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Accumulation and sources of heavy metals in urban topsoils: a case study from the city of Xuzhou, China

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Abstract The knowledge of the variability, the anthropogenic versus natural origin and corresponding environmental risk for potentially harmful elements in urban topsoils is of importance to assess human impact. The aims of the present study were: (1) to assess the distribution of heavy metals (Sn, Li, Ga, Ba, Fe, Mn, Co, Be, Ti, Al, Hg, Cr, Sb, As, Bi, Pd, Pt, Au, Ni, Cd, Zn, Cu, Pb, Se, Mo, Sc and Ag) in urban environment; (2) to discriminate natural and anthropogenic contributions; and (3) to identify possible sources of pollution. Multivariate statistic approaches (principal component analysis and cluster analysis) were adopted for data treatment, allowing the identification of three main factors controlling the heavy metal variability in Xuzhou urban topsoils. Results demonstrate that Hg, Cr, Sb, As, Bi, Pd, Pt, Au, Ni, Cd, Br, Zn, Cu, S, Pb, Se, Mo, Sc and Ag could be inferred to be tracers of anthropogenic pollution, whereas Al, Ti, Ga, Li, V, Co, Pt, Mn and Be were interpreted to be mainly inherited from parent materials. Iron, Ba, Sn, Pd and Br were interpreted to be affected by mixed sources.

Keywords Heavy metals · Urban topsoil pollution · Multivariate statistics · Xuzhou

Introduction

Human activities, in general, exert a substantial impact on the environment. Population and industrial centers pollute the air, water and soil, causing a decline in the quality of the environment. People living in industrial cities are particularly exposed to this decline in environment quality leading to human health concerns. Urban environment affects and are affected by natural cycles, where air, water and soil are altered by products ultimately returned to the environment in the form of emissions and wastes.

In the past few decades, heavy metals have been the subject of much attention because of their peculiar pollutant characteristics: (1) they do not decay with time, unlike many organics and radionuclides; (2) they can be necessary or beneficial to plants at certain levels, but can also be toxic when exceeding specific thresholds; (3) they are always present at a background level of non-anthropogenic origin, their input in soils being related to weathering of parent rocks and pedogenesis; and (4) they often occur as cations, which strongly interact with the soil matrix; consequently, heavy metals in soils, even at high concentrations, may be present in inert and not harmful forms, but they can become mobile as a result of changing environmental conditions or by saturation beyond the buffering capacity of the soil. This situation is referred to as a ‘‘chemical timing bomb’’ (Facchinelli et al. 2001).

Heavy metals in urban soils have been shown to be very useful tracers of environmental pollution (Davies et al. 1984; Burguera et al. 1988; Bacon et al. 1992; Kelly et al. 1996; Manta et al. 2002). Urban soils are known to have peculiar characteristics such as unpredictable layering, poor structure, and high concentrations of heavy metals (Tiller 1992). They are the ‘‘recipients’’ of large amounts of heavy metals from a variety of sources including industrial wastes, vehicle emissions, coal burning waste and other activities. In areas where pubic gardens and parks are exposed to significant pollution levels, dust from the ground may have toxic effects as a consequence of inhalation or ingestion by humans, particularly children, which poses major health hazards (Culbard et al. 1988). Furthermore, any contamination of urban soils could cause, in turn, groundwater contamination because metals of the polluted soils tend to be more mobile than those of unpolluted ones (Steinmann et al. 1997; Wilcke et al. 1998; Manta et al. 2002).

Around the world many studies have evaluated the heavy metals concentrations in urban soils. Concentrations of heavy metals in urban topsoils vary considerably across cities (Table 1) depending, among other factors, on the density of industrial activities in the area and technologies employed, as well as on local weather conditions and wind patterns (Ordòñez et al. 2003).

As the amount of bromine in urban topsoils is frequently linked with traffic and the amount of sulfur is associated with coal burning, we discuss bromine and sulfur together with heavy metals.

