley lies on the outskirts of the Greater Bilbao Area (in the Basque Country, Northern Spain). The valley has long been the site of numerous industries with a high potential for air pollution (e.g., foundries and non-ferrous metal-working plants), coexisting side by side with small farms. Air pollution in the area was very high from the 1960s to the end of the 1980s.

Preliminary soil sampling yielded pollutant concentrations ranging between 42 and 1600 ppm for lead and between 0.7 and 54.4 ppm for cadmium. A risk assessment was carried out taking into account soil/dust ingestion and home-grown produce consumption (Cambra *et al.*, 1995). Pollutant intake calculations were made on the basis of soil results, soil-to-plant transfer factors found in the bibliography, and a local nutrition survey. The risk assessment showed that the exposure of residents in the most polluted area could be high and that there was a need for further study to assess exposure with sufficient accuracy to make intervention decisions possible.

Accordingly, there appeared to be a need for the present study for the purposes of: (1) assessing more precisely the risk to which local residents are exposed, (2) delimiting areas that might require environmental clean-up, and (3) determining whether the health status of residents in the area should be monitored.

Materials and Methods

Soil Sampling. Samples were taken at a total of 189 natural soil points covering a surface area measuring 310 Ha. Of those, 109 were planted with vegetables and other produce, and another 76 had a farmhouse or dwelling nearby. Each sample consisted of six subsamples taken at random in a 20×20 m^2 area at a depth of between 0 and 15 cm. A manual sampler was used. Lead and cadmium were determined by Plasma Emission Spectrophotometry (ICP/OES) after sample digestion under ISO/DIS 11466 conditions. To ensure methodological comparability, certified standard materials were used. Contrast analyses were performed in an accredited laboratory with satisfactory results (Ondoan, S. Coop.-IHOBE 1995).

Plant Sampling. Sampling was stratified according to the level of lead and cadmium in the soil. Gardens and cultivated areas were classified into seven groups according to Cd content (3–4, 4–5.5, 5.5–8, 8–13, 13–18, 18–23, >23 ppm), and in turn — except in the groups on either end where further classification was impossible — each one was further classified according to the lead content of the soil (<300, 300–475, 475–650, 650–825, 825–1000, 1000–1500, >1500 ppm). The plant species were divided into five groups, because it was impossible to take samples of all the species produced in the area, due to the high economic cost involved and the cost in time entailed by a sampling period that would have lasted an entire year. The groups (leafy vegetables, potatoes, cabbage, fruit, and bulbs/roots) were formed taking into account homogeneity of soil/plant transfer factors in the different species. Sampling was performed in June 1996 and samples were taken from 21 different gardens.

Vegetables were washed with tap and distilled water and Cd and Pb determinations in the edible parts of the plant were made following dry-ashing at 450°C by atomic absorption spectrophotometry with graphite furnace and Zeeman background correction system. The quantification limits were 0.005 mg/kg for Pb and 0.0005 mg/kg for Cd.

Statistical Methods. Plant content-soil content relations were studied by multiple linear regression using the SPSS 6.1.2 statistics package.

Assessment of Risk for the Resident Population. Farm resident exposure, including an estimate of Pb and Cd intake through direct soil/dust ingestion and through consumption of home-grown produce, was calculated using the following values:

(a) Soil ingestion: (1) Children of up to 15 kg body weight, 200 mg/day soil ingestion rate as the upper bound (USEPA, 1991) and 110 mg/day as the central-estimate value. This value is the mean of the average results reported in the studies reviewed (Hawley, 1985; Binder *et al.*, 1986; Clausing *et al.*, 1987; Calabrese *et al.*, 1987; La Goy, 1987; Calabrese and Staneck, 1989; Van Wijnen *et al.*, 1990; Davies and Waller, 1990; Calabrese and Staneck, 1991; Staneck *et al.*, 1995; Calabrese *et al.*, 1997). (2) Adults of up to 70 kg body weight, soil ingestion rate of 100 mg/day (USEPA, 1991). We assume an exposure frequency of 365 days/year. In order to assign to each farm a soil content value for each of the pollutants, we used the results obtained from soil samples taken within 30 m of the farmhouse.

