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Concentrations of potentially toxic metals in urban soils of Seville: relationship with different land uses

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Abstract

Fifty-two samples of surface soils were taken in the urban area of Seville, to assess the possible influence of different land uses on their metal contents and their relationship with several soil properties. The samples corresponded to five categories or land uses: agricultural, parks, ornamental gardens, riverbanks, and roadsides. Sequential extraction of metal according to the procedure proposed by the former Community Bureau of Reference (BCR) was carried out, and pseudo-total (*aqua regia* soluble) metal contents were determined. Lower organic C, total N and available P and K contents were found in riverbank samples, probably due to the lack of manuring of those sites, left in a natural status. In contrast, significantly higher electrical conductivity was found in those sites, due to the tidal influence of the nearby Atlantic Ocean. Other land uses did not show significant differences in the general properties. Concentrations of Cu, Pb and Zn, both *aqua-regia* soluble and sequentially extracted, were clearly higher in soils from ornamental gardens, whereas the concentrations in the riverbank samples were slightly lower than the other categories. In contrast, other metals (Cd, Cr, Fe, Mn, Ni) were uniformly distributed throughout all land uses. A strong statistical association is found among the concentrations of Cu, Pb, Zn and organic C, suggesting that the larger contents of these metals in ornamental gardens are partly due to organic amendments added to those sites more frequently than to other kinds of sites. Considering the conclusions of previous studies, heavy traffic can also contribute to those 'urban' metals in urban soils. Periodic monitoring of the concentrations of urban metals in busy city centres and of the quality of amendments added to soils of recreational areas are recommended.

Introduction

The environmental quality of soils as related to their contents in potentially toxic metals (PTM) is being studied since long ago. Although the concern for high contents of PTM in soil is justified because of the possibility of their transfer to animals and man through plants, lately the existence of high metal contents in non-agricultural soils is receiving growing attention. This is especially true in the case of soils in urban areas that are used for residential or recreational activities. In such areas, soils come easily in direct contact with humans, especially children, so that soil metals can be directly transferred to them. That is why some national regulations on maximum acceptable concentrations of PTM in soils consider specific values for parks and residential areas. Some examples are given in Table 1 (Swedish Environmental Protection Agency, 1997; Ministero dell'Ambiente, 1999; Canadian Council of Ministers of the Environment, 2001; Ministère del' Environnement du Québec, 2001; Sociedad Pública de Gestión Ambiental, 2002). As can be seen, wide variations are found among different countries. Many other countries or regions have defined guideline values for metal concentrations in soils, but no distinction is made for any particular use.

Table 1. Maximum acceptable limits (mg kg^{-1}) established in various countries or regions for the PTM studied here in soils of residential/recreational areas.

Country/Region	Cr	Ni	Pb	Zn	Cu
Québec	250	100	500	500	100
Canada	64	50	140	200	63
Italy	150	120	100	150	120
Sweden [a]	120	35	80	350	100
Basque country	400	500	450	–	–

[a] "KM", land with sensitive use (residential areas, kindergarten, agricultural, etc.).

In previous studies of urban soils in Seville (SW Spain) (Madrid *et al*., 2002, 2004), evidence was given of a certain degree of pollution in some specific spots of the city. However this degree of pollution was not especially high when compared to some other European cities (Hursthouse *et al*., 2004; Madrid *et al*., submitted for publication). However, only soils in the main parks of the city were included in those studies. In the work described here, a new sampling campaign has been designed with the aim of obtaining a more uniform distribution of samples within the city, including a selection of the various land uses or categories that can be distinguished for soils within the city.

Materials and methods

Surface soil (0–10 cm) samples were taken at 52 sites of the city. The sampling protocol has been described elsewhere (Madrid *et al*., 2004). Figure 1 shows the location of the sampling points.

Five site categories or land uses were distinguished for sampling:

Fig. 1. Location and use of the sampling sites.

