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# Cadmium and Lead Contents in Suburban and Urban Soils from Two Medium-Sized Cities of Spain: Influence of Traffic Intensity

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Increasing agricultural, industrial and urban activities are currently leading to increased heavy metal levels in soils with respect to those in natural systems (Nriagu and Pacyna 1988; Kabata-Pendias and Pendias 1992; Alloway 1995). For many years the evaluation of these metals in cultivated soils has attracted the attention of many researchers (Frank et al. 1976; De Boo 1990), but their evaluation in urban and suburban soils, where most of the population lives or works and is continually in contact with such soils, has been only begun a few years ago (Thornton 1990; Mielke et al. 1991; Mielke 1994; Sucharová and Suchara 1995; Markus and Bratney 1996). Urban soils may receive extremely large inputs of toxic metals from different anthropogenic sources but especially from automobile emissions (Ho and Tai 1988; Garcia and Millán 1998). In this sense, from the point of view of human health Cd and Pb are cause for great concern owing to their persistence and potential toxicity (Ragan and Mast 1990).

Most studies on heavy metals levels in urban soils have usually been carried out in large cities with dense traffic or a high degree of industrialisation and few studies have addressed this issue in smaller cities (Thornton 1990). In this work we determined the contents in total and easily soluble Cd and Pb in urban and suburban soils of two cities (Salamanca and Valladolid) chosen from the Region of Castilla-León of Spain, differing in surface area, demography and their degrees of industrial development with a view to assessing the effects of these differences on the enrichment in Cd and Pb of the urban soils. Within Spain, Valladolid can be considered a medium-size city with increasing industrial development while Salamanca is small and there is little development in this sense (Junta de Castilla y León 1998). A further aim of this work is to fill a gap in the data concerning heavy metal contents in Spanish urban soils, which has been many times observed in the international literature.

## MATERIALS AND METHODS

Thirty-three samples of soils were collected in Salamanca and 40 in Valladolid from suburban areas close to roads and from urban areas (median strips of main arterial roads inside the cities and park soils). No samples were taken from parks in Salamanca because the Cd and Pb contents in these soils had already been determined by the authors in a previous work (Sánchez-Camazano et al. 1994a). Also, the total Cd and Pb contents in natural uncontaminated soils from the

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province of Salamanca and of Valladolid have already been determined by the authors (Sánchez-Camazano et al. 1994b, 1998; Sanchez-Martin et al. 1999). The samples were taken in July, 1996 from the soil surface (1- 10 cm) and comprised a mixture of at least three separate subsamples collected at 1 m intervals from one another. The chemical and physicochemical characteristics of the samples were determined (data not shown). The City of Salamanca (SA) has a surface area of 39 km2 and a population of 186156 and the surface of the City of Valladolid (VA) is 198 km2 and its population is 347997 (Junta de Castilla y Leon 1998). The data on mean daily traffic intensity (MDTI) taken at the points closest to the sampling zones range between 1586 and 17728 vehicles/day in SA, and between 1668 and 33540 vehicles/day in VA (Junta de Castilla y Leon 1995).

One gram of finely powdered sample (<0.75 mm) was dissolved in 7.5 mL of HC1 and 2.5 mL of HN03, using a CEM model MDS-2000 microwave oven and volume was brought up with 25 mL of distilled water. Soluble Cd and Pb were extracted by shaking 2 g of soil in 15 mL of 1 N NH4AcO, pH 7, for 1 h. Total and soluble Cd and Pb were determined in triplicate in all cases. Cd and Pb were determined by atomic absorption spectrophotometry using a Varian AA 1475 instrument coupled to a GTA-95 graphite furnace, also from Varian. Standard solutions containing the same acid matrix as the samples were made up at Cd concentrations ranging between 0 and 20 µg/L and at Pb concentrations between 0 and 10 µg/mL. Analyses were performed according to the recommendations provided in the Varian Manual for Analytical Methods ( Rothery 1982). Analytical accuracy was checked with BCR reference materials: BCR 141 (calcareous loam soil) and CRM 30 (river sediment) and was expressed with a variation coefficient of <10%. Special care was taken in preparing the samples and in determining the analytes so as to avoid contamination through deposition of pollutants from the laboratory atmosphere, glassware or potentially contaminated reagents. The precision of the methods was assessed by performing experiments 10 times for a single soil sample (RSD= 6.75% for total Cd determination; RDS= 3.76% for total Pb determination).