The present study was carried out as a preliminary survey on Xuzhou urban topsoil contamination. The aims of the study were: (1) determine concentrations of heavy metals, sulfur and bromine (Sn, Li, Ga, Ba, Fe, Mn, Co, Be, Ti, Al, Hg, Cr, Sb, As, Bi, Pd, Pt, Au, Ni, Cd, Br, Zn, Cu, S, Pb, Se, Mo, Sc and Ag); (2) to define their natural or anthropogenic source; (3) to identify possible non-point source of contamination. A multivariate statistical and Geographical Information System (GIS)-based approach was adopted to assist the interpretation of geochemical data. Principal component analysis (PCA) and derivative methods have been widely used in geochemical applications to identify pollution sources and to apportion natural versus anthropogenic contribution (Facchinelli et al. 2001). Clustering analysis (CA) is often coupled to PCA to check results and provide grouping of individual parameters and variables. GIS is increasingly used in environmental pollution studies because of its ability to identify non-point source contaminants (Corwin et al. 1996) and also provides a very important role in geochemical anomaly separation by giving a good visual aid in reasoning natural sources from anthropogenic sources.

Materials and methods

The study area (the city of Xuzhou) is located in the northwestern part of Jiangsu, one of the provinces of China, the geographical position being 33°43′–4°58′N, 116°22′–118°40′E. Xuzhou (China) is an industrial city and current urban population exceeds 1,200,000 inhabitants. The main wind direction is from the northwest, although, in general, wind velocities are low, even approaching zero at times. This enhances the deposition of particulates within the city.

A total of 21 topsoil samples (depth = 0–10 cm) were collected within the city of Xuzhou (Fig. 1). Sampling sites were selected where chemicals (such as fertilizers, pesticides) and sewage sludge had not been used. To avoid effects due to the differential uptake of metals by vegetation, sampling was carried out where plants with superficial roots are not present. At each sampling point, three sub-samples, with a 20×20 cm surface, were taken and then mixed to obtain a bulk sample. Such a sampling strategy was adopted in order to reduce the possibility of random influence of urban waste. All the samples were collected with a stainless steel spatula and kept in PVC packages.

Table 1 Average heavy metal concentrations (mg/kg) in urban soils from different cities in the world

City	Hg	Pb	Zn	Cu	Cd	Cr	Co	Ni	V	Sb	Mn	Reference
Rome		330.8		0.31								1
Pittsburg	0.51	398			1.2							2
Boston		800										3
Warsaw		57	166	31	0.73	32	51	12			337	4
Hamburg		218.2	516	146.6	2.0	95.4		62.5			750	5
Salamanca		53.1			0.53							6
Coruña		309	206	60	0.3	39	11	28		3		7
Central Madrid		621										8
Madrid		161	210	71.7		74.7	6.42	14.1	30			9
Bangkok		47.8	118	41.7	0.29	26.4		24.8			340	10
Aberdeen		944	58.4	27			23.9	6.4	14.9		286	11
Birmingham		570										12
Glasgow		216	207	97	0.53							13
London boroughs		294	183	49	1.0							14
Hong Kong		93.4	168	24.8	2.18							15
Manila		213.6	440	98.7	0.57	114			20.9		1,999	16

References: *1* Angelone et al. (1995); *2* Carey et al. (1980); *3* Spittler et al. (1979); *4* Czarnowska (1980); *5* Lux (1986); *6* Sánchez-Camazano et al. (1994); *7* Cal-Prieto et al. (2001); *8* Pellicer 1985; *9* De Miguel et al. (1998); *10* Wilcke et al. (1998); *11* Paterson et al. (1996); *12* Department of the Environment (1982); *13* Gibson et al. (1986); *14* Culbard et al. (1998); *15* Li et al. (2001); *16* Pfeiffer et al. (1988)