(a) Areas of agricultural use (*AG*) (6 points),
(b) Parks: major recreational extensions with a predominance of green areas (*PA*) (22 points),
(c) Ornamental gardens: smaller sites with ornamental plants, usually within highly urbanised areas, especially squares (*OR*) (7 points),
(d) Riverbanks: uncultivated sites, close to the Guadalquivir river, not receiving any special care (*RB*) (5 points), and
(e) Roadsides: small green areas, or parts of parks, located less than 10 m away from the edge of roads with high traffic density (*RS*) (12 points).

General soil properties (particle-size distribution, pH, electrical conductivity, carbonate and organic matter contents, total N, available P and K) were estimated by standard procedures (Madrid *et al.*, 2004). Sequential extraction of soil metals was carried out by a modified version of the original 3-step BCR (nowadays Standards, Measurement and Testing Programme) procedure (Mossop and Davidson, 2003). Metals not solubilised in this sequential extraction procedure are not considered to be of environmental significance (Hooda and Halloway, 1994). Metals extracted by *aqua regia* in the residual fraction were determined by a microwave oven technique (Kingston and Haswell, 1997). The *aqua-regia* extractable metal contents have been sometimes called the 'pseudo-total contents', and those metals not soluble in *aqua regia* are considered to be mostly bound to silicate minerals and unimportant for estimating the mobility and behaviour of elements (Niskavaara *et al.*, 1997; Chen and Ma, 2001).

Results

General soil properties

Table 2 summarises the results for the general descriptive properties of the samples. Wide ranges in most variables are observed regardless of whether all samples are included or if the different categories are considered separately. A picture of the distribution of most variables can be seen in the box plots of Figures 2a–d.

Figure 2a shows that the contents in carbonate and in clay fraction seem to vary at random without any noticeable trend for a given land use. The same conclusion is reached for CEC in Figure 2b. In contrast, organic C for the riverbank (*RB*) samples (Figure 2b) is significantly lower than for the other categories. In Figure 2c the N contents are also lower for this same land use, whereas electrical conductivity is clearly higher. As the sites for *RB* samples do not receive any amendment and thus are in a more or less 'natural' situation, they are likely to have less organic matter and less N than all other sites, which in most cases receive frequent care. It is worth to remember that natural soils in SW Spain are often poor in organic matter compared to other, cooler European areas. Also, the tidal effects from the not too distant Atlantic ocean, which are noticeable well upstream along the estuary of Guadalquivir river, make it likely that some increase in salinity can be detectable in soils close to the river. Therefore these differences for the *RB* samples are not surprising. The lack of amendments must also cause that the contents of nutrients as P or K are significantly lower for this category (Figure 2d).

Metal concentrations

Considering that some differences in general properties have been just shown to exist among the various land uses chosen, it is convenient to check whether different land uses also cause differences in contents of pollutants, either in total concentrations or the most available fractions. With this purpose, the *aqua regia*-soluble metals were considered a good estimate of the total metal contents, and the BCR sequential extraction procedure was chosen as a way of estimating the environmentally more available fractions of some metals. Table 3 gives a summary of the data for the sum of the first 3 fractions of the metals considered and the total (actually 'pseudo-total', see the Materials and Methods section) contents (3 fractions plus the residual fraction). The means of the sum of 3 fractions are also expressed as per cent of the total contents.

The box plots in Figures 3a–c show the distribution of the sum of the 3 first fractions of the sequential extraction. It is readily noticeable that Cu, Pb and Zn extracted from the *OR* samples (Figure 3a) are clearly above the data for the other land uses. Data for these metals in *RB* samples seem to be lower than for the other categories, but overlapping of the interquartile ranges is

Table 2. Summary of the general properties of the samples studied.