(b) Vegetable consumption: Mean consumption values for adults taken from the Nutrition Survey conducted by the Basque Government Department of Health, 1994, per type of plant. Pulses and fruit are not grown in the area in question. We assumed that 100% of the vegetables and potatoes consumed were home grown. No statistics are available on food consumption among children in the population, so our estimates of ingestion for this age group were made using the adult statistics of consumption per kilo body weight and day. Exposure through home-grown produce was calculated, per soil pollutant interval and group of vegetables, multiplying the average pollutant content in the samples collected by the consumption of the group in question. The total intake per soil contaminant interval was calculated as the sum of the five groups of vegetables.

Water exposure was not assessed, because all the dwellings in the area receive mains water whose cadmium and lead content is under the standard detection limit for drinking water (i.e., less than 1 and 5 ppb, respectively), and because lead leaching from lead piping does not appear to be high in this area (Cirarda, 1998).

We used the tolerable daily intake (TDI) values recommended by WHO: 1 μg/kg/day of Cd (FAO/WHO, 1993) and 3.5 μg/kg/day of Pb (FAO/WHO, 1993). Risk was characterized by calculating the hazard quotients, HQ (Intake per route/Tolerable intake) and the hazard index, HI (sum of HQs) (USEPA, 1989).

RESULTS

A total of 69 plant samples were taken to determine their lead and cadmium content. These results, by vegetable group, are shown in Figures 1 to 4.

We have tested linear regression models with the pollutant content of the plant as the dependent variable, and with the pollutant content of the soil, the pH value, and the clay and organic matter content of the soil, as independent variables. We verified the conditions for application of the models: normality of residual values, homocedasticity and identification of influential (Cook distance greater than 1) and outlier values (studentized residuals greater than 2). The values of the parameters defining the statistically significant models are given in Table 1. As can be seen,

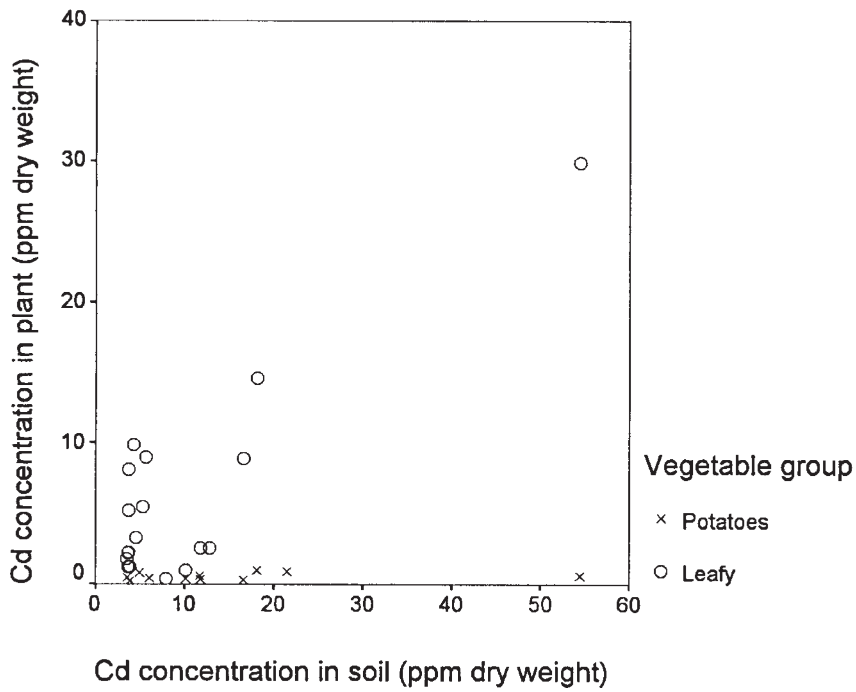

FIGURE 1

Plant and soil cadmium concentrations: leafy, potatoes.