## RESULTS AND DISCUSSION

Tables 1 and 2 show the extreme and mean contents of total and soluble Cd and Pb in the urban and suburban soils from the cities studied. They also show the metal contents in soils from parks of SA (Sánchez-Camazano et al. 1994a) and in natural uncontaminated soils from the provinces of SA and VA (Sánchez-Camazano et al. 1994b, 1998; Sanchez-Martin et al. 1999). The contents in natural soils are considered as the reference values for the assessment of the degree of pollution of the urban and suburban soils from the two cities studied.

The total Cd contents in soils from SA range from 0.07 µg/g to 0.95 µg/g (mean 0.41 µg/g). The highest mean content was found in park soils (0.53 µg/g); in soils from suburban zones the minimum and maximum levels were higher in soils taken from farther away from the road, although the mean contents coincided at samples taken at 1 m from the road and those taken at 10 m. In the soils from VA, the total Cd contents range from 0.07 µg/g to 0.59 µg/g (mean 0.28 µg/g) and the highest Cd content was found in soils from median strips (0.38 µg/g). In soils from SA, 86% of the samples had Cd contents above the mean value found in natural soils (0.19 µg/g) and in 61% of the samples the contents were higher than the maximum value found in natural soils (0.33 µg/g). Also, the soils from VA, 78% of the samples had contents higher than the mean Cd level detected in natural soils (0.16 µg/g) and in 23% of the samples the contents were higher than the maximum level found in natural soils (0.43µg/g). The results obtained indicate that the samples studied can be considered to be polluted by Cd to a greater or lesser extent.

The mean contents in NH4AcO-soluble Cd (expressed as percentages of the content in total Cd) were 14.7% (SA soils) and 17.0% (VA soils) (Table 1). These percentages are lower than those obtained in natural soils (18.3% and 19.0%, respectively), indicating that soluble Cd decreases in urban and suburban soils. Only in soils from median strips, the mean contents in soluble Cd were higher than those found in natural soils, indicating that pollution by Cd elicits an enrichment in soluble Cd in these soils. Crisanto Herrero and Lorenzo Martin (1993) also reported an enrichment in soluble Cd in cultivated soils from the province of Salamanca treated for many years with inorganic fertilisers.

**Table 1.** Cd total (µg/g) and soluble fraction (%) in suburban and urban soils from the cities of Salamanca and Valladolid

| Soils/Number | Total Cd | | | Soluble fraction | | |
|---|---|---|---|---|---|---|
| | Range | Mean | Mode | Range | Mean | Mode |
| SALAMANCA | | | | | | |
| Total samples (49) | 0.07-0.95 | 0.41 | 0.52 | 1.95-38.2 | 14.7 | 12.1 |
| Suburban (24) | 0.07-0.64 | 0.31 | 0.26 | 4.60-38.2 | 12.9 | 6.40 |
| Roadside[a] (12) | 0.07-0.63 | 0.31 | 0.28 | | | |
| Close-roadside[b] (12) | 0.13-0.64 | 0.31 | 0.26 | | | |
| Median strip (9) | 0.27-0.87 | 0.47 | 0.43 | 14.2-38.2 | 26.4 | 26.8 |
| Park (16) | 0.21-0.95 | 0.53 | 0.52 | 1.95-25.3 | 10.6 | 13.8 |
| Natural (29) | 0.07-0.33 | 0.19 | 0.13 | 2.90-45.4 | 18.3 | 11.3 |
| VALLADOLID | | | | | | |
| Total samples (40) | 0.07-0.59 | 0.28 | 0.17 | 2.30-90.0 | 17.0 | 18.0 |
| Suburban[a] (11) | 0.17-0.59 | 0.35 | 0.34 | 2.30-25.3 | 11.7 | 10.8 |
| Median strip (8) | 0.24-0.48 | 0.38 | 0.28 | 5.85-41.2 | 20.3 | 12.5 |
| Park (21) | 0.07-0.48 | 0.21 | 0.17 | 5.33-90.0 | 18.6 | 24.2 |
| Natural (30) | 0.05-0.43 | 0.16 | 0.11 | 4.20-44.0 | 19.0 | 18.2 |