	Category	Mean	SD	Range
pH ($CaCl_2$)	All	7.21	0.13	6.65–7.42
	AG	7.32	0.05	7.22–7.38
	OR	7.19	0.13	6.98–7.31
	PA	7.16	0.14	6.65–7.37
	RB	7.34	0.09	7.20–7.42
	RS	7.20	0.05	7.13–7.30
EC/dS m^{-1}[a]	All	0.16	0.09	0.06–0.67
	AG	0.19	0.06	0.12–0.28
	OR	0.09	0.02	0.06–0.11
	PA	0.16	0.06	0.08–0.27
	RB	0.31	0.21	0.14–0.67
	RS	0.14	0.05	0.08–0.24
CCE/g kg^{-1}[b]	All	205	74	4–362
	AG	238	20	210–258
	OR	218	60	162–337
	PA	197	94	4–362
	RB	242	31	195–275
	RS	176	63	19–241
Org. C/g kg^{-1}	All	19.7	9.9	4.9–46.7
	AG	19.0	11.3	5.7–31.7
	OR	25.9	5.2	17.3–31.6
	PA	21.2	10.6	7.8–46.7
	RB	10.4	6.0	4.9–20.1
	RS	17.7	8.9	7.3–36.7
N/g kg^{-1}	All	1.8	1.0	0.4–5.2
	AG	2.0	1.2	0.5–3.3
	OR	2.2	0.5	1.7–3.0
	PA	2.1	1.2	0.7–5.2
	RB	0.7	0.3	0.4–1.3
	RS	1.6	0.7	0.7–3.3
Avail. P/mg kg^{-1}	All	53	69	3–299
	AG	119	126	12–299
	OR	117	58	54–205
	PA	35	44	5–199
	RB	8	5	3–15
	RS	30	41	4–152
Avail. K/mg kg^{-1}	All	360	249	48–1553
	AG	383	138	227–566
	OR	584	463	224–1553
	PA	366	182	173–839
	RB	127	48	48–169
	RS	303	173	59–577
Clay/g kg^{-1}	All	270	102	72–469
	AG	291	37	234–339
	OR	211	85	105–379
	PA	273	107	72–458
	RB	257	149	77–469
	RS	294	101	140–426
CEC/cmol kg^{-1}[c]	All	15.4	7.2	2.1–35.4
	AG	13.8	3.4	9.7–17.8
	OR	14.7	2.7	9.9–17.5
	PA	17.5	8.1	5.9–35.4
	RB	11.0	6.9	2.1–19.8
	RS	14.9	8.4	5.2–28.7

[a] Electrical conductivity.
[b] Calcium carbonate equivalent.
[c] Cation exchange capacity.

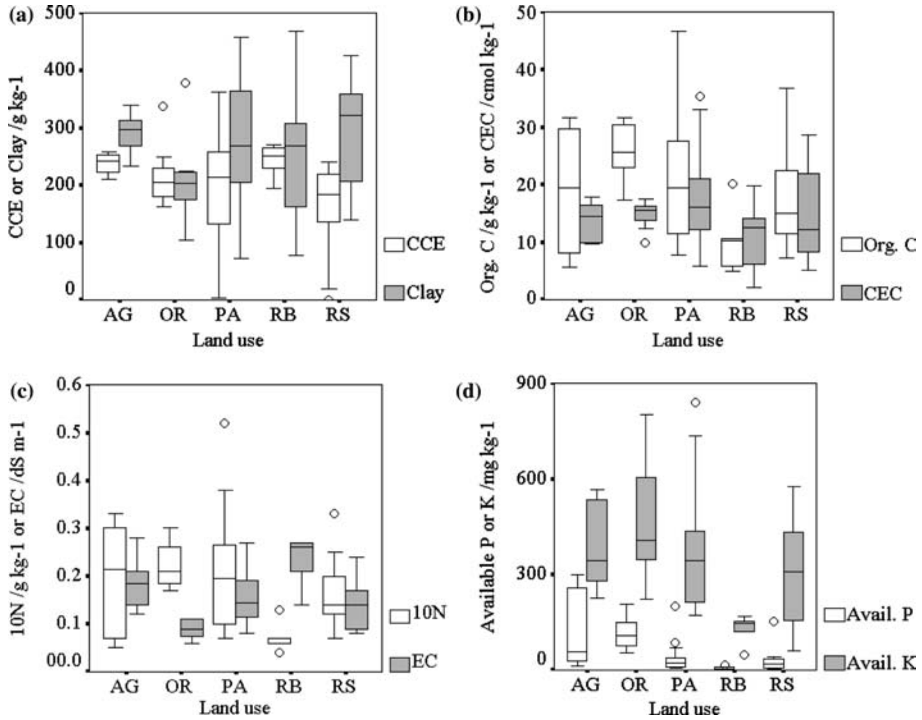

Fig. 2. Box plots for some general descriptive properties of the soil samples. Boxes, interquartile range (IQR). Dots, outliers (values below $Q_1 - 1.5 \times IQR$ and above $Q_3 + 1.5 \times IQR$). Whiskers, range excluding the outliers. Medians are marked in each box. (a) CCE and clay contents; (b) Org. C and CEC; (c) N and EC; (d) Available P and K. In the land use axis, 'AG' stands for agricultural, 'OR' for ornamental garden, 'PA' for park, 'RB' for riverbank, and 'RS' for roadside samples.

