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PHYTOREMEDIATION OF POLLUTED SOILS: RECENT PROGRESS AND DEVELOPMENTS

Origin and spatial distribution of metals in urban soils

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Abstract

Purpose This study assessed soils from 36 parks and gardens (Vigo City, NW of Spain) where there are different degrees of traffic intensity and activity.

Materials and methods The soils were characterised, and the content of Ba, Ca, Cr, Cu, Fe, Mg, Mn, Na, Ni, Pb, Si, Sr and Zn was analysed. Further assessment determined the geoaccumulation index, enrichment factor and the contamination degree by metals with adverse effects on human health and environmental quality.

Results and discussion The results reveal the existence of a moderate degree of contamination by Ba, Pb and Cu, which contribute the most to soil contamination due to the influence of industrial areas and main transport routes. Correlation and cluster analyses suggest that the metals included in the study have three possible origins: "natural" (Na and Si), "mixed" (two groups with different source intensity: Ca and Sr and Cr, Fe, Mg, Mn and Ni) and two possible "urban" sources: traffic (Cu, Pb, Zn) and mixed (Ba).

Conclusions None of the soils can be classified as strongly contaminated but more than 61 % of the moderate contamination degree determined in the studied soils is explained by the Ba, Cu and Pb contents.

Keywords Enrichment factor · Geoaccumulation · Pollution · Metals · Sources · Urban soil

1 Introduction

Urban soils are increasingly attracting the attention of numerous investigators, who are focusing on soils, vegetation and demographic and economic factors, mainly due to the fact that almost 49 % of the world population lives in urban areas, and it is estimated that this figure will be increased to 84 % in 2050. In Europe, 75 % of the population lives in urban or peri-urban areas covering only 9 % of the total land surface, and in 2020 this value will rise to 80 % (Ajmone-Marsan and Biasioli 2010; Picket et al. 2011). These soils are being subjected to severe anthropogenic pressure because of urban development. There are different "urban factors" which directly or indirectly affect their physical, chemical and biological properties (Picket et al. 2011). The urban soils are created by the processes of urbanisation, and they are materials that have been manipulated, disturbed or transported by man's activities in the urban environment. Their physical, chemical and biological properties are generally unfavourable as rooting medium (Craul 1992). Therefore, urban soils are different from agricultural or natural ones as these are soils that were transformed by supply of amendments and fertilisation, ploughing, sowing, etc., which makes them fertiles, suitables for life development, particularly plants for animal and human consumption, and for livestock production. Hence, the conditions of the urban soils vary from one city to another depending on the urban structure, the distribution of vehicles and the types of fuels used (Kabata-Pendias 2010; Minguillón et al. 2014).

Soils play an important role as they act not only as sources but also as sinks of various contaminants. Unlike in the atmosphere or water masses, contaminants are accumulated in soils

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for long periods due to their interaction with colloidal particles (Metreveli and Frimmel 2007). Furthermore, green areas are essential components of urban ecosystems, not only because of their capacity to purify toxic substances but also because of their role in the microclimate (e.g. dampen temperature and water content variations), among other factors (De Kimpe and Morel 2000; Li et al. 2013).

The natural processes and anthropogenic activities, such as the atmospheric emissions of industries and transport, the construction waste disposal and landfills (Puskás and Farsang 2009) influence the metal content in urban soils. The proximity of humans and animals to contamination sources in urban areas (industries, transport infrastructures, etc) increases the risk of developing various diseases through the inhalation, ingestion or dermal exposure, enhancing mortality and morbidity rates (Rydin et al. 2012; Minguillón et al. 2014). Therefore, they are commonly used as indicators of environmental quality and pollution (Loska et al. 2004; Ajmone-Marsan and Biasioli 2010; Szolnoki et al. 2013).

After the ban on leaded petrol in most industrialised countries around the end of the 1990s, Pb is no longer the only indicator of pollution and heavy traffic in urban environments. As a result, in recent years, Ba, Cd, Cu, Ni, Pt, Pd or Zn have been introduced as other indicators of traffic-related contamination (Monaci and Bargagli 1997; Sutherland 2000; Zechmeister et al. 2005). For example, the origin of Ba is attributed to enrichment due to brake linings in cars and later emissions due to frictional processes (Fernández–Espinosa and Ternero-Rodríguez 2004).

To control risks and determine the degree of soil contamination with respect to natural or background values, different legislations include generic reference or intervention levels (GRL in air, water or soil). They allow to estimate the soil quality and to study any risk above accepted levels for human health or protection of ecosystems.

The objectives of this study were (1) to study the concentrations of several elements (Ba, Ca, Cr, Cu, Fe, Mg, Mn, Na, Ni, Pb, Si, Sr and Zn), focussing on those with implications for environmental implications in the urban soils from Vigo (NW, Spain); (2) to determine the degree of soil quality and (3) to identify the possible origin (natural and/or anthropogenic) of the metals in the soils for obtaining the spatial distribution of the possible pollutants.

2 Materials and methods

2.1 Study area

The city of Vigo (296,479 inhabitants in 2013) is located in the NW of Spain (datum ETRS89/UTM 29 N 42.2333 N, -8.7166 W) on the shores of the Ría de Vigo. It is the largest city in the region, and the 14th largest in Spain. Important commercial and industrial activities are operating in this area, in the SW of the city there is a car manufacturing plant whilst in the NW and NE the most noteworthy companies are involved in shipbuilding (dockyards). There is also an important commercial and fishing port as well as an airport situated 9 km E of the city (Fig. 1). The number of vehicles is around 170,000 (±50 % diesel), and the average daily traffic at the end of 2013 was approximately 500,000. The climate ranges from oceanic to Mediterranean, with mild temperatures (an average of 14.8 °C) and high precipitation (1450 mm year^{-1}). The dominant winds are from the SW. The geology of the area is mainly granite rock in the W and gneiss in the E, the latter occupying more than 50 % of the surface.

2.2 Sampling and analytical method

A total of 36 soil samples were collected (0–20 cm depth) from green areas scattered around the city of Vigo. The main focus when sampling was paid to the city centre during a dry period (1–15 October 2013) (Fig. 1). In order to obtain a good perspective of the evolution of the city, samples were taken from at least 10-year-old areas that had not undergone important changes. They were also selected attending to the degree and type of vegetation coverage as this is an important factor that influences the soil properties and functions. The soil structure, organic matter content, water holding capacity, pH, etc. are directly related to the plant coverage and also influence the retention of the different pollutants. The selected soils were also sampled attending to the intensity of surrounding traffic and the distance to different areas of activities (residential, industrial, riverside walks, etc.) (Table 1). Most of these green areas are subjected to some kind of human intervention (irrigation, fertilisation and periodical changes in ornamental vegetation, trampling of the herbaceous vegetation by pedestrians, etc.).

