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# Lead and Cadmium Contamination Levels in Edible Vegetables

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Ever since the research of Kehoe et al. (1933), with each passing day food is being attributed with a greater role as one of the major sources in the exposure of man to lead. Lead and cadmium are found in all foods, in widely varying amounts (Reilly 1980). Vegetables absorb these metals from the ground, as well as from deposits on the parts of the vegetables exposed to air from the polluted environment. In the case of lead, the principal source of contamination in vegetables comes from the atmosphere, due to the use of alkyl-lead derivates, such as antidetonators in liquid carburants (Ganje and Page 1972). The use of residual waters for the irrigation of the vegetables, as well as the use of certain fertilizers constitute the principal source of cadmium pollution in vegetables according to Jaakkola et al. (1979). For Buchauer (1973), the air-exposed parts, and, more specifically, the leaves, are the principal entrance ways of lead and cadmium; nevertheless, Haghiri (1973) feels that the principal access is through the root.

Plants absorb lead and cadmium from the earth and atmosphere, although the differences between species are enormous (Dedolph et al. 1970). Jaakkola et al. (1979) stress the great capacity of spinach to accumulate cadmium.

The objective of this research is to reveal the level of lead and cadmium pollution in fresh vegetable samples from the Cordovan fertile lowland region of the Guadalquivir River and to establish a base level of contamination to serve as a reference point in further studies. At the same time, the possible health risks for the consumer are discussed.

## MATERIALS AND METHODS

A total of 235 vegetable samples belonging to 20 different species have been analyzed: chard (*Beta vulgaris* var. *cicla* L.);

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artichoke (Cynara Scolymus L.); garlic (Allium sativum L.); celery (Apium graveolens L.); cardoon (Cynara cardunculus L.); onion (Allium cepa L.); cauliflower (Brassica oleracea var. botrytis DC); endive (Chichorium endivia L.); spinach (Spinacea oleracea L.); green beans (Phaseolus vulgaris SAVI); lettuce (Latuca sativa L.); potato (Solanum tuberosum L.); parsley (Petroselinum sativum HOFFM); green pepper (Capsicum anuum L.); leek (Allium porrum L.); radish (Raphanus sativus L.); beetroot (Beta vulgaris var. rapa DUM); cabbage (Brassica oleracea var. capitata DC); tomato (Lycopersicon esculentum MILL); and carrot (Daucus carota var. sativa L.).

We have established five groups based on the edible parts according to the Spanish Alimentary Code (Código Alimentario Español 1967).

Group 1.- Roots and Tubercles (carrot, beetroot, radish, celery, and potato).
Group 2.- Bulbs (onion, leek, and garlic).
Group 3.- Leaves and Soft Stalks (spinach, lettuce, parsley, chard, endive, and cardoon).
Group 4.- Cabbages (different types of cabbages and cauliflower).
Group 5.- Fruits and Similar Garden Produce (tomato, pepper, green bean, and artichoke).

The different anatomical parts are considered separately in some of these products, for example, leaves, roots, stalks, pulp, skin, etc.

The vegetables were washed in fresh running water to eliminate dust, dirt, possible parasites or their eggs and then they were washed again with deionized water. The washed samples were placed on filter paper to eliminate excess moisture. Once dry, each sample was weighed (20 grams) and dried in a oven at a temperature lower than 80ºC for 24-36 hours (Torija Isasa 1982). From the dried sample, 1 gram was placed in a muffle furnace at 450ºC for 24-36 hours. The ashes were collected with a $ClH-NO_3H$ mixture at 50 %, at a 1:1 ratio and filtered in Whatman GF/C paper (Martínez Para 1979). The filtering was carried out to a volume of 5 mL with the acid mixture used and was followed by atomic absorption spectrophotometry. In the nebulization system, the impact bead was used and the sample solution was aspirated into an air-acetylene flame. The determinations were carried out on a Perkin-Elmer Model 2380 atomic-absorption spectrophotometer equipped with a lead hollow cathode lamp, operated at 283.3 nm and a cadmium hollow cathode lamp, operated at 228.8 nm.

The sensitivity obtained was 0.29 mg/L for lead and 0.019 mg/L for cadmium. The criteria of the American Chemical Society (1980) and Mottola (1984) were used to calculate the detection limit. The concentration limits (minimum detectable concentrations on the fresh weight) obtained were 52 ug/kg for lead and 6.04 ug/kg for cadmium.

The lead and cadmium concentrations in the sample, expressed in mg/kg of fresh weight, are obtained through the expression:

$$\text{Pb or Cd concentration (mg/kg)} = \frac{5 \times \text{dry weight} \times A}{a \times \text{fresh weight}}$$

where:
dry weight = weight in g of the dry sample at 80 ºC
fresh weight = weight in g of the sample before drying at 80 ºC
A = absorbance reading
a = slope of the standard curve

## RESULTS AND DISCUSSION

A summary of the results are in Table 1.