noticeable. In all other cases, no significant differences can be seen among land uses, with the exception of Cr in the *RB* samples (Figure 3b), that seems significantly below the other categories. Fe and Mn (Figure 3c), which can be considered unlikely to be strongly influenced by human activities, are clearly invariant among categories. Similar conclusions are reached if the total *aqua-regia* concentrations or any of the three fractions of the BCR procedure are separately considered (not shown). This fact means that the different land uses do not cause significant changes among the three BCR fractions. The similarity in behaviour of the total concentrations and the sum of the BCR fractions is a logical consequence of the constancy observed in the latter for a given metal when expressed as per cent of the total, as can be seen in Table 3.

Principal component analysis

The possible associations among the general soil properties can be objectively checked by principal component analysis (PCA). Table 4 shows the results of Varimax-rotated PCA, represented by the correlation coefficients between the various variables and the first 3 components, which account for about 72% of the total variance. The first component accounts for a 36.2% of the total variance and shows the association of those variables most related to soil fertility (N, P, K and organic C). The second factor, with 19.2% of the variance, is related with soil mineralogy (CEC and clay content). The third factor seems to include the influence of salts, represented by EC and CCE. Figure 4 shows the distribution of the sampling points when represented on the plane defined by the first two factors. It can be seen that all *OR* samples are located on the positive side of the axis for component 1, whereas *RB* samples are all located on the opposite side of that axis. The other categories are almost uniformly distributed with respect to component 1, and none of them show any tendency with respect to either axis. The result for land uses *OR* and *RB* is again a consequence of the more intense

Table 3. Means and ranges of the sum of the first three sequentially extracted fractions and of the total contents (3 fractions plus residual).

	Land use	Mean, 3 fracs.		Range, 3 fracs.	Mean, total	Range, total
		/mg kg^{-1}	/% of total	/mg kg^{-1}	/mg kg^{-1}	/mg kg^{-1}
Cd	All	2.17	74	0.17–3.85	2.89	0.18–4.85
	AG	2.20	74	1.88–2.42	3.04	2.38–3.53
	OR	2.27	77	1.67–2.80	2.98	1.97–3.80
	PA	2.31	76	0.17–3.85	3.01	0.18–4.85
	RB	2.62	78	1.67–3.50	3.27	2.45–4.00
	RS	1.65	67	0.17–2.72	2.37	0.58–4.22
Cr	All	12.7	33	4.4–26.5	38.4	14.0–86.6
	AG	12.1	35	10.3–13.8	35.0	30.7–41.2
	OR	10.9	31	8.8–13.5	36.7	28.6–53.5
	PA	14.6	36	5.4–26.5	40.5	26.5–86.6
	RB	5.5	21	4.4–6.2	27.4	14.0–37.5
	RS	13.7	33	7.1–23.5	42.1	28.9–61.2
Cu	All	22.9	34	2.2–126	54.5	10.7–198
	AG	13.3	30	4.9–22.3	42.4	23.4–67.4
	OR	51.9	47	22.7–77.6	107	51.9–155
	PA	25.2	36	2.2–126	56.8	14.1–198
	RB	5.3	23	3.2–7.5	24.2	10.7–37.2
	RS	13.8	30	3.3–67.4	38.4	17.4–116
Fe	All	520	3	60–2000	16400	8700–29200
	AG	326	2	184–629	15400	14700–16900
	OR	487	3	60–933	15600	12900–18200
	PA	477	3	63–1140	15600	11100–22200
	RB	452	3	206–791	16000	8700–20200
	RS	742	4	246–2000	19000	14000–29300
Mn	All	294	75	128–701	391	187–781
	AG	300	76	247–377	394	320–480
	OR	278	74	198–408	372	300–515
	PA	287	76	140–701	368	222–765
	RB	282	75	128–427	373	187–548
	RS	318	71	139–434	451	236–781
Ni	All	4.57	21	1.60–12.7	21.2	11.1–46.7
	AG	2.54	13	1.60–3.57	19.5	14.4–22.9
	OR	3.80	23	2.23–7.75	16.4	12.6–19.4
	PA	5.00	23	1.92–12.7	21.6	12.1–46.7
	RB	3.16	16	1.92–5.58	20.8	11.1–27.1
	RS	5.85	24	1.90–11.1	24.2	15.6–32.4
Pb	All	79.8	63	12.6–263	156	23.0–725
	AG	76.5	64	33.9–210	119	52.6–331
	OR	169	48	65.6–263	411	162–725
	PA	83.1	67	15.6–259	146	23.4–702
	RB	22.2	59	15.1–37.6	37.4	23.0–55.1
	RS	47.3	64	12.6–135	92.1	24.3–476
Zn	All	54.4	40	5.5–186	120	38.8–326
	AG	27.2	29	15.3–40.9	91.6	63.8–137
	OR	124	53	56.5–186	229	139–326
	PA	56.1	42	11.2–154	121	38.8–288
	RB	19.8	28	5.5–51.4	65.5	39.8–92.0
	RS	38.8	39	14.1–145	91.4	46.4–231