After the elimination of the superficial layer of vegetation (2–3 cm depth), the samples were taken using an Eijkelkamp Mod.04.20.SA sampler (three subsamples per sampling point) and stored in polyethylene bags to be transported to the laboratory. They were air dried, sieved (<2 mm) and homogenised to make up a compound sample, which was then subdivided into three subsamples for further analysis.

After sieving, the coarse component of the soil was determined (Eriksson and Holmgren 1996). The fraction <2 mm was analysed for soil pH, total Kjeldahl-N and organic matter content (OM). They were determined, respectively, with a pH electrode in 2:1 water/soil extracts, according to Bremner and Mulvaney (1982) and following the Walkley and Black (1934) procedure. Available P was determined as described by Olsen and Sommers (1982). Exchangeable cations (Ca^{2+}, K^+, Mg^{2+}, Na^+ and Al^{3+}) were extracted with 0.1 M $BaCl_2$ (Hendershot and Duquette 1986). The concentrations of Al, Ca, K, Mg and Na and available P were analysed by ICP-OES (Perkin Elmer Optima 4300DV).

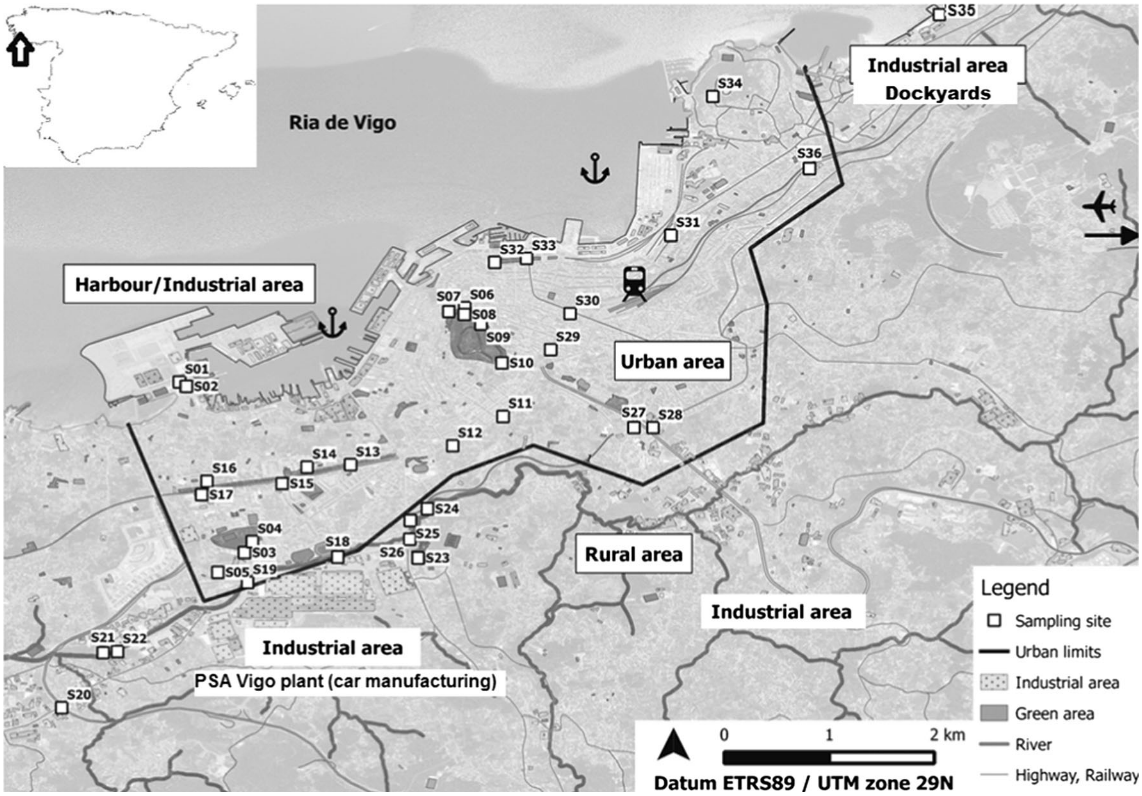

Fig. 1 Sampling sites of the soils from 36 parks and gardens of Vigo City

The total concentrations of Ba, Ca, Cr, Cu, Fe, Mg, Mn, Na, Ni, Pb, Si, Sr and Zn in the soils were determined by means of X-ray fluorescence (XRF) (Siemens; model SRS-3000, Hannover, Germany). For each sample, three 40-mm-diameter pellets were prepared by compacting 5 g of the ground sample under 25 Mg of pressure. The overall accuracy and precision of the analytical procedures for total contents (XFR method) were verified through the analysis of two standard reference materials (SRM 2710a Montana I Soil and 2711a Montana II Soil) from the National Institute of Standards and Technology (NIST). Spikes and duplicates were also used as a part of our quality control. The blank determinations were also performed in triplicate throughout all the experiments (data included as supplementary data, Table S1, see Electronic Supplementary Material).

2.3 Statistical and spatial analysis

All the analyses were carried out in triplicate, and each parameter is expressed as the average on a dry weight basis. Pearson correlation analyses ($p<0.05$, $p<0.01$) were applied in order to identify the relationships between the metal contents. Cluster analyses (CA) were performed in order to determine the different geochemical groups by means of the complete link method; the results are presented in a dendrogram with Euclidean distances. The statistical analyses were carried out with R 3.0.3 (R Development Core Team 2014). The distribution maps were performed with QGIS 2.0.2 (QGIS Development Team, 2014), using the BCN25-BTN25 (1:25,000) cartography from the Instituto Geográfico Nacional de España (2014) and the inverse distance weighting (IDW) method.

2.4 Assessment of pollution

Different environmental quality indicators were used, such as the geoaccumulation index (Igeo), the enrichment factor (EF), the contamination factor (c^i_f) and the contamination degree (Cdeg) (Table 2), on the basis of the concentrations of the earth's crust in accordance with several studies (Loska et al. 2004; Srinivasa Gowd et al. 2010). As there can be endogenous concentrations which are higher than in the terrestrial average (Loska et al. 2004), in this study, the different environmental quality indicators where calculated on the basis of the background values (Table 3) and GRL for the Galician soils (Macías and Calvo de Anta 2009).