[a] Samples taken at 1-3m from the road; [b] Samples taken at 10 m from the road.

The mean contents of total Cd in suburban and urban soils from SA and VA lie in the ranges reported by Garcia and Millán (1998) (0.07-2.22 µg/g) for roadside soils in Guipúzcoa (Spain), province with a lot of industrial activity, by Mielke et al. (1991) (<0.25->2.0 µg/g) for urban soils from 5 cities in Minnesota, and by Carey et al. (1980) (0.1-8.0 µg/g) for suburban and urban soils from five cities in the U.S., although the Cd concentrations found in the soils studied here are close to the minimum levels reported by these authors. There is no uniform procedure throughout the European Union for setting contamination thresholds for heavy metals in urban soils. However, the Cd levels found here are lower than the critical value of 5 µg/g indicating the need for further research according to the Dutch guideline for several soil metal pollutants (Moen et al. 1986), and are also lower than the "trigger concentration" (15 µg/g) of the guideline of the United Kingdom for urban soils (ICRCL 1987).

The total Pb contents fluctuate over a wide range in the soils studied (Table 2). In soils from SA they ranged between 17.7 and 1480 µg/g (mean 165 µg/g and mode 67.9 µg/g) and in soils from VA the range is 18.l- 1117 µg/g (mean 151 µg/g and mode 90.0 µg/g). The highest mean contents corresponded to the soils from the median strips in both cities (580 µg/g and 352 µg/g, respectively), the Pb concentrations in these soils always being higher than the maximum value found in natural soils. The results obtained indicate that 76% of the samples from SA, and 78% of the samples from VA have Pb contents higher than the maximum levels found in natural soils, pointing to soil pollution by this element.

**Table 2.** Pb total (µg/g) and soluble fraction (%) in suburban and urban soils from the cities of Salamanca and Valladolid

| Soils/Number | Total Pb | | | Soluble fraction | | |
|---|---|---|---|---|---|---|
| | Range | Mean | Mode | Range | Mean | Mode |
| SALAMANCA | | | | | | |
| Total samples (49) | 17.7-1480 | 165 | 67.9 | 0.08-5.34 | 1.30 | 0.14 |
| Suburban (24) | 17.7-353 | 85.4 | 62.8 | 0.08-4.74 | 1.17 | 0.27 |
| Roadside[a] (12) | 33.2-353 | 122 | 92.8 | | | |
| Close-roadside[b] (12) | 17.7-89.5 | 48.0 | 45.9 | | | |
| Median strip (9) | 87.2-1480 | 580 | 435 | 0.30-5.34 | 2.09 | 1.64 |
| Park (16) | 20.1-92.6 | 53.1 | 45.3 | 0.08-3.97 | 1.05 | 0.09 |
| Natural (29) | 8.33-44.0 | 24.6 | 24.4 | 0.05-1.82 | 0.51 | 0.32 |
| VALLADOLID | | | | | | |
| Total samples (40) | 18.1-1117 | 151 | 90.0 | 0.03-2.96 | 0.96 | 0.50 |
| Suburban[a] (11) | 19.2-477 | 95.9 | 56.6 | 0.03-2.36 | 0.77 | 0.71 |
| Median strip (8) | 51.0-1117 | 352 | 144 | 0.14-1.99 | 1.16 | 1.30 |
| Park (21) | 18.1-233 | 103 | 93.0 | 0.22-2.96 | 0.99 | 0.59 |
| Natural (30) | 4.44-50.3 | 23.0 | 19.4 | 0.25-4.39 | 1.09 | 0.69 |

[a] Samples taken 1-3 m from the road; [b] Samples taken 10 m from the road.