care that *OR* sites probably receive, including manure and organic amendments, whereas *RB* sites are in its natural status.

Again, possible associations among metals can be objectively ascertained by PCA. In Table 5 results are shown for the first three principal

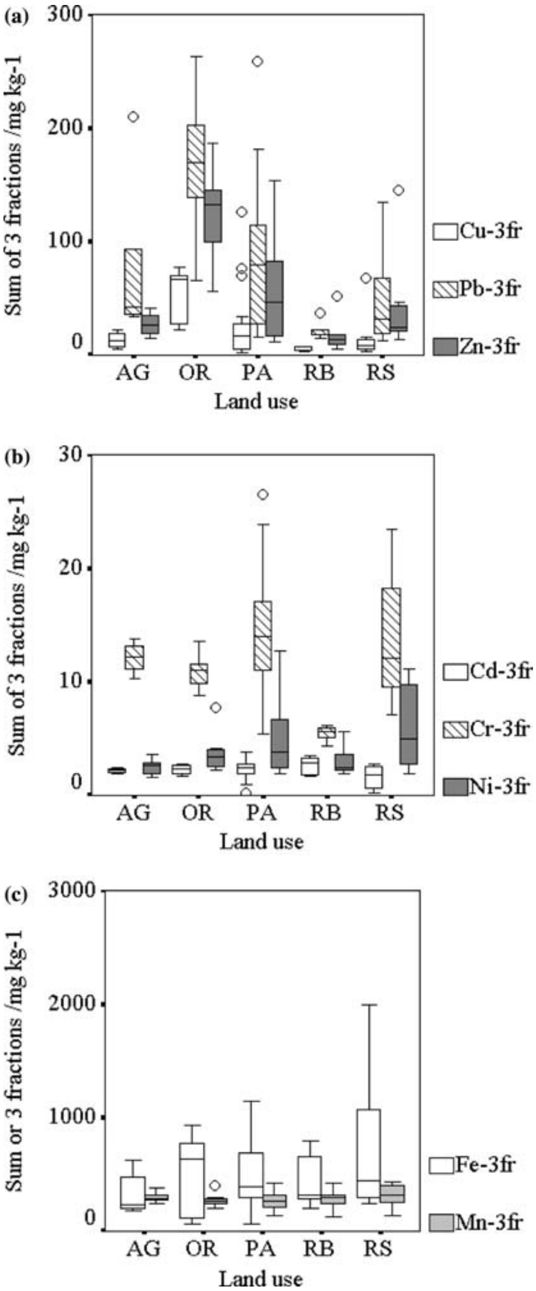

Fig. 3. Box plots for the sum of the first three sequentially extracted fractions of several metals. Symbols as in Figure 2. (a) Cu, Pb, Zn; (b) Cd, Cr, Ni; (c) Fe, Mn.