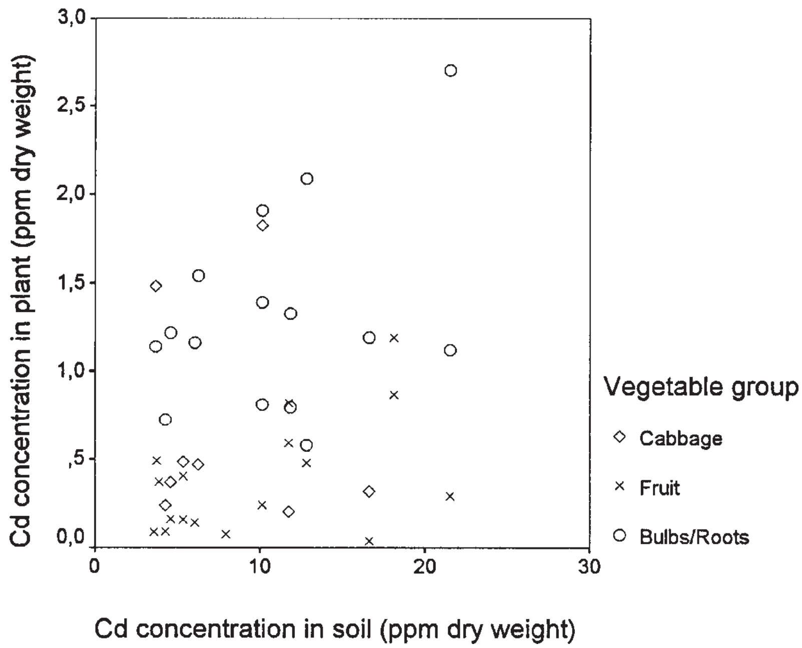

FIGURE 2

Plant and soil cadmium concentrations: cabbage, fruit, bulbs.

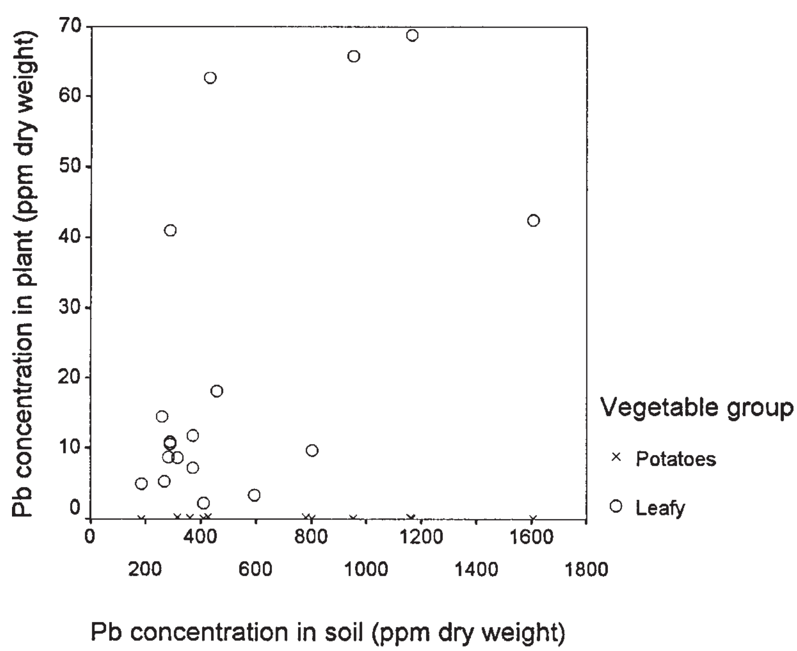

FIGURE 3

Plant and soil lead concentrations: leafy potatoes.

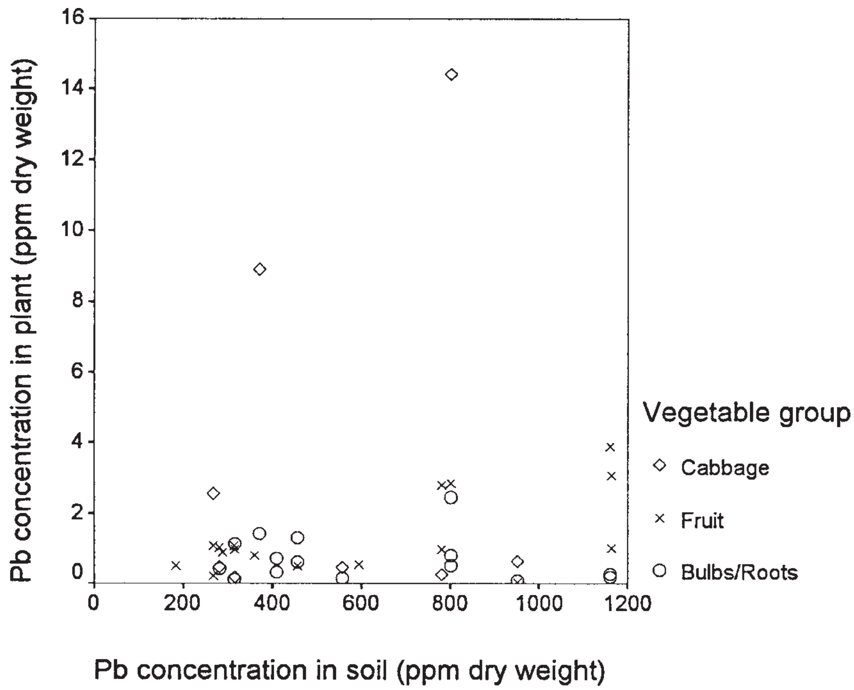

FIGURE 4

Plant and soil lead concentrations: cabbage, fruit, bulbs.

the linear models are better suited for cadmium than for lead. An outlier was found among broad beans results; elimination of the outlier improves the r^2, but does not affect the slope of the line.