2.4.1 Geoaccumulation index

The geoaccumulation index introduced by Müller (1979) was calculated on the basis of the background levels (Loska et al. 2004) by using the following equation:

$$\text{Igeo} = \log_2 \times (C\text{n}/1.5 \times B\text{n})$$

Table 1 Characteristics of the soils from 36 study sites

Soil	Land use	ADT	DT	Vegetation cover [lawn (mainly *Festuca sp.*, *Lolium perenne*). dominant species tree cover]
S1	I	>20,000	2	Lawn
S2	I	>20,000	2	Lawn
S3	GA	5362±836	2	Lawn. *Eucalyptus globulus*
S4	P	500±256	2	Lawn. *Salix atrocinerea*, *Acer platanoides*
S5	AA	13,110±387	2	*Cortaderia selloana*
S6	GA	8785±1265	3	Ornamental plants
S7	GA	6302±445	4	Lawn. *Acer pseudoplatanus*, *Camellia japonica*
S8	GA	6302±445	80	Lawn. *Quercus robur*, *Cedrus deodara*
S9	GA	2056±107	3	Lawn. *C. deodara, Pinus pinaster*, *Ficus carica*
S10	GA	2056±107	3	Lawn. *Pinus pinaster*
S11	GSC	20,045±870	1	Ornamental plants. *Aesculus hippocastanum*
S12	GSC	24,767±3155	1	Ornamental plants. *A. hippocastanum*
S13	BP	13,728±1002	4	Lawn. *Cedrus libani, Liquidambar styraciflua*
S14	BP	13,728±1002	5	Lawn. *Populus alba*, *Betula pubescens*, *Sorbus aucuparia*
S15	GSC	15,108±1890	1	*Thuja sp.*
S16	BP	13,728±1002	4	Lawn. *P. alba*
S17	GSC	15,108±1890	1	*Thuja sp.*
S18	RF	12,039±811	4	Lawn
S19	I	13,419±600	3	Lawn
S20	I	16,385±2345	3	None
S21	I/RF	±600	40	Lawn
S22	I/RF	±600	40	Lawn
S23	P	14,387±1728	3	Ornamental plants. *Hydrangea sp.*
S24	GA	16,385±2345	2	Lawn. *Fagus sylvatica*
S25	UP	14,387±596	3	Lawn. *Platanus× hispanica*, *P. alba*
S26	UP	14,387±596	3	Lawn. *P.* × *hispanica*, *P. alba., Hydrangea sp.*
S27	GA	14,451±1728	3	Lawn. *Q. robur*
S28	AA	17,213±3105	2	None
S29	GSC	22,000±1250	1	Ornamental plants
S30	GSC	22,542±871	1	Ornamental plants
S31	GA	8331±2851	4	Lawn. *S. atrocinerea*, *Fortunella* sp.
S32	UP	10,172±440	4	Lawn. *C. japonica*
S33	UP	10,172±440	4	Lawn. *C. japonica, A. hippocastanum*
S34	GA	>1000	1	Lawn. *Tilia tomentosa*
S35	I	>1000	10	None. *Juglans regia*
S36	P	>30,000	2	Lawn. *A. pseudoplatanus*

ATD average daily traffic (average vehicles/day), *DT* distance to road (metres), *I* industrial area, *GA* green area, *P* playground, *AA* abandoned area, *GSC* green street corridor, *RF* riverside footpath, *BP* bicycle path, *UP* urban park

*C*n is the total concentration measured in each soil and for each metal, *B*n is the background value for each metal and the factor of 1.5 is used to correct any possible lithological variations.

As it was previously indicated, Igeo was calculated on the basis of the regional background values (Table 3). The different Igeo values are classified into six degrees or classes (Table 2) where Igeo$\geq$5 indicates an enrichment of at least 100-fold with respect to the background value.

2.4.2 Enrichment factor

The enrichment factor (EF) proposed by Buat-Menard and Chesselet (1979) is based on the standardisation of the

Table 2 Assessment of pollution

Geoaccumulation indexes[a]			Enrichment factor[b]		Contamination factor[c]		Contamination degree[c]	
Igeo	Class	Pollution intensity	EF value	Enrichment category	c^if		C_{deg}	
<0	0	Unpolluted	EF<2	Minimal enrichment	$c^if<1$	Low contamination	$C_{deg}<8$	Low contamination degree
0–1	1	Unpolluted to moderately polluted	2<EF<5	Moderate enrichment	$1\leq c^if<3$	Moderate contamination factor	$8\leq C_{deg}<16$	Moderate contamination degree
1–2	2	Moderately	5<EF<20	Significant enrichment	$3\leq c^if<6$	Considerable contamination factor	$16\leq C_{deg}<32$	Considerable contamination degree
2–3	3	Moderately to Strongly	20≤EF<40	Very high enrichment	$c^if\geq6$	Very high contamination factor	$C_{deg}\geq32$	Very high contamination degree
3–4	4	Strongly	EF≥40	Extremely high enrichment				
4–5	5	Strongly to extremely polluted						
≥5	6	Extremely						

[a] Müller (1979)

[b] Buat-Menard and Chesselet (1979)

[c] Hakanson (1980)

Table 3 Physical and chemical characteristics of the 36 urban soils of Vigo City

	pH		pH	OM	P	N		ECEC	Exchangeable cations ($Cmol_{(+)}kg^{-1}$)				
	H_2O		KCl	$mg\ kg^{-1}$	$mg\ kg^{-1}$	$mg\ kg^{-1}$		$Cmol_{(+)}kg$	Al^{3+}	Ca^{2+}	K^+	Mg^{2+}	Na^+
Mean	5.87		5.21	29,600	25.66	1061		10.09	0.22	7.79	0.29	0.95	0.23
Median	5.75		5.00	29,000	18.00	1060		9.20	0.10	6.90	0.30	0.85	0.20
SD	0.67		0.76	12,900	24.76	650		6.31	0.36	5.41	0.21	0.65	0.16
Min	4.30		3.70	7000	4.00	60		0.00	0.00	0.00	0.00	0.00	0.00
Max	7.80		7.40	68,000	146.90	3400		31.50	2.20	30.20	0.90	2.80	0.90
CV	0.11		0.15	4.4	0.96	0.40		0.63	1.65	0.69	0.72	0.69	0.68
Skewness	0.80		0.95	6.1	2.69	0.60		1.24	3.36	1.81	0.87	0.87	1.37
Kurtosis	1.60		1.01	4.4	8.68	0.61		2.97	14.53	6.22	0.51	0.63	3.35
25th percentile	5.50		4.70	18,500	10.70	1300		6.93	0.00	5.43	0.10	0.50	0.20
75th percentile	6.20		5.70	37,800	28.50	1900		13.20	0.30	9.95	0.40	1.30	0.30
Total elements concentrations content ($mg\ kg^{-1}$)													
	Ba	Ca	Cr	Cu	Fe	Mg	Mn	Na	Ni	Pb	Si	Sr	Zn
Mean	516.75	4345.97	68.55	66.12	27,944.15	6053.88	531.55	7214.39	32.01	96.29	272,944.4	58.82	149.03
Median	528.48	3412.76	65.10	61.60	25,756.28	5841.28	517.20	6634.00	28.00	81.62	272,500	55.61	149.98
SD	152.68	5044.32	28.62	33.83	10,257.92	2154.88	165.84	3766.78	12.29	62.33	16,260.28	19.40	35.57
Minimum	237.82	1734.62	33.48	22.56	11,745.64	2488.96	168.09	2108.00	11.50	34.38	248,000	29.22	59.05
Maximum	1145.04	32,433.36	195.30	208.22	52,017.71	9878.06	879.24	1,7856.00	60.00	259.11	314,000	139.63	234.35
CV	0.30	1.16	0.42	0.51	0.37	0.36	0.31	0.52	0.38	0.65	0.06	0.33	0.24
Skewness	1.84	5.23	2.47	2.30	0.47	0.13	0.21	1.02	0.72	1.49	0.50	2.18	−0.14
Kurtosis	7.45	29.39	10.25	8.13	−0.59	−0.95	−0.30	1.10	−0.25	1.32	−0.15	8.05	0.17
25th percentile	424.99	2666.47	51.15	42.95	19,576.07	5841.28	517.20	6634.00	28.00	81.62	259,500	48.91	121.86
75th percentile	572.52	4229.65	79.52	83.07	36,500.77	8147.46	646.50	8928.00	40.00	103.64	282,250	66.16	178.11
BL	220	–	80	45	–	–	850	–	65	55	–	–	100
A	7	–	2	3	–	–	0	–	0	12	–	–	0