A variance analysis reveals that the differences found in contamination levels among the vegetable species being studied are highly significant in the case of lead ($F = 12.4$) and cadmium ($F = 9.42$). As a result, we emphasize the importance of the species factor in the accumulation of both heavy metals in the place of origin and coincide with the observations previously made by Dedolph et al. (1970) and Jaakkola et al. (1979). Even within the same vegetable species, the differences in the level of contamination in the different anatomical parts have been pointed out (Buchauer 1973; Haghiri 1973). In this sense, and through a statistical study at a specified level using the Student's t-test we found statistically significant differences ($p \leq 0.001$) in the lead and cadmium levels in the different anatomical parts of the beetroot samples ($t \leq 5.52$ for the Pb and $t \leq 6.01$ for the Cd), radish ($t \leq 4.07$ for the Pb and $t \leq 3.97$ for the Cd), potato ($t \leq 5.35$ for the Pb and $t \leq 5.61$ for the Cd), and celery ($t \leq 5.48$ for the Pb and $t \leq 4.41$ for the Cd). We would like to point out that we have not found an explanation for this in the bibliography consulted. In any case, we think that the existence of said differences at an intraspecific level is a consequence of a specified source of contamination of natural or anthropogenic origin. In general terms, we believe this to be of natural origin, since the low levels found (Table 1) and the existence of highly significant correlations ($p \leq 0.001$) between the lead and cadmium content in the vegetables analyzed, eliminate the possibility of the existence of important anthropogenic contaminating sources and suggest irrigation water as the contaminating vehicle.

According to Pozo et al. (1985) the average pollution of the Guadalquivir River water in 1984-85 at it passed through Cordova was $2.30 \pm 1.56$ ug/L in the case of lead and $0.49 \pm 0.13$ ug/L for cadmium.

Of the five groups established according to the Código Alimentario Español (1967), the group presenting the highest levels of lead and cadmium contamination was actually that made

Table 1. Lead and cadmium levels (mg/kg) in vegetable samples at fresh weight

| species | n | LEAD | | CADMIUM | |
|---|---|---|---|---|---|
| | | range | X ± SD | range | X ± SD |
| chard leaves | 11 | 0.115 - 0.226 | 0.193 ± 0.031 | 0.008 - 0.015 | 0.012 ± 0.002 |
| chard roots | 11 | 0.213 - 0.473 | 0.435 ± 0.395 | 0.013 - 0.092 | 0.027 ± 0.024 |
| artichoke leaves | 6 | 0.260 - 0.519 | 0.369 ± 0.092 | 0.017 - 0.033 | 0.024 ± 0.005 |
| artichoke hearts | 6 | 0.340 - 0.514 | 0.403 ± 0.058 | 0.023 - 0.034 | 0.026 ± 0.004 |
| garlic | 6 | 0.079 - 0.190 | 0.141 ± 0.044 | <0.006* - 0.012 | 0.008 ± 0.003 |
| celery leaves | 9 | 0.418 - 0.853 | 0.685 ± 0.134 | 0.024 - 0.052 | 0.040 ± 0.008 |
| celery stalks | 9 | 0.153 - 0.543 | 0.298 ± 0.148 | 0.009 - 0.035 | 0.020 ± 0.010 |
| cardoon | 12 | 0.196 - 0.460 | 0.326 ± 0.086 | 0.012 - 0.029 | 0.021 ± 0.006 |
| onion | 7 | 0.092 - 0.170 | 0.138 ± 0.033 | 0.006 - 0.011 | 0.009 ± 0.002 |
| cauliflower | 10 | 0.074 - 0.296 | 0.162 ± 0.079 | <0.006* - 0.019 | 0.009 ± 0.006 |
| endive | 14 | 0.118 - 0.305 | 0.223 ± 0.049 | 0.007 - 0.020 | 0.013 ± 0.003 |
| spinach | 10 | 0.137 - 0.297 | 0.195 ± 0.043 | 0.008 - 0.021 | 0.011 ± 0.004 |
| green beans | 11 | 0.027 - 0.403 | 0.158 ± 0.100 | <0.006* - 0.026 | 0.010 ± 0.006 |
| lettuce | 9 | 0.117 - 0.242 | 0.178 ± 0.042 | 0.006 - 0.015 | 0.010 ± 0.003 |