Ingestion Results

Both direct dust/soil ingestion and vegetable ingestion were calculated. Soil data from 44 farms were used. Figures 5 to 8 show, per soil content interval, the hazard quotients and hazard index for the population of young children and adults, respectively, in the area. Cadmium ingestion ranged between 30 and 160% of TDI for adults and between 35 and 230% in children. In the case of lead, exposure calculations ranged between 20 and 130% of TDI in adults and 90 to 690% in children.

Discussion

The question we set out to answer was whether living in a contaminated area and consuming home-grown vegetables posed a health hazard, and, if so, what pollut-

TABLE 1
Plant Lead and Cadmium Content: Linear Regression Model Results

	GROUP/SPECIES	INDEPENDENT VARIABLES	N	r^2	$r^2_{adjusted}$	p	PARAMETERS (SE)
Cd	LEAFY	SOILCD	18	0.739	0.721	0.000	B 0.5069 (0.0755) C 1.0926 (1.158)
	CHARD	SOILCD	12	0.804	0.780	0.001	B 0.519 (0.0808) C 1.525 (1.451)
	CHARD	SOILCD OM	11	0.860	0.830	0.000	B SOILCD. 0.5331 (0.076) B OM -1.075 (0.5782) C 7.67 (3.668)
	ONIONS	SOILCD	6	0.717	0.647	0.03	B 0.1052 (0.03304) C 0.1453 (0.3955)
	ONIONS	SOILCD PH	6	0.860	0.780	0.04	B SOILCD. 0.0961 (0.0264) B PH 0.3993 (0.2134) C -2.51 (1.456)
	PEPPERS	SOILCD	4	0.978	0.968	0.01	B 0.05265 (0.005487) C 0.2279 (0.06094)
	BROAD BEANS	SOILCD	7	0.677	0.613	0.02	B SOILCD. 0.0108 (0.0033) C 0.0681 (0.03366)
Pb	PEPPERS	SOILPB	4	0.776	0.664	0.1	B 0.00222 (0.0008445) C 0.0189 (0.60864)
	BROAD BEANS	SOILPB	7	0.801	0.761	0.006	B 0.003403 (0.000759) C - 0.2643 (0.475)
	BROAD BEANS (*)	SOILPB	6	0.978	0.973	0.0002	B 0.003506 (0.00026) C - 0.11812 (0.1643)
	BROAD BEANS	SOILPB OM	7	0.896	0.844	0.01	B SOILPB 0.004021 (0.00069) B OM - 0.2868 (0.149) C 1.171 (0.8389)

SOILCD: Cadmium content of the soil
SOILPB: Soil lead concentration
OM: Soil organic matter
PH: Soil pH
B: Slope of the the regression line
C: Point of intersection on the Y axis
(*) The model parameters are arrived at by eliminating an outlier value.

ant levels would warrant environmental intervention or health measures. According to our intake calculations, suitability for growing vegetables and devoting the land to residential purposes depends, respectively, on the magnitude of exposure due to plant consumption and to dust/soil ingestion, because absorption of the metals in question through the skin may be discarded, and inhalation, under normal conditions of particle suspension from the land, is negligible compared with soil and food ingestion.

To assess the risk arising from vegetable consumption over the entire contaminated area, we used the results of lead and cadmium content in 69 vegetables grown in 21 gardens. There are two factors whose effects on the results are difficult to assess: the random error resulting from the small size of the sample, and the fact that, due to urgency of demand, all the samples were taken in June, meaning that only the vegetables grown during that month are represented, with some at a stage of development other than that at which they are normally consumed.