A number of samples exceeding the Generic Reference Level for urban soils in the Galician soils (600 mg kg^{-1} for Ba, 100 mg kg^{-1} for Cr, 100 mg kg^{-1} for Cu, 1500 mg kg^{-1} for Mn, 100 mg kg^{-1} for Ni, 100 mg kg^{-1} for Pb, 500 mg kg^{-1} for Zn) (Macías and Calvo de Anta 2009), *CV* coefficient of variation, *SD* standard deviation, *CC* coarse component, *OM* organic matter, *P* available phosphorus, *N* total Kjeldahl nitrogen, *CEC* effective cation exchange capacity, *BL* background level (mg kg^{-1}) for Galician soils (Macías and Calvo de Anta 2009), elements measured against one used as a reference (Al, Fe, Mn, Sc and Ti). Some authors (Covelli and Fontolan 1997; Loska et al. 2004) indicated that it is better to use than other environmental indices. In this work, Fe was used as a reference value for the standardisation following the criteria of Daskalakis and O'Connor (1995) and Acevedo-Figueroa et al. (2006). They indicated that (1) the geochemistry of Fe and many trace metals are similar both in oxic and anoxic conditions; (2) its geochemistry is similar to that of many trace metals and (3) its natural sediment concentration tends to be uniform. As a result, there are five categories defined for the different EF values (Table 2). The EF was calculated according to the following equation:

$$\mathrm{EF} = (\mathrm{Metal}/\mathrm{Fe})_{\mathrm{soil}}/\ (\mathrm{Metal}/\mathrm{Fe})_{\mathrm{background}}$$

2.4.3 Contamination factor and contamination degree

The global assessment of the contamination in the soil was carried out by means of the contamination factor (c^{i}_{f}) and the degree of contamination (*C*deg). According to Hakanson (1980), c^{i}_{f} is the contamination factor for each element and *C*deg represents the sum of all the contamination factors. Each contamination factor was calculated following the equation:

$$c^{i}_{f} = c^{i}_{0\text{-}1}/\ C^{i}_{n}$$

$c^{i}_{0\text{-}1}$ is the average metal content from at least five sampling points; c^{i}_{n} is the pre-industrial concentration of each metal (the regional background levels, Table 3). Table 2 shows the classes proposed by Hakanson (1980) into which both the contamination factor and the degrees of contamination values are included.

3 Results and discussion

3.1 Properties of urban soil samples

The values of the different soil characteristics are shown in Table 3. The high content of coarse component (18.2± 7.3 %; 5–50.4 %) is due to the materials used to refill the areas planned for gardens as there are different artefacts and rubble coming from those materials. The $\mathrm{pH_{H2O}}$ is between 4.30 and 7.79 (average 5.87); the highest values are from soils close to beach areas (S2, S35) and the influence of $CaCO_3$ from the mollusc shells probably influence the pH of the soils. In fact, most of the soils are moderately to slightly acid as a result of the influence of the geological parent material of the soil (granite rocks), which was attenuated by the construction waste materials among other factors (Li et al. 2013).

The average of the N Kjeldahl concentration is 1061± 650 mg $\mathrm{kg^{-1}}$ (60–3400 mg $\mathrm{kg^{-1}}$). The values are enough to support the growth of vegetation (Li et al. 2013) and similar to those measured in soils of other cities such as Anji, China (540–1310 mg $\mathrm{kg^{-1}}$; Yang et al. 2014), Seville (400–5200 mg $\mathrm{kg^{-1}}$; Ruiz-Cortés et al. 2005) and Denver-Boulder, Co, USA (2100 mg $\mathrm{kg^{-1}}$; Golubiewski 2006). Nevertheless, the OM values are, in general, low 29,600± 12,900 mg $\mathrm{kg^{-1}}$ (and lower than those found by other authors for urban soils in Torino (5200–82,700 mg $\mathrm{kg^{-1}}$; Biasioli et al. 2006) or Seville (8400–80,500 mg $\mathrm{k^{-1}}$; Ruiz-Cortés et al. 2005).

The average of the available P contents is 25.65± 24.75 mg $\mathrm{kg^{-1}}$ (4–146.90 mg $\mathrm{kg^{-1}}$), and they are congruent not only with the soil types but also with the undergoing processing and modifications on a periodic basis. However, in some soils (S29, S30, S31), the content of the available P is over 70 mg $\mathrm{kg^{-1}}$, and there are different reasons for that as an overuse of phosphate fertilisers, the OM content or because they are in more disturbed soils (transport, industry, etc.) (Pouyat et al. 2007; Li et al. 2013).