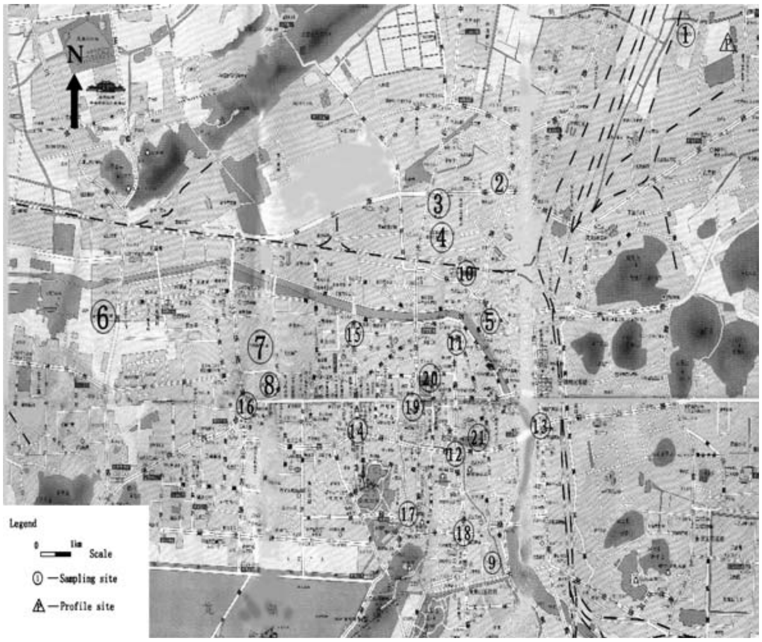

Fig. 1 Map of the Xuzhou city (Jiangsu) with location of sampling sites of topsoils

The soils samples were air-dried and sieved through a 2-mm sieve. The major element concentrations for Ti, Fe, Cr, Al, Ga, Pb and S were determined by X-ray fluorescence spectrometry (XRF, Philips PW1400 apparatus), on bulk-sample pressed, boric-acid backed pellets. The accuracy of determinations was checked by using certified reference materials. Analytical errors were below 1% for Al, below 3% for Ti, Fe, and below 10% for Pb, Ga and Cr.

Samples (approx. 0.2 g) were dissolved in a hot HF–HNO_3–HCl acid mixture (approx. 15 mL), and refluxed with the acid mixture if the sample was only partly dissolved. Sc, Ba, Li, Cd, Be, Br,Co, Cu, Mn, Ni, V, Zn, Mo, Pt, Pd and Au concentrations were measured by inductively coupled plasma mass spectrometry (ICP-MS). The elements As, Sb, Se, Hg, Bi, Ag and Sn were determined by inductively coupled plasma atom emission spectrometry (ICP-AES). All calibration standards were prepared in the acid matrix used for the soil samples. Caution was used in preparing and analyzing samples to minimize contamination from air, glassware and reagents, which were all of suprapur quality. Replicated measures of standard reference materials (ESS-1 and ESS-2, provided by China Environmental Monitoring General Station), reagent blanks and duplicated soil samples (approx. 20 of the total number of soil samples was used for this purpose) randomly selected from the set of available samples were used to assess contamination and precision. The analytical precision, measured as relative standard deviation, was routinely between 5% and 6%, and never higher than 10%.

Results and discussion

Accumulation of heavy metals

Summary statistics for the analyzed elements in all the studied samples are presented in Table 2. Compared to their levels in natural soils of China, Zinc, Cd, As, Hg, Sb, S, Br, Sn and Ag concentrations are higher for topsoils, while the concentrations of Fe, Se, Sc, Ba, Bi, Pb, Cu, Ni and Cr are only slightly higher for topsoils. High concentrations coupled with high standard deviation values suggest anthropogenic sources for these elements.

The Mn, Mo, Be, Ti, Al, Ga, V, Li and Co concentrations do not exceed arithmetic values in natural soils of China. Moreover, these metals display quite homogeneous distributions across the city and therefore lower standard deviations, thus suggesting a major natural source.

It is noteworthy that concentration of As in sample 2 surpasses the intervention value for Dutch legislation. It is important to take action to find out the probable pollution source and carry out of As remediation in this area if necessary.

Unleaded gasoline was introduced in China during the late 1980s, when a new generation of cars was equipped with catalytic converters. Exploiting the catalytic properties of the noble metals (platinum, palladium and rhodium), the converters reduce the emission of CO, not combusted hydrocarbons and NO_X, favoring redox reactions and leading to the formation of less dangerous compounds (CO_2, N_2, H_2O). However, the introduction

Table 2 Descriptive statistics of the concentrations (mg/kg) of the elements in urban topsoils from the city of Xuzhou (Pt, Au, Pd: μg/kg, unless otherwise stated)