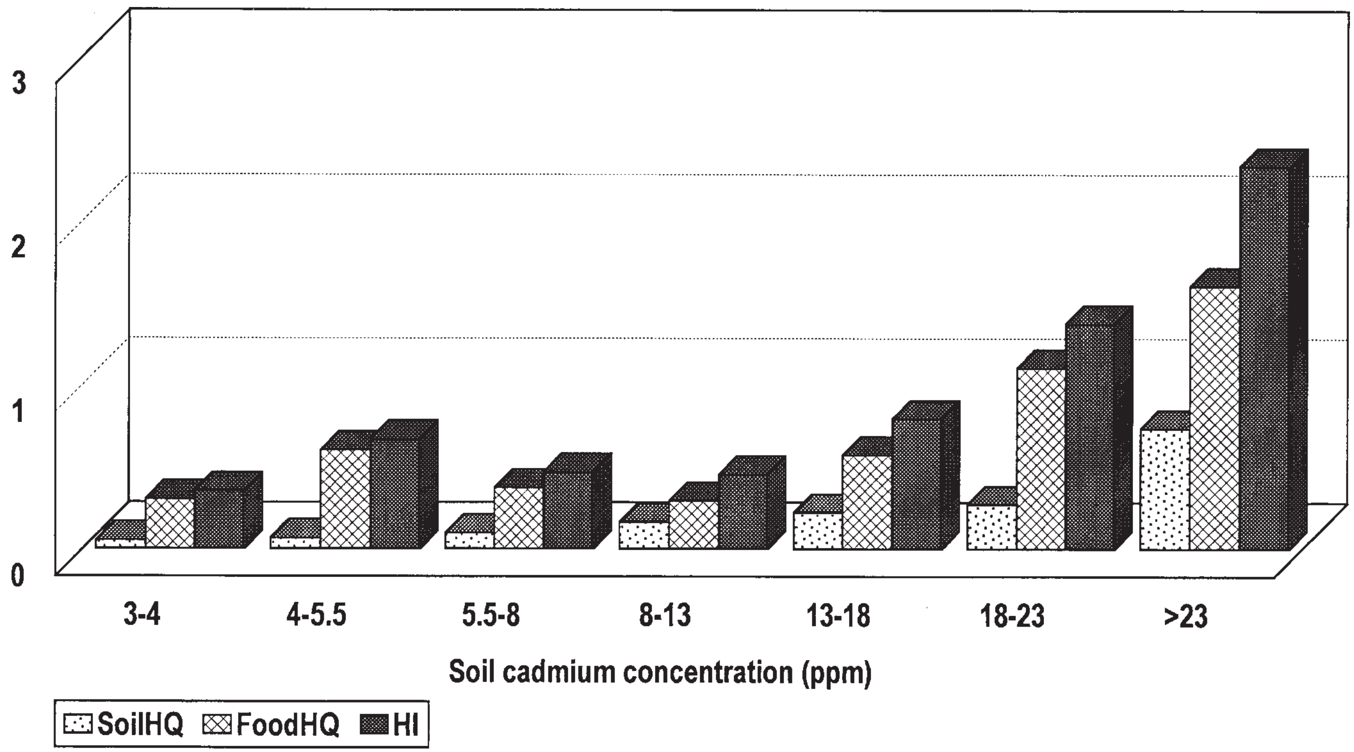

FIGURE 5

Soil and food hazard cadmium quotients and hazard index for children.