The average of the cation exchange capacity (CEC) of the soils is 10.56 $\mathrm{cmol_{(+)}}$ $\mathrm{kg^{-1}}$ (it ranges from 4.10 to 31.36 $\mathrm{cmol_{(+)}}$ $\mathrm{kg^{-1}}$). In most of the soils, this value is lower than 15 $\mathrm{cmol_{(+)}}$ $\mathrm{kg^{-1}}$, but there are some exceptions (31.36 $\mathrm{cmol_{(+)}}$ $\mathrm{kg^{-1}}$; S35) detected in areas near beaches where depositions of mollusc shells would influence the CEC as in the case of pH. In fact, the content of exchangeable Ca^{2+} is high in the soils from these areas, although in most of the soils it is also the principal exchangeable cation and the contents vary from 0.99 to 30.17 $\mathrm{cmol_{(+)}}$ $\mathrm{kg^{-1}}$). In addition, the exchangeable complex saturation is generally high (average value 96.29 % and range 49.02–100 %). In any case, the CEC is similar to the urban soils from other cities like Seville (2.1–35.4 $\mathrm{cmol_{(+)}}$ $\mathrm{kg^{-1}}$; Ruiz-Cortés et al. 2005), Torino (4.7 to 26.3 $\mathrm{cmol_{(+)}}$ $\mathrm{kg^{-1}}$; Biasioli et al. 2006) or Beijing (4.23–20.29 $\mathrm{cmol_{(+)}}$ $\mathrm{kg^{-1}}$; Xia et al. 2013).

As urban soils are subjected to garden-tending processes, they usually reveal higher values for pH, OM, N, available P, CEC and exchangeable complex saturation than rural or natural soils. The direct addition of fertilisers, pruning waste, fallen leaves and organic residues originating from vegetation and animals influence the soil urban properties, but they also receive indirect additions of N and P through construction waste materials and the emissions of vehicles (Pouyat et al. 2007; Lorenz and Lal 2008).

Attending to the previous results, the urban soils from Vigo have low pH and OM contents that may increase the mobility and availability of different contaminants. On the other hand, a high amount of available P may counteract this effect by reducing mobility (Kabata-Pendias 2010).

3.2 Metal concentrations

Tables 3 and 4 show the total concentrations of the analysed metals and the corresponding degree of enrichment with respect to the background level. The average values for enrichment factor are 2.64 (Ba), 2.43 (Cr), 1.91 (Cu), 0.68 (Mn), 0.52 (Ni), 2.07 (Pb) and 1.73 (Zn).

The total content of Ba varies between 237.82 and 1145.04 mg kg^{-1} (an average of 516.75 mg kg^{-1}), and seven soils exceed the corresponding GRL for urban soils (600 mg kg^{-1}) (Fig. 2). Nevertheless, the average Igeo value (0.59) indicates that, in general, the soils are not contaminated or that the contamination is moderate. The EF data (0.93–4.57) also indicates a moderate enrichment in Ba.

The total contents of Cr range between 33.48 and 195.30 mg kg^{-1}(average 68.55 mg kg^{-1}) and only two soils (S9 and S27) exceed the GRL for urban soils (100 mg kg^{-1}) (Fig. 2). With the exception of S9 (0.70), all the soils reveal negative Igeo values (an average of ¯0.90) reflecting no contamination. However, the EF values show a moderate enrichment (they range from 1.18 to 3.63). The results are in agreement with Kabata-Pendias (2010) as Cr is an element with low mobility under oxidation conditions and with pH close to neutral.

According to the background levels, Cu contents are rather high, that vary from 22.56 to 208.22 mg kg^{-1}, (average 66.12 mg kg^{-1}) but only three soils exceed the GRL (100 mg kg^{-1}) (S3, 104.11 mg kg^{-1}; S4, 208.22 mg kg^{-1} and S14, 130.14 mg kg^{-1}) (Fig. 3). The average Igeo value was ¯0.18 (¯1.84 to 0.70) showing that in general, there is no contamination but according to EF values few soils were classified as with significant enrichment (EF=6.61).

Lead contents are high according to the background levels. They range from 34.38 to 259.11 mg kg^{-1} (average 96.29 mg kg^{-1}), and 12 soils exceed the GRL (100 mg kg^{-1}) (Fig. 4). Although the average Igeo value is indicative of uncontaminated soils (¯0.02), it is necessary to highlight the existence of 11 soils classified as Class 1 and five soils with moderate contamination (Class 2). In addition, the average EF values indicate moderate enrichment (0.48–7.88).

The levels of Mn (Table 3) are below the GRL (1500 mg kg^{-1}) (Fig. 3). Furthermore, both the average Igeo values (¯1.34) and the EF (0.68) reveal no contamination or minimum enrichment (Table 4). As with Mn, the levels of Ni are below the GRL (100 mg kg^{-1}) (Fig. 4) but they are also lower than the regional background level (65 mg kg^{-1}). They range between 11.50 and 60 mg kg^{-1} (average 32.01 mg kg^{-1}) and accordingly the average Igeo

Table 4 Geoaccumulation indexes (Igeo), enrichment factor (EF), contamination factors (c^{i}_{f}) and contamination degree (*C*deg) of metals in the urban soils of the Vigo city

Geoaccumulation index							
	Ba	Cr	Cu	Mn	Ni	Pb	Zn
Minimum	−0.4	−1.84	−1.58	−2.92	−3.08	−1.26	−1.34
Maximum	1.79	0.7	1.62	−0.53	−0.7	1.65	0.64
Mean	0.59	−0.9	−0.18	−1.34	−1.71	−0.02	−0.06
Class	1	0	0	0	0	0	0
Enrichment factor							
	Ba	Cr	Cu	Mn	Ni	Pb	Zn
Minimum	0.93	1.18	0	0.36	0.34	0.48	0.71
Maximum	4.57	3.63	6.61	1.43	0.91	7.88	4.17
Mean	2.64	2.43	1.91	0.68	0.52	2.07	1.73
Class	Moderate	Moderate	Minimal	Minimal	Minimal	Moderate	Minimal
Contamination factor							
	Ba	Cr	Cu	Mn	Ni	Pb	Zn
Minimum	1.08	0.42	0.5	0.2	0.18	0.63	0.59
Maximum	5.2	2.44	4.63	1.03	0.92	4.71	2.34
Mean	2.35	0.86	1.47	0.63	0.49	1.75	1.49
Categories	Moderate	Low	Moderate	Low	Low	Moderate	Moderate
Contamination degree							
Minimum	3.59						
Maximum	21.29						
Mean	9.03						
Classes	Moderate contamination degree						