The mean $NH_4AcO$-soluble Pb contents, expressed as percentages of the total Pb contents, are 1.30% in soils from SA and 0.96% in soils from VA (Table 2) These percentages indicate that the soluble Pb fraction is low, as in natural soils (0.51% and 1.09%, respectively). Nevertheless, in the soils studied, the greater or lesser degree of pollution by Pb gives rise, in general, to an enrichment in soluble Pb that is greater in soils from median strips.
The total Pb concentrations in suburban and urban soils from SA and VA lie

within the range reported by Garcia and Millan (1998) (35-1548 µg/g) for suburban soils in Guipúzcoa (Spain) and by Culbard et al. (1988) (28-1260 µg/g) for soils from parks in Great Britain. Also, the mean Pb contents are within the ranges given by Carey et al. (1980) (2.1-11700 µg/g) for suburban and urban soils from five cities in the U.S., and by Markus and Bratney (1996) (22-20278 µg/g) for urban soils in Glebe, Australia. Although the maximum concentrations found in the soils from SA and VA are very distant from the maximum levels reported by these authors, according to the Dutch guidelines (Moen et al. 1986), in 25% of the samples from both cities the total Pb contents are higher than those requiring investigation (150 µg/g). Additionally, in isolated cases soils from median strips in SA and VA have Pb contents equal to or higher than those specified in this legislation for polluted soils requiring cleaning (600 µg/g). Nevertheless, according to the United Kingdom guideline none of the the samples studied would reach the trigger concentration limit (2000 µg/g) (ICRCL 1987).

In order to determine the influence of the soil properties on the distribution of Cd and Pb, we calculated simple correlations between total and soluble Cd and Pb contents and the soil parameters. No significant correlations were found in this respect when the soils from SA or VA and Cd contents were considered. This lack of correlations with the soil properties could be due to the fact that Cd is deposited onto the polluted soils and is not clearly incorporated into soil dynamics. It should be noted that in natural soils Sánchez-Camazano et al. (1994b, 1998) reported significant correlations between soluble Cd and pH and the cation exchange capacity (CEC) in soils from SA and between total Cd and carbonates in soils from VA. A significant correlation did emerge between the soil organic matter content and soluble Pb ($r=0.57$; $p<0.001$) and total Pb ($r=0.43$; $p<0.05$) in soils from VA. This correlation has also been reported by Cala Rivero et al. (1985) in cultivated soils from of Vega de Aranjuez and by Sánchez-Camazano et al. (1998) in natural soils from the province of Valladolid.

Significant correlations were found between total Cd and soluble Cd and between total Pb and soluble Pb in soils from SA and soils from VA (Table 3). Garcia and Millan (1998) also reported a significant correlation between total Cd and DTPA-extracted Cd in roadside soils from Guipúzcoa (Spain). These results indicate that soluble Cd and Pb contents in soils can be used as indicators of pollution by these metals. Highly significant correlations are also found between soluble Cd contents and total and soluble Pb contents in soils from SA and in soils from VA, indicating that both polluting elements must come from the same source (Akhter and Madany 1993).

A significant correlation is also found between the soluble Cd contents and the MDTI on considering soils from SA, together with a positive but non-significant relationship between the MDTI and the total Cd contents (Table 3). These correlations indicate the effect of traffic on the Cd contents of soils in SA. However, in soils from VA, no significant correlation was found between total Cd and the MDTI. Ho and Tai (1988) did not find a correlation between traffic density and Cd levels in roadside soils, although they did detect one in grass. Also, a significant correlation can be seen between total Pb contents and the MDTI (Table 3) in soils from SA, whereas in soils from VA this correlation is not seen. The pollution due to Cd and Pb in soils from VA may be the consequence of the impact of traffic but may also derive from other sources of pollution related to the greater industrial development in this city. The impact of traffic on the pollution of soils by Pb has been addressed by many authors, who in some cases have found a significant correlation between the total Pb contents in soils and traffic volume (Ho and Tai 1988; Garcia and Millán 1998).