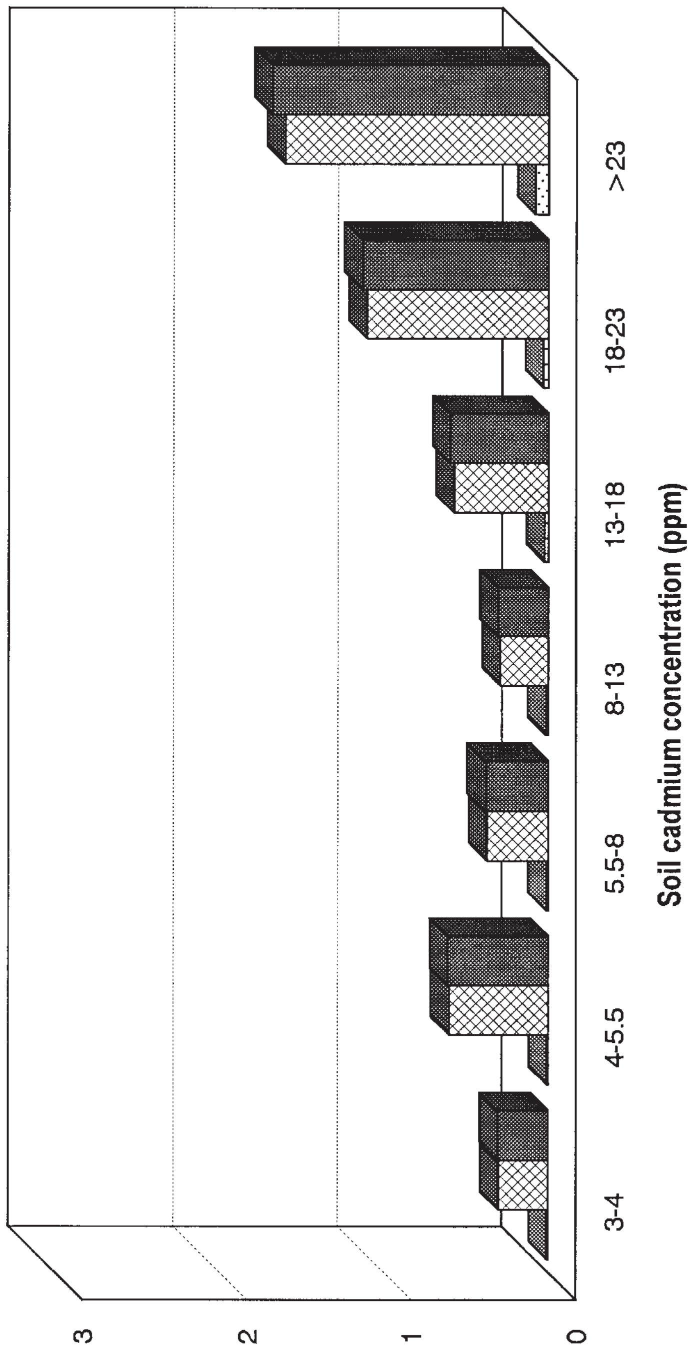

FIGURE 6

Soil and food hazard cadmium quotients and hazard index for adults.