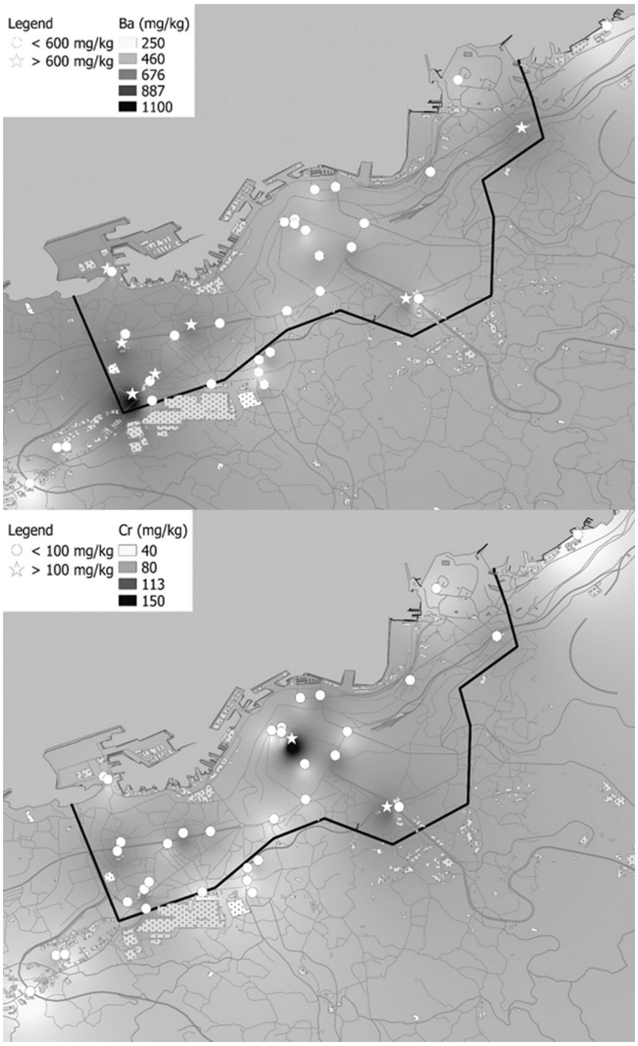

Fig. 2 Spatial distribution of Ba and Cr concentrations in the urban soils of Vigo City

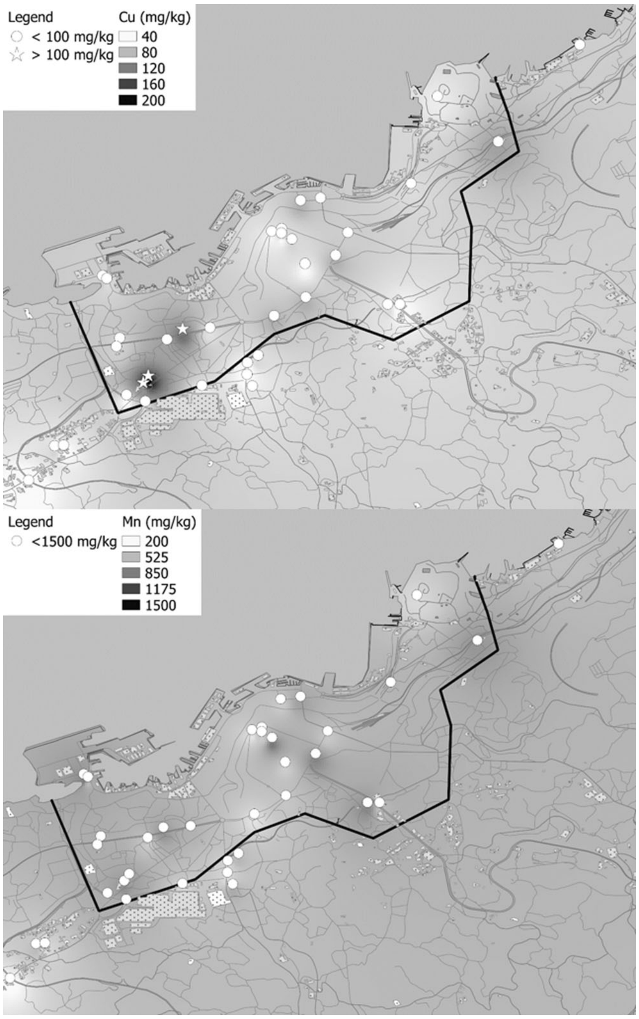

Fig. 3 Spatial distribution of Cu and Mn concentrations in the studied soils

(¯1.71) values indicate there is no contamination and the EF values (0.51) that there is minimum enrichment or deficient.

Zinc contents are relatively high (ranging from 116 to 268 mg kg^{-1} and an average of 192.92 mg kg^{-1}) (Fig. 5) compared to the background levels, but they do not exceed the GRL (500 mg kg^{-1}). In fact, according to the average Igeo values, there is contamination but according to the EF values, there is Zn moderate enrichment in the 12 soils where EF>2.

In general, the metal contents in the studied soils are below their corresponding GRL and although some soils have high contents suggesting contamination, none of them can be classified as strongly contaminated or significantly enriched. Comparing the results with those obtained in the urban soils from other Spanish cities, in general, the metal contents in Vigo are lower than in cities with more population like Seville (Biasioli et al. 2007) or Madrid (De Miguel et al. 2007), but also in smaller cities with potentially polluting industrial activity like Avilés (Gallego et al. 2002) where there is a Zn smelter or A Coruña (Cal-Prieto et al. 2001) or Huelva (Guillén et al 2012), both with a petrochemical complex.

3.3 Contribution of particular metals to the contamination of soils in Vigo

In order to assess the contamination of the soils, the contamination factor and the contamination degree were calculated. As it was previously indicated by the Igeo and EF values, the new results also reveal low contamination factor (c^{i}_{f}) for Cr, Mn and Ni, and moderate for Ba, Cu, Pb and Zn (Table 4). The degree of metal contamination (C*deg*) indicates moderate contamination (9.03) (Table 4). The contribution of each metal to this index is as follows: Ba (26 %), Pb (19.38 %), Cu (16.57 %), Zn (16.50 %), Cr (9.49 %), Mn (6.92 %) and Ni (5.45 %).