Sample size = 21	Al(%)	Fe(%)	Ti	Ag	Se	Sc	Ga	Ba	Li	Bi
Mean	6.05	3.37	3638.2	0.28	0.42	17.4	15.2	485	35.6	0.42
Median	5.87	3.29	3540	0.19	0.31	17.0	15	470	36	0.32
SD	0.64	0.48	268.8	0.24	0.27	2.63	1.83	54.1	5.67	0.39
Minimum	5.13	2.66	3241	0.06	0.13	13	12	425	25	0.23
Maximum	8.04	4.15	4349	1.1	1.0	25	18	628	47	2.1
Range	2.91	1.49	1108	1.04	0.87	12	6	203	22	1.87
Kurtosis	3.29	−1.3	1.06	6.37	−0.29	2.46	−1.1	2.4	−0.07	19.6
Skewness	1.54	0.17	1.09	2.37	1.06	0.85	−0.14	1.7	−0.066	4.3
Unpolluted soils[a]	6.62	2.94		0.13	0.29	11.1	17.5	469	32.5	0.37
Sample size = 21	V	Pb	Cu	Zn	Cd	Ni	Co	Cr	As	Hg
Mean	76	43.3	38.2	144.1	0.54	34.3	11.7	78.4	39.8	0.29
Median	74	36	32	102	0.42	30	11	72	13	0.18
SD	10.3	26.1	16.2	90.1	0.6	17.6	2.59	21.6	123	0.32
Minimum	62	16	17	53	0.11	23	8.9	63	8.7	0.02
Maximum	101	120	80	380	2.9	104	19	162	577	1.3
Range	39	104	63	327	2.79	81	10.1	99	568.3	1.28
Kurtosis	0.16	2.68	0.72	1.06	12.06	13.06	2.01	11.64	20.9	4.07
Skewness	0.84	1.7	1.19	1.39	3.16	3.39	1.48	3.16	4.5	1.97
Unpolluted soils[a]	82.4	26	22.6	72.4	0.097	26.9	12.7	61	11.2	0.065
Target value		150	100	500	5	100	50	250	30	2
Intervention value		600	500	3000	20	500	300	800	50	10
Sample size = 21	Sb	Mn	Mo	Be	Sn	Br	S(%)	Pt	Au	Pd
Mean	3.46	543.1	1.51	1.79	5.13	4.35	0.05	2.5	4.4	2.3
Median	0.96	508	1.2	1.7	4.2	3.9	0.04	2.9	3.5	2.8
SD	11.3	116	0.9	0.19	2.3	1.8	0.034	1.1	4.8	0.6
Minimum	0.79	430	0.71	1.5	2.2	2.4	0.01	1.0	0.8	1.1
Maximum	53	902	4.9	2.2	11	8.7	0.14	4.7	24	3.7
Range	52.2	472	4.19	0.7	8.8	6.3	0.13	3.7	23.2	2.6
Kurtosis	20.9	3.7	10.1	−0.49	1.2	1.39	1.57	−1.23	15	1.3
Skewness	4.58	1.84	2.89	0.49	1.3	1.34	0.96	0.11	3.6	−1.05
Unpolluted soils[a]	1.21	583	2.0	1.95	2.6	5.4				
Target value			100		50					
Intervention value			200		300					

[a] Arithmetic mean values of different natural soils of China (China Environmental Monitoring General Station 1990); Target and intervention values for Dutch legislation

of catalytic converters is now causing a considerable rise in the matrices such as soil, urban water and vegetation near areas subjected to intense vehicular traffic (Varrica et al. 2003; Helmers et al. 1998). Pt, Au and Pd concentrations in Xuzhu urban topsoils vary in the range 1.0–4.7, 0.8–24 and 1.1–3.7 ng/g, respectively. The observed values are similar to the earth's crust indicating these three have less impact on the Xuzhou urban environment.

PCA and statistical evaluation of anthropogenic pollution

To reduce the high dimensionality of the sample/variable space, a PCA was applied to the available dataset of heavy metals, Br and S. The obtained factors were rotated using a varimax normalized algorithm, which allows an easier interpretation of the principal component loadings and maximization of the variance explained by the extracted factors.