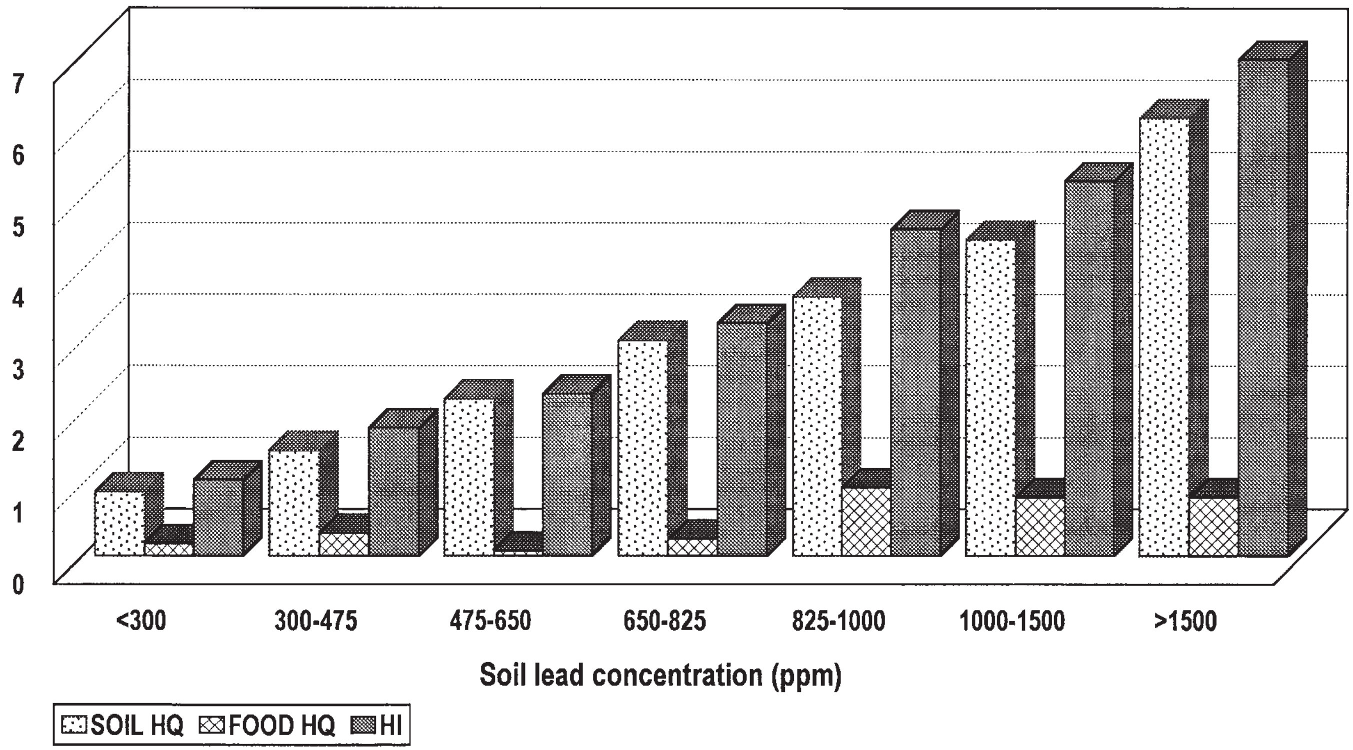

FIGURE 7

Soil and food lead hazard quotients and hazard index for children.

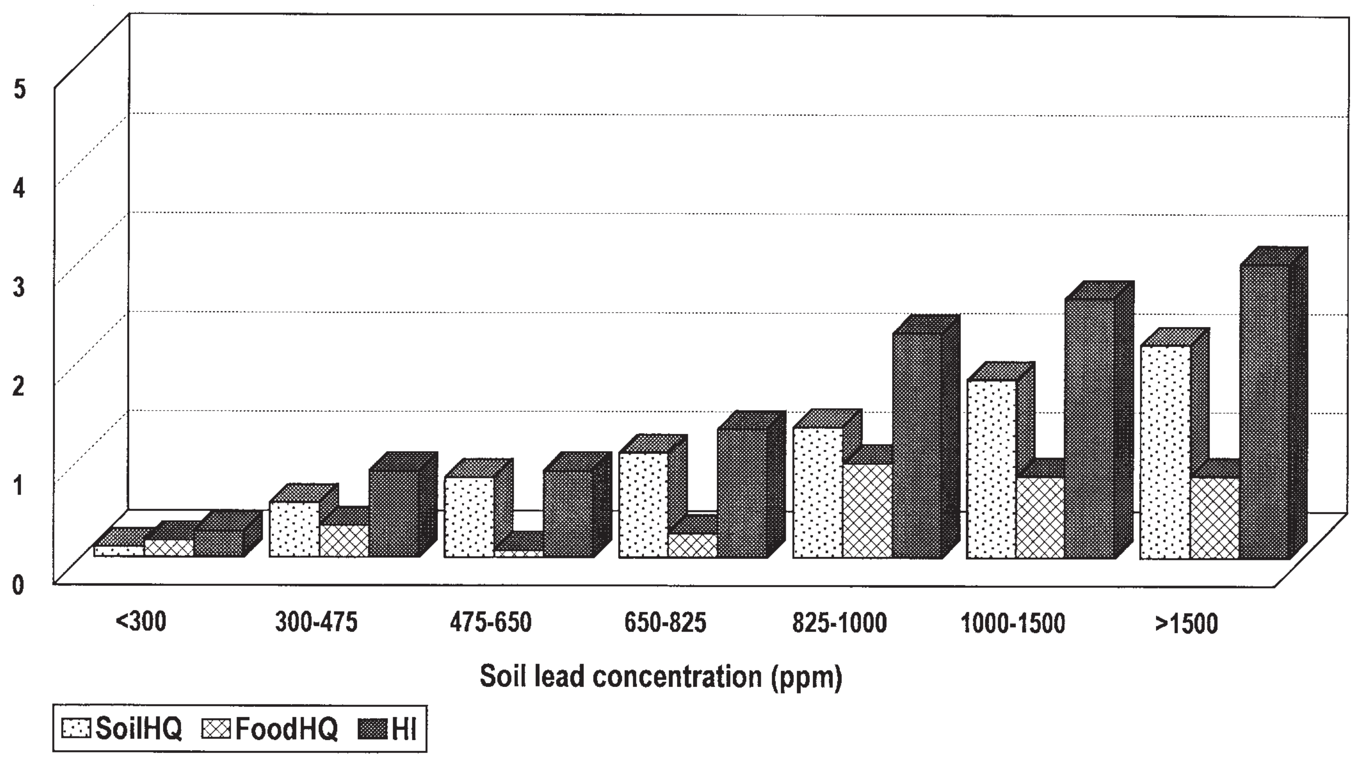

FIGURE 8

Soil and food lead hazard quotients and hazard index for adults.