components after Varimax rotation. As a clear relationship has been previously reported between some soil metals and organic matter (Madrid *et al.*, 2004), organic C was also included in the analysis. About 75% of the variance is accounted for by the three first factors of the analysis. The first component groups together the data for Cu, Pb and Zn, regardless of whether the residual fraction is included or not. These metals have been often reported as of 'urban' origin in urban soils, so that component 1 can be considered as 'anthropogenic'. The second component includes the data for Mn and Cd and the most available fractions of Ni, with a weak contribution of the available fraction of Fe. Cr data and the total contents in Fe and Ni are included in component 3. When represented on the plane of the first two factors (Figure 5), the points for OG are located near the axis for component 1, whereas most other sampling points are distributed close to the axis for component 2.

Discussion

Although no strong differences are found among most properties of soils with different uses in the urban area of Seville, some minor effects are noticeable. These effects are often attributable to the treatments that many soils within the city receive, which are lacking in some areas left in a more or less 'natural' status. Examples of the latter are some spots along the riverbanks. In those cases some of the observed differences can be attributed to the tidal influence of the sea.

In some soils supporting ornamental plants, where gardening practices include frequent addition of organic amendments, a significant increase in some metals is observed as compared to other land uses. This effect is observed for the (pseudo) total as well as for the more easily accessible fractions of Cu, Pb and Zn. The observed contents are in some sites high enough as to be considered as moderately polluted, especially in the case of Pb. This is noticeable when data for ranges for total contents of these three metals are compared with the guideline values for several countries given in Table 1. Other metals studied are scarcely, or not at all, affected by the various uses. This different behaviour brings about a strong association of the contents of Cu, Pb and Zn, as shown by PCA.

Two different reasons can be responsible for the observed increase in those three metals in OR soils. The most obvious one is that the organic amendments used might have moderately high contents in such metals. No data are available for

Table 4. Correlation coefficients between several descriptive soil properties and the first 3 factors resulting from principal component analysis after Varimax rotation, eigenvalues and percent variance accounted for. Communalities show the proportion of each variable variance accounted for by the factors.

Variable	Factor 1	Factor 2	Factor 3	Communalities
Organic C	**0.890**	0.074	0.042	0.799
CEC	0.262	**0.827**	−0.259	0.819
EC	−0.179	−0.038	**0.794**	0.664
CCE	0.369	−0.037	**0.775**	0.738
Clay	−0.071	**0.881**	0.114	0.794
N	**0.889**	0.142	0.062	0.814
Available P	**0.765**	−0.143	0.034	0.606
Available K	**0.698**	0.157	−0.036	0.513
Eigenvalue	2.895	1.533	1.319	
% variance	36.2	19.2	16.5	
Cumulative		55.4	71.9	

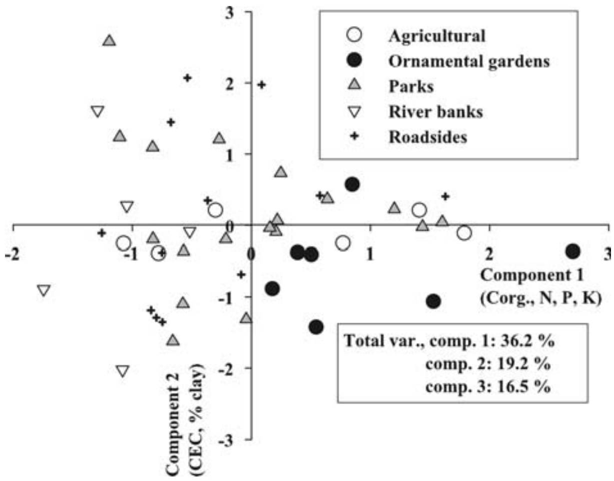

Fig. 4. Scatter of the sampling points on the plane of the first two principal components of the PCA for the general descriptive properties after Varimax rotation.

the quality of amendments used by the Parks and Gardens Service of Seville Municipality, but the association found between organic matter contents and those three metals points to that direction. However, other pollutants that are likely to be present in low quality organic amendments (*e.g*. Cr, Ni) are clearly not related with organic matter in our soils and do not present high contents.