Three principal components were extracted from the available dataset and explained a total variance of approximately 70.187% (Table 3). In Table 3, the factor loadings for the three extracted factors are reported allowing an interpretation of the master variables. Factor 1 is dominated by Al, Ti, Ga, Li, V, Co, Pt, Mn and Be. The distribution of these elements is mainly controlled by natural parent materials, the first factor can be inferred as a ''lithogenic factor''. Factor 2 loaded by Ag, Se, Sc, Pb, Cu, Zn, Cd, Au, Ni, S and Mo can be identified as a tracer of anthropogenic pollution source, which is associated with traffic. The influence of traffic is characterized by Zn and to a lesser extent by Cu and Pb (Miguel et al. 1997). Zinc compounds have been employed extensively as antioxidants (e.g. zinc carboxylate complexes and zinc sulfonates) and as detergent/dispersant improves for lubricating oils (Drew 1975). Tyre wear has also contributed significantly to the zinc load in urban topsoils. Oxidation of lubricating oils upon exposure to air at high temperatures results in the formation of organic

Table 3 Values of the three extracted factor loadings for the studied heavy metals, Br and S

Elements	Factor 1	Factor 2	Factor 3
Al	**0.951**	−0.120	1.338E-02
Fe	**0.602**	**0.469**	0.379
Ti	**0.945**	−0.192	−3.23E-02
Ag	−5.05E-02	**0.628**	0.179
Se	−9.59E-02	**0.768**	0.377
Sc	−0.361	**0.812**	0.120
Ga	**0.684**	0.403	1.247E-02
Ba	**0.513**	**0.496**	0.115
Li	**0.820**	0.411	6.989E-03
Bi	−5.05E-02	0.155	**0.968**
V	**0.985**	−7.96E-02	5.67E-02
Pb	8.135E-02	**0.729**	**0.491**
Cu	0.204	**0.889**	0.230
Zn	0.148	**0.913**	6.784E-02
Cd	0.109	**0.528**	−8.27E-03
Ni	0.253	**0.607**	−0.119
Co	**0.870**	−2.62E-03	0.283
Cr	7.508E-03	0.234	**0.910**
As	−7.86E-02	1.760E-02	**0.975**
Hg	0.123	9.482E-02	**0.753**
Sb	−9.12E-02	1.760E-02	**0.974**
Mn	**0.913**	−1.05E-02	6.769E-02
Mo	6.113E-04	**0.274**	5.311E-02
Be	**0.956**	0.191	8.044E-02
Sn	**0.372**	**0.349**	−2.58E-02
Br	**0.503**	**0.671**	−0.122
S	−0.262	**0.618**	−1.30E-02
Pt	**−0.630**	0.190	0.184
Au	−0.125	**0.395**	−0.278
Pd	−0.211	3.295E-02	**−0.560**
Cumulative loading	27.639%	52.093%	70.187%

Values of dominant elements in each factor reported in bold
Extraction method: PCA, *Rotation method*: Varimax with Kaiser normalization

acids, alcohols, ketones, aldehydes and other organic compounds that are corrosive to metals. This corrosive action causes wear of those metal parts which come into contact with the oil and which in many cases consist of zinc, copper and cadmium-bearing alloys or, as in the case of sinterised materials used in the oil pump of automobiles, of nickel, copper and Mo (Miguel et al. 1997). This process results ultimately in the release of those metals to the urban environment and their accumulation in the urban topsoils.

Factor 3 loaded mainly by Bi, Cr, As, Hg and Sb can also be identified as a tracer of anthropogenic pollution source which is linked with the coal-burning or other activities. The origin of Sb is associated with the fly-ash from the coal combustion (Miguel et al. 1997) and Hg in urban topsoils is often linked with the coal-burning industry (Liu 1997).

Inter-element relationships (Table 4) can provide interesting information on element sources and pathways (Manta et al. 2002). Table 4 shows that there are significant correlations between elements of Factor 2 and Factor 3, which in turn also demonstrate the conclusions drawn from the above-mentioned theory.

Iron, Ba, Sn, Pd and Br show high values in Factor 1, but it is also represented in Factor 2. An explanation for this may be that the distributions of these elements are affected by mixed sources.

Conceptually, Factor 2 and Factor 3 condense the information of the elements as tracers of anthropogenic pollution. The calculated scores of Factor 2 and Factor 3 were plotted on the maps of the urban area of Xuzhou (Fig. 2) to gather an "average image" of the anthropogenic pollution in the city. The two maps indicate the occurrence of a diffuse pollution related to anthropogenic activity with a good correspondence of metal contaminations and sites of major deposition of atmospheric particles generated primarily from traffic. Such an urban distribution map can be used for direct comparison with thematic maps and database using GIS software.