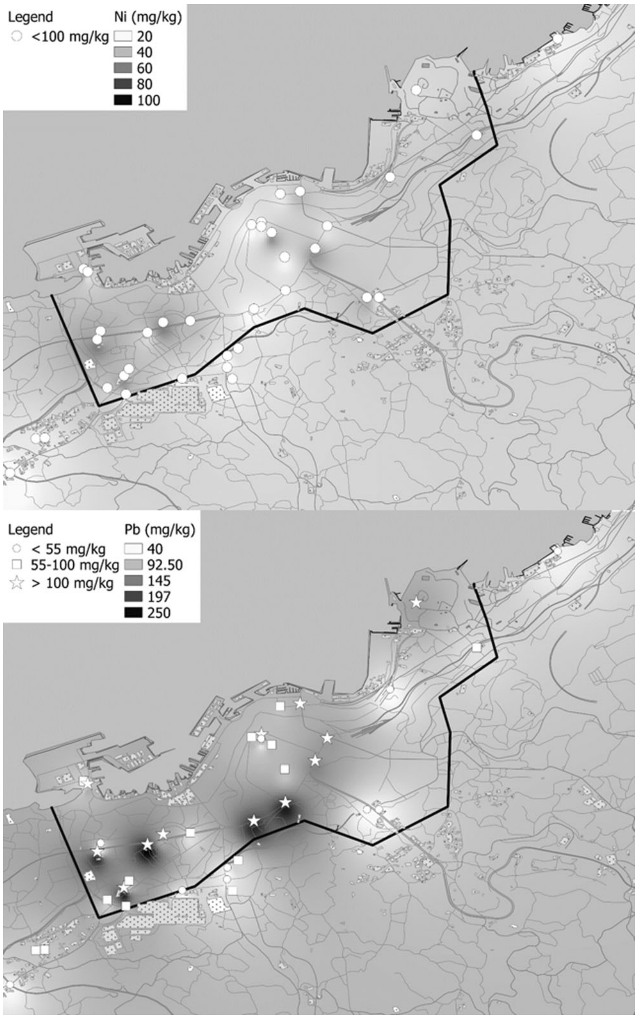

Fig. 4 Spatial distribution of Ni and Pb concentrations in the studied soils

3.4 Correlation of metals and source identification

By means of GIS techniques, it is possible to represent in maps the spatial distribution of the elements in the soils. These techniques also allow to relate them to "hot-spots" (Rodríguez-Salazar et al. 2011) by means of using interpolation methods such as Kriging or IDW. Inverse distance weighting (IDW) uses easier parameters than Kriging, giving rise to results that are more easy to define and to understand. When there are outliers or very different values (Azpurua and Ramos 2010), IDW interpolation gives better results than the model Kriging or SPLINE. As urban soils are very heterogeneous environments, with very high spatial variability in physical, chemical and biological properties, IDW is a better choice than Kriging. In fact, different studies about metal distribution in urban soils (Zhang 2006; Lee et al. 2006; Carr et al. 2008; Slavik et al. 2012; Karim et al. 2014) indicated that using IDW is a good choice for identifying a contamination pattern, or determining the variation of soil metal concentration.

The sampling points, transport routes and industrial zones are indicated in Fig. 1, and the pollution hotspot can be identified with the distribution maps (Figs. 2, 3, 4 and 5). They suggest a strong influence of industries and traffic on soil metal accumulation as was also indicated for Shanghai urban soils by Shi et al. (2008). The distribution of Ba (Fig. 2), Cu (Fig. 3) and Pb (Fig. 4) was mainly associated to transport routes and to the city's industrial areas. Chromium seems to be somewhat more localised (S6, S7 and S8, Fig. 2), and the garden-tending processes (protectors or wooden fences treated with chromate) is the most probable origin. On the other hand, the distribution of Mn, Ni and Zn is more diffuse (Figs. 3, 4 and 5, respectively) and the distribution maps also showed there are no hotspot in accordance with the previously environmental factors that revealed no contamination.

The correlation (Table 5) and cluster (Fig. 6) analyses were performed with the total metal content of the soils. Together with the Igeo, EF and C*deg* (Table 4), it is possible to explain the relationships between the metals and their probable origins. Is it possible also to distinguish between "natural", "mixed" and "urban" origins. Two possible sources in both mixed and urban contents were identified:

i). Natural: the cluster of natural origin is made up of Na and Si. Their origin in the soils is supposed to be lithogenic as to the parent material is mainly granite.

ii). Mixed: the two sources identified are mainly natural and urban. There is a cluster made up of Ca and Sr. Both elements, but also Mg, were not included in the "natural" cluster, therefore, their origin was supposed to be mixed (anthropogenic and natural). They probably undergo different geochemical changes with respect to the original sources (De Miguel et al. 1997). They could be resuspended in the air and latter deposited in the soils since they are present in particles of cement from building materials and concrete manufacturing companies (Santacatalina et al. 2012).

The second cluster classified as mixed origin is made up of Mg, Cr, Fe, Mn and Ni (Fig. 6). Except for Cr, the other studied metals are always below the GRL for the studied soils. The concentration of each element that made up the mixed origin cluster is positive and significantly correlated with the concentration of the other elements that belong to the same cluster (Table 5). These facts suggest the existence of a common source (natural or anthropogenic). It is worth highlighting that, except for Cr, the enrichment factors are low, suggesting that a priori the origin is lithogenic (Massas et al. 2013). However, the negative correlation between their contents and the ones of the elements identified as natural origin (Na and Si), along with no correlation with the ones of Ca (Table 5), indicate a mixed origin.

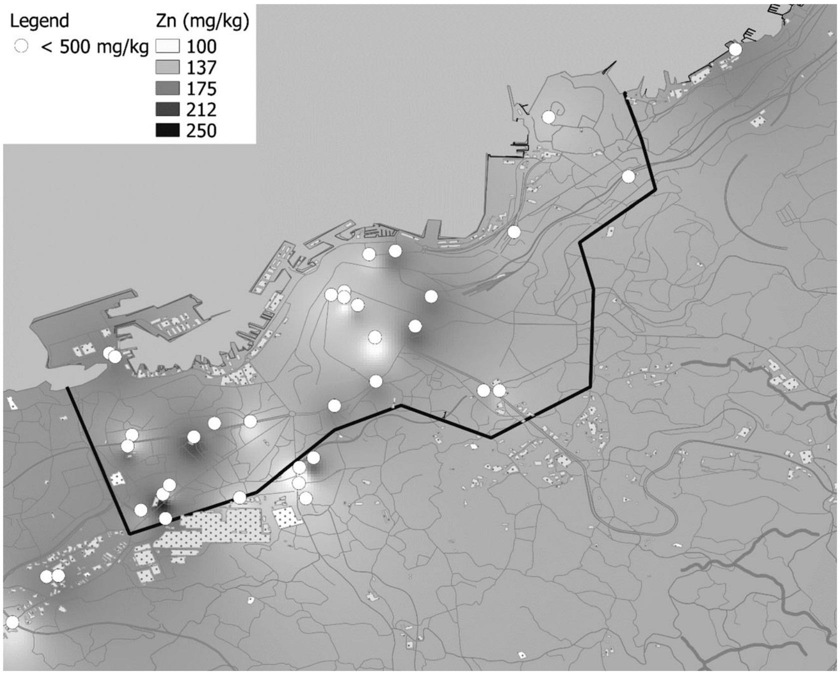

Fig. 5 Spatial distribution of Zn concentrations of the studied samples in Vigo City

iii). Urban: the urban origin cluster is made up of Ba, Cu, Pb and Zn. The maps (Figs. 2, 3, 4 and 5, respectively) show the distribution of these elements in the study area and the hot spots for those that are higher than the GRL (7 soils of Ba, 3 soils of Cu and 12 soils of Pb). There are no hot spots for Zn as the contents did not exceed the GRL. Nevertheless, the Zn contents are relatively high and the EF values indicate there is moderate enrichment. As a result, the urban or anthropogenic origin is deduced for Ba, Cu, Pb and Zn as the Igeo, EF and c^{i}_{f} values indicated there is enrichment and moderate contamination of the soils by Ba, Cu, Pb and Zn.