In several of the plant species sampled, the linear models are well suited to characterizing the relation between levels of lead and cadmium in vegetables and in soil (Table 1). We considered that the use of regression functions is indicated in cases of a determination coefficient greater than 0.5. Draft regulation III/5125/97 of the European Commission sets a content limit of 0.1 ppm for lead in vegetables, 0.2 ppm for lead in leafy vegetables, and 0.1 ppm for cadmium in vegetables. Using these values as our plant pollution value and solving the simple linear regression equations of Table 1 for soil content, we arrive at 1 ppm cadmium for leafy vegetables, 11.4 ppm cadmium for onions, 24 ppm cadmium for green peppers, 286 ppm lead for broad beans, and 663 ppm lead for green peppers. However, the applicability of these results is questionable for several reasons. The plant lead and cadmium value utilized is taken from a piece of draft legislation and is not yet a statutory value in Spain. With respect to the models, the amount of data is small, interspecies variation is very high, and plant absorption depends on other factors besides soil pollutant concentration. In the case of leafy vegetables, which are widely consumed in the area, the model utilized is unreliable because the cadmium result obtained, for a plant concentration of 0.1 ppm, is under the lower limit of the range of soil content values introduced into the model. Nevertheless, according to the public health protection level adopted in the Commission's project, it appears that the maximum level of soil cadmium allowable for the production of leafy vegetables would be very low.

Accordingly, we followed a different approach to arrive at soil pollutant limits beyond which crop intervention should begin. In the case of cadmium, we took as critical value the midpoint of the soil content previous to the one reaching a hazard quotient of 1 for food ingestion. In the case of lead, we took as critical value that of the interval where exposure occurs from food consumption with a hazard quotient of 0.25. This difference is due to the fact that for cadmium, the predominant exposure of the general population is due to vegetable consumption, whereas with lead this is not the case, and we feel it is necessary to reserve a fraction of the TDI, which we have set at 75%, so that the vegetable-consuming population will have a safety margin to allow for other exposure pathways. The values obtained were 15 ppm Cd and 650 ppm Pb. The applicability of these values may be only local, and it should be stressed that they are critical values beyond which vegetables should not be grown. They are not maximum allowable values, as it is likely that even in soil with 15 ppm cadmium, the vegetables grown will have a higher cadmium content than recommendable for public health protection.

The suitability of a site for residential use (one-family homes) is determined by the exposure of young children to lead through direct ingestion of dust/soil. We have used two estimates: a central and an upper bound estimate. The soil lead limit for residential use obtained with the central estimate and the assumptions described in the methodology section, assigning 100% of the TDI to this pathway, was 475 ppm. This value should be considered as the limit beyond which measures must be taken to diminish lead exposure in all homes with small children, without thereby implying that dwellings with lower lead content of the soil are acceptable, because all children with above-average soil ingestion levels would very likely be above the recommended TDI. The soil lead content calculated for residential use assuming 200 mg/day soil ingestion was 262 ppm.

The 262 ppm lead value was surpassed in 37 dwellings, while 11 (approximately 14900 m^2) exceeded the lead value of 475 ppm. In the case of cadmium, 15 ppm cadmium was exceeded in 13 sampling units, of which 9 were under cultivation (18600 m^2). The areas with cadmium and lead pollution overlapped. Although the undesirable effects of these pollutants occur in different age groups (small children in the case of lead, and adults following a long period of exposure in the case of cadmium), this overlap increases the cost/benefit ratio, making clean-up operations all the more justifiable.

In the study area there is a risk of high exposure to lead and cadmium due to plant consumption, and to overexposure to lead in young children arising from the residential use of land. Therefore, it appears necessary to conduct an epidemiological study to achieve an individual and population assessment of the degree of adult exposure to cadmium and lead through consumption of vegetables grown in the most contaminated area, and exposure to lead among the children living in dwellings with soil lead levels higher than 262 ppm. People living in homes with soil lead levels higher than 475 ppm should be informed of the individual precautions that can be taken. Urgent intervention is required in gardens and cultivated areas with cadmium content of the soil exceeding 15 ppm.

The methodology followed has enabled us to delimit the contaminated area where an environmental clean-up or intervention project should begin, and has also pointed to the need for an epidemiological assessment among the area's inhabitants. This epidemiological study has already been carried out and its results will be published soon.

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