Another possibility was already suggested in a previous paper (Madrid *et al*., 2002). The *OR* sites are all close to the busy historic centre of the city, where traffic density has been very heavy for many years. Thus the high contents in some metals, especially Pb, could be related with engine exhausts, tyre wear, etc. Madrid *et al*. (2002) showed that multiple correlation of those metal contents with traffic density and distance to the nearby roads was low, but not negligible. However, some of the soils with highest contents in Pb are located in small, out-of-the-way squares with limited access of motor vehicles.

Although neither possibility can be fully confirmed, both can contribute to the moderate pollution found in *OR* soils. The second one is becoming less significant in what Pb is concerned after shift to unleaded petrol, but it is still likely that other metals related with traffic, *e.g*. Cu, Zn, etc. (De Miguel *et al*., 1997), are still being added to the environment in busy city centres in Europe.

Table 5. Correlation coefficients between the sum of the sequentially extracted fractions for several metals (excluding and including the residual) and organic C and the first 3 factors resulting from principal component analysis after Varimax rotation, eigenvalues and percent variance accounted for. Communalities show the proportion of each variable variance accounted for by the factors.

Variable	Factor 1	Factor 2	Factor 3	Communalities
Organic C	0.616	−0.269	0.059	0.456
Cd all fractions	0.095	**−0.923**	0.174	0.891
Cr all fractions	0.137	0.117	**0.884**	0.815
Cu all fractions	**0.950**	−0.087	−0.039	0.912
Fe all fractions	−0.120	0.265	**0.807**	0.736
Mn all fractions	−0.181	**0.682**	0.497	0.745
Ni all fractions	−0.365	0.281	**0.710**	0.715
Pb all fractions	**0.930**	−0.079	−0.061	0.876
Zn all fractions	**0.922**	−0.183	0.001	0.884
Cd 3 fractions	0.091	**−0.902**	0.051	0.824
Cr 3 fractions	0.149	−0.253	**0.772**	0.683
Cu 3 fractions	**0.933**	−0.069	−0.117	0.890
Fe 3 fractions	−0.098	0.567	0.128	0.347
Mn 3 fractions	−0.192	**0.735**	0.376	0.718
Ni 3 fractions	−0.173	**0.728**	0.104	0.570
Pb 3 fractions	**0.838**	−0.155	0.027	0.727
Zn 3 fractions	**0.930**	−0.110	−0.052	0.880
Eigenvalues	5.753	3.908	3.008	
% variance	33.8	23.0	17.7	
Cumulative		56.8	74.5	

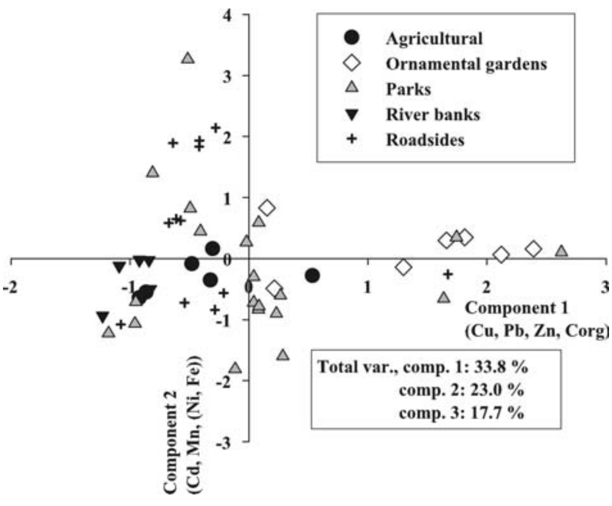

Fig. 5. Scatter of the sampling points on the plane of the first two principal components of the PCA for the sequentially extracted metals after Varimax rotation.

A periodic monitoring of metal contents in such areas is thus advisable. Also, especial care must be taken concerning the quality of amendments used in soils of urban, recreational areas, in order to maintain a good environmental quality of such areas.

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