**Table 3.** Correlation matrix for metal contents and MDTI data

| | SA-SOILS | | | | VA-SOILS | | | |
|---|---|---|---|---|---|---|---|---|
| | Total Cd | Soluble Cd | Total Pb | Soluble Pb | Total Cd | Soluble Cd | Total Pb | Soluble Pb |
| SA SOILS | | | | | | | | |
| Total Cd | 1 | | | | | | | |
| Soluble Cd | 0.49[a] | 1 | | | | | | |
| Total Pb | 0.30 | 0.81[c] | 1 | | | | | |
| Soluble Pb | 0.21 | 0.68[c] | 0.95[c] | 1 | | | | |
| VA SOILS | | | | | | | | |
| Total Cd | | | | | 1 | | | |
| Soluble Cd | | | | | 0.37[a] | 1 | | |
| Total Pb | | | | | 0.42[a] | 0.82[c] | 1 | |
| Soluble Pb | | | | | 0.44[a] | 0.80[c] | 0.96[c] | 1 |
| MDTI | 0.32 | 0.48[a] | 0.39[a] | 0.28 | -0.24 | 0.16 | 0.10 | 0.10 |

[a]Significant at <0.05 level; [c]Significant at 0.001 level

To compare the degree of pollution by Cd and Pb in the soils from Salamanca and Valladolid, the mean Cd and Pb contents in the polluted soils were expressed as multiples of the mean levels of these metals in natural soils (Figure 1). According to the results, pollution due Cd is higher in VA than in SA in suburban soils, possibly owing to the influence not only of the traffic intensity but also of other sources of pollution of industrial origin located close to the zones studied (paint, fertiliser and automobile factories) (Page and Binham 1973). In soils from parks, pollution is higher in SA; this could be explained taking into account the smaller extension of the parks in this city and their greater age and use (Sánchez-Camazano et al. 1994a). Pollution is similar in soils from median strips as a result of traffic intensity in both cities. The highest Pb contents corresponded in the two cities to median strip soils, where the impact of pollution due to traffic intensity is highest, especially in SA where the streets are narrower and the median strips are older. In suburban soils, and especially in park soils, the contamination indices are higher in VA than in SA, pointing to the effect of the greater degree of

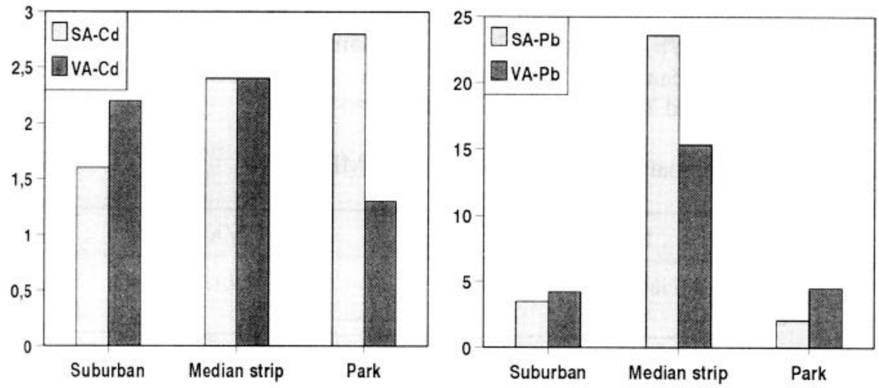


**Figure 1.** Mean Cd and Pb contents in soils from SA and VA as multiples of the mean contents of these metals in naturals soils.

development of this city (VA), with a larger population, more traffic, and a greater number of industries and commercial centres.

The results obtained in the present work point to an enrichment in the levels of Cd and Pb in the urban and suburban soils of the two cities studied. This increase derives from the impact of traffic and human and industrial activities, as also reported by other authors (Mielke et al. 1991). Nevertheless, the levels reached in the soils up till now, are considerably lower than the threshold concentrations admitted in European guidelines.

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