Hierarchical clustering analysis

In order to discriminate distinct groups of elements studied as tracers of natural or anthropogenic source, an explorative hierarchical CA was performed on the available dataset. The obtained results (Fig. 3) enabled the identification of two main groups of elements, discriminating Sn, Li, Ga, Ba, Fe, Mn, Co, Be, Ti, Al and clay (group 1) from Hg, Cr, Sb, As, Bi, Pd, Pt, Au, Ni, Cd, Br, Zn, Cu, S, Pb, Se, Mo, Sc and Ag (group 2). This result is consistent with elemental relationships indicating that the metals of Group 1 strongly correlate with the clay (Table 5), thus supporting a natural origin for these elements (Wilcke et al. 1998). Group 2 includes metals dominated by an anthropogenic input. This result obtained by CA is in agreement with the results of PCA.

Table 4 Correlation matrix for elements in Factor 2 and Factor 3

Factor 2

	Pb	Zn	Cu	Cd	Ag	Se	Br
Pb	1	0.672[b]	0.740[b]	0.305	0.462[a]	0.826[b]	0.564[a]
Zn		1	0.963[b]	0.725[b]	0.477[a]	0.671[b]	0.758[b]
Cu			1	0.712[b]	0.525[b]	0.710[b]	0.721[b]
Cd				1	0.222	0.304	0.544[a]
Ag					1	0.391	0.072
Se						1	0.492
Br							1

Factor 3

	As	Cr	Sb	Hg	Bi
As	1				
Cr	0.887[b]	1			
Sb	1.00[b]	0.888[b]	1		
Hg	0.728[b]	0.590[b]	0.725[b]	1	
Bi	0.988[b]	0.915[b]	0.988[b]	0.715[b]	1

[a] Correlation is significant at the 0.05 level (1-tailed)
[b] Correlation is significant at the 0.01 level (2-tailed)

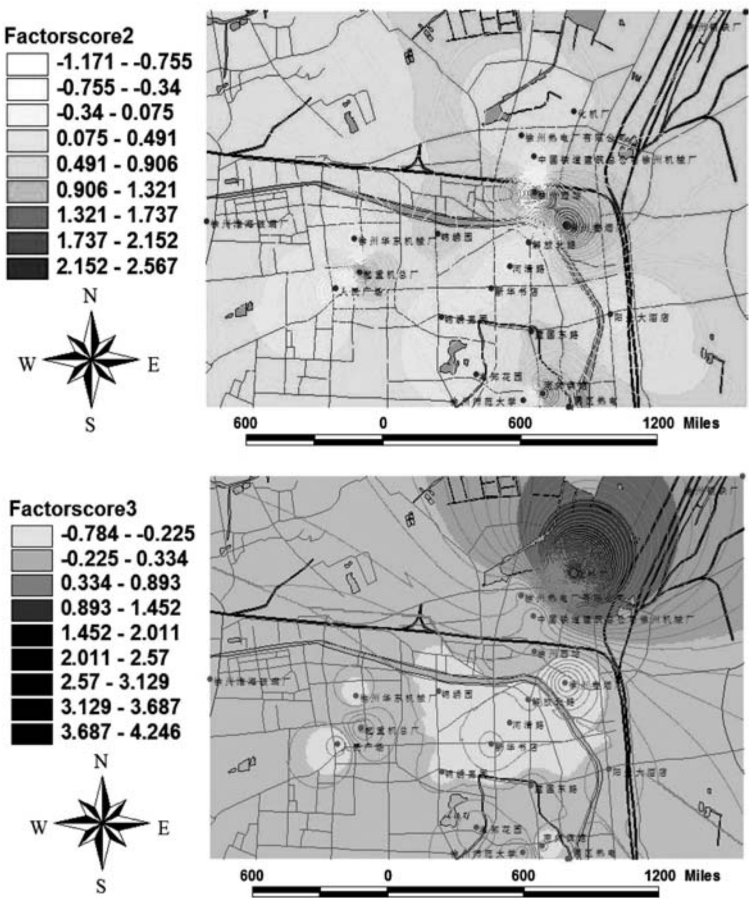

Fig. 2 Maps of the estimated "average pollution" in the city of Xuzhou (*upper panel* for Factor 2 and *lower panel* for Factor 3)