* concentration limit 0.006 mg/kg

Table 1 (cont). Lead and cadmium levels (mg/kg) in vegetable samples at fresh weight

| species | n | LEAD | | CADMIUM | |
|---|---|---|---|---|---|
| | | range | X ± SD | range | X ± SD |
| potato pulp | 8 | 0.128 - 0.262 | 0.195 ± 0.046 | 0.008 - 0.017 | 0.013 ± 0.003 |
| potato skin | 8 | 0.293 - 0.451 | 0.340 ± 0.055 | 0.019 - 0.028 | 0.022 ± 0.003 |
| parsley | 10 | 0.109 - 1.009 | 0.412 ± 0.259 | 0.006* - 0.068 | 0.027 ± 0.018 |
| green pepper | 10 | 0.065 - 0.088 | 0.075 ± 0.007 | < 0.006 - 0.006 | 0.006 ± 0.005 |
| leek leaves | 5 | 0.153 - 0.390 | 0.256 ± 0.089 | 0.011 - 0.024 | 0.016 ± 0.005 |
| leek stalks | 5 | 0.120 - 0.259 | 0.171 ± 0.058 | 0.007 - 0.016 | 0.010 ± 0.003 |
| leek roots | 5 | 0.661 - 1.626 | 1.094 ± 0.402 | 0.041 - 0.111 | 0.069 ± 0.027 |
| radish leaves | 9 | 0.220 - 0.595 | 0.404 ± 0.131 | 0.012 - 0.033 | 0.021 ± 0.007 |
| radish roots | 9 | 0.129 - 0.266 | 0.202 ± 0.050 | 0.008 - 0.018 | 0.013 ± 0.003 |
| beet leaves | 6 | 0.429 - 0.684 | 0.545 ± 0.084 | 0.029 - 0.043 | 0.036 ± 0.005 |
| beet roots | 6 | 0.130 - 0.415 | 0.228 ± 0.097 | 0.008 - 0.026 | 0.015 ± 0.006 |
| cabbage | 10 | 0.091 - 0.349 | 0.230 ± 0.067 | 0.006* - 0.022 | 0.014 ± 0.004 |
| tomato | 7 | 0.045 - 0.119 | 0.082 ± 0.028 | < 0.006 - 0.008 | 0.007 ± 0.002 |
| carrot | 6 | 0.107 - 0.359 | 0.191 ± 0.090 | 0.007 - 0.024 | 0.012 ± 0.006 |

* concentration limit 0.006 mg/kg

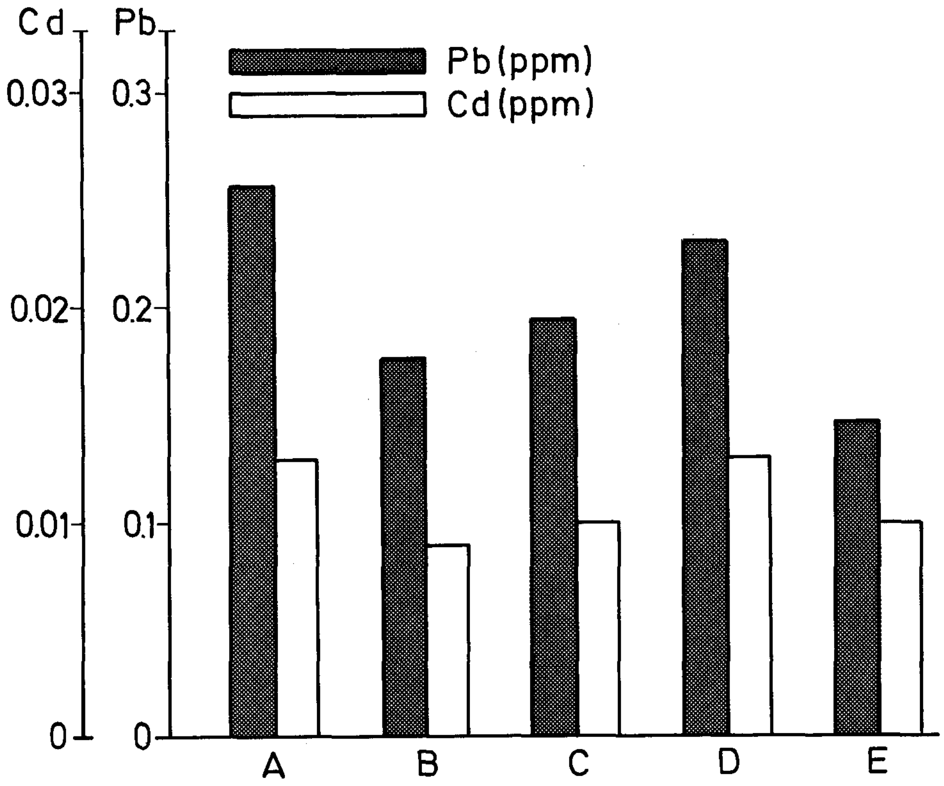


Figure 1. Mean concentrations (mg/kg) of the five established groups of vegetables: A (leaves and soft stalks), B (bulbs), C (cabbages), D (roots and tubercles), E (fruit and similar garden produce).