Moreover, according to the cluster analysis (Fig. 6), it is possible to identify two urban sources (one for Ba and another one for Cu, Pb and Zn). In addition, the positive correlation between their concentrations (Table 5) also suggests the common origin. The distribution map of Pb (Fig. 4) shows that traffic is the main probable origin as Pb from petrol is the main source of Pb in soils

Table 5 Pearson's correlation matrix for metal concentrations in the urban soils of Vigo City

	Ba	Ca	Cr	Cu	Fe	Mg	Mn	Na	Ni	Pb	Si	Sr	Zn
Ba	1.00												
Ca	−0.27	1.00											
Cr	0.20	−0.24	1.00										
Cu	0.25	0.00	0.22	1.00									
Fe	0.39*	−0.35*	0.78**	0.28	1.00								
Mg	0.69**	−0.28	0.59**	0.45**	0.71**	1.00							
Mn	0.47**	−0.05	0.78**	0.44**	0.82**	0.83**	1.00						
Na	−0.45**	0.27	−0.43**	−0.38	−0.70**	−0.53**	−0.58**	1.00					
Ni	0.47**	−0.19	0.73**	0.43**	0.83**	0.79**	0.82**	−0.70**	1.00				
Pb	0.15	−0.10	0.10	0.35*	−0.01	0.12	0.03	−0.15	0.24	1.00			
Si	−0.22	−0.16	−0.50**	−0.35*	−0.70**	−0.47**	−0.66**	0.62**	−0.67**	−0.07	1.00		
Sr	0.52**	0.33*	−0.21	0.03	−0.20	0.09	−0.04	−0.04	−0.05	0.05	−0.10	1.00	
Zn	0.25	0.18	0.24	0.38*	0.23	0.45**	0.40*	−0.22	0.42**	0.63**	−0.37*	0.12	1.00

*$p<0.05$; **$p<0.01$

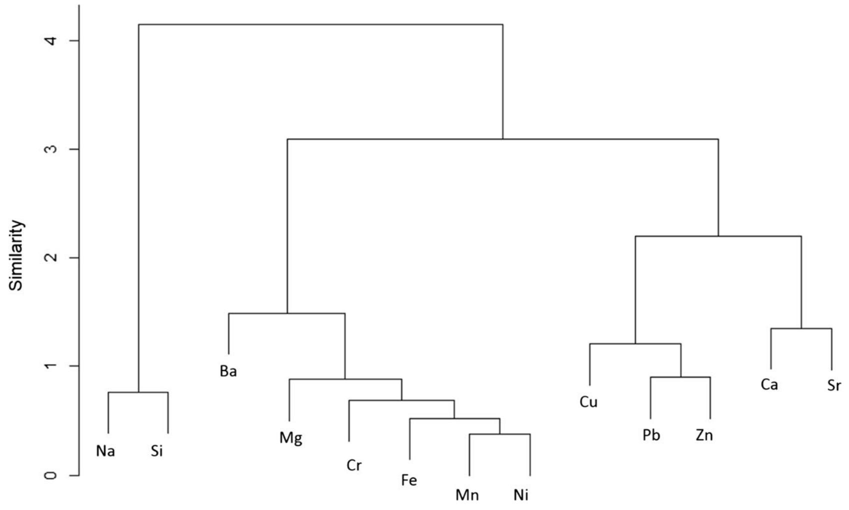

Fig. 6 Dendrogram of the cluster analysis (*CA*) based on correlation coefficients using the complete link method in the urban soils of Vigo City

(43.51 % average) (Rodríguez-Seijo et al. 2015). The Pb comes from the additives used in petrol, that were later banned, and their accumulation in the soil during decades, as well as from the combustion of fuel oil, the wear and corrosion of metal vehicle parts and urban fixtures, paints, etc. Furthermore, the main sources of Cu and Zn are lubricants, abrasion processes of tyres, brakes, etc. and corrosion of metal parts in contact with fuel. Other sources of Zn in urban environments are vulcanised tyres and galvanised components of vehicles, street furniture, roofs, etc. (De Miguel et al 1997; Sutherland 2000; Saeedi et al 2012).

The origin of Ba in the soils is also assigned to an urban origin. The total Ba contents are high as it was previously indicated. For decades, the importance of Ba in areas with intense road traffic was ignored, but different processes may give rise to an increase of Ba contents in soils. Barium is a constituent of alloys, paint, ceramic, glass or plastic cements but the friction processes of tyres and brakes also release Ba to the environment (Monaci and Bargagli 1997; Sutherland 2000; Fernández–Espinosa and Ternero-Rodríguez 2004; Minguillón et al. 2014). Even traffic is an important source of Ba; according to several authors, Ba can be forming part of very fine particles and it can be transported long distances. As a result, its origin in the soils is mixed, from natural and anthropogenic sources, as it was already stated in previous studies (De Miguel et al. 1997; Zechmeister et al. 2005; Chen et al. 2014).

Three groups were obtained; the first one named the “urban” group is influenced by transport. The second is composed of elements with industrial origin or mixed and the third one encompasses elements from natural origin. Nevertheless, the elements included in the second and third group can vary according to geology and industrial activity (De Miguel et al. 1997; Wei et al. 2010; Rodríguez-Salazar et al. 2011, Saeedi et al. 2012; Oliva and Fernández-Espinosa 2007).

4 Conclusions

In general, the metal contents in the studied soils are below their corresponding GRL, and although some soils have high metal concentrations suggesting contamination, none of them can be classified as strongly contaminated or significantly enriched.

Barium, Cu and Pb explain more than 61 % of the moderate contamination degree of the studied soils and they come from the same source.

There are three main different sources that determine the metal contents: “natural”, “mixed” and “urban” origins. The urban origin is mainly explained by traffic.

The obtained results will allow to help in the selection of “hot-spots” to carry out more in-depth studies into bioavailability.

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Compliance with ethical standards This research does not contain any studies with human or animal subjects. Informed consent was obtained from all individual participants included in the study.

Conflict of interest The authors declare that they have no competing interest.

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