up of leaves and soft stems (Figure 1). This fact agrees with the observations of Havre and Underdal (1976) in the consideration of the aerial vegetable zones as the most important entry point for the metals under study. However, as can be observed in the same figure, the next lower contamination level corresponds to the group of roots and tubercles. Due to this, it is necessary to take into account the theory of Haghiri (1973) to a certain point in considering the root as an important entry point for these metals into the vegetables. Nevertheless, this only occurs in the leek samples, and for this reason we consider that the contamination in the vegetables being studied seems to be produced to a greater degree by the aerial zone than by the root. This could be explained by irrigation by sprinkling in the zone from which the samples proceed.

Bowen (1966) found that vegetables cultivated in pollution free areas have a cadmium level oscillating between 0.01 and 0.1 mg/kg. These levels are 3 or 4 times higher in those vegetables from intensively cultivated zones of great productivity maintained by the addition of fertilizers and insecticides (Hutchinson et al. 1974). Based on the results obtained in our present research, we consider that the point of origin of the vegetables analyzed shows slight contamination with respect to the heavy metals which are the object of this study.

Noting the average contamination levels obtained for lead and cadmium and keeping in mind that the estimations with repect to the percentage of vegetables forming part of the average nutritional portion of 1500 g are 27 % in Spain (Instituto Nacional de Estadística 1985), we can deduce that the daily intake of lead and cadmium through these vegetables is on the order of 113 ug per day for lead and 7 ug/day for cadmium. The majority of the estimates made in various countries suggests that the daily intake in adults, between food and beverages, oscillates between 100 to 500 ug of lead and 10 to 30 ug of cadmium (Reilly 1980). We feel that reasonable consumption of these vegetables does not pose a health risk for the consumer since their levels are much lower than the levels proposed by FAO/WHO (1978).

REFERENCES

American Chemistry Society (1980) Guidelines for data adquisition and data quality evaluation in environmental chemistry. Anal Chem 52:22-42

Bowen HJM (1966) Trace elements in biochemistry. Academic Press, London and New York

Buchauer MJ (1973) Contamination of soil and vegetation near a zinc smelter by zinc, cadmium, copper and lead. Environ Sci Technol 7:131-135

Código Alimentario Español (1967) Separatas del Boletín Oficial del Estado. Gaceta de Madrid.

Dedolph RG, Ter Haaer R, Holtzman R, Lucar HJ (1970) Sources of lead in perennial reygrass and radishes. Environ Sci Technol 4:217-223

FAO/WHO (1978) Lista de dosis máxima de contaminantes recomendada por la Comisión Mixta FAO/OMS del Codex Alimentarius. Roma

Ganje TJ, Page AL (1972) Lead concentrations of plants, soil and air near highways. Cal Agric 26:7-9

Haghiri F (1973) Cadmium uptake by plants. J Environ Qual 2:93-96

Havre GN, Underdal B (1976) Lead contamination of vegetation grown close to roads. Acta Agric Scan 26:18-24

Hutchinson TC, Czuba M, Cunningham L (1974) Lead, cadmium, zinc copper and nickel distributions in vegetables and soils of and intensely cultivated area and levels of copper, lead and zinc in the growers. Traces Substances in Environmental Health 8:81-93. A Symposium, DD Hemphill Ed University of Missouri. Columbia

Instituto Nacional de Estadística (1985) Encuesta de presupuestos familiares. Estudio sobre nutrición 5 DL M-32466-85. Spain

Jaakkola A, Korkmam J, Koski TJ (1979) The effect of cadmium contained in fertilizers on the cadmium content of vegetables. J Sci Agric Soc Finland 51:158-162

Kehoe RA, Thaman F, Cholak J (1933) Lead absorption and excretion in relation to the diagnosis of lead poisoning. J Ind Hyg 15:320-327

Martinez Para MªC, Masoud TA, Torija Isasa MªE (1979) Determinación de cobre y hierro en hortalizas por absorción atómica. Anal Bromatol 31:189-193

Mottola H (1984) La cinética en la química analítica, en: Métodos cinéticos de análisis. Pérez y Valcarcel Ed. Córdoba (España)

Pozo R, Polo L, Jodral M, Jordano R, Zurera G, Rincón F (1985) Estudio conjunto para el control de la contaminación de las aguas: Estudio de la contaminación en el sistema hidrográfico del Rio Guadalquivir. Proyecto de investigación nº 33021 del CSIC, Spain

Reilly C (1980) Metal contamination of food. Applied Science Publishers. London

Torija Isasa MªE, Martinez Rincón MªC (1982) Plomo y cadmio como contaminantes de hortalizas en fresco. Anal Bromatol 34:71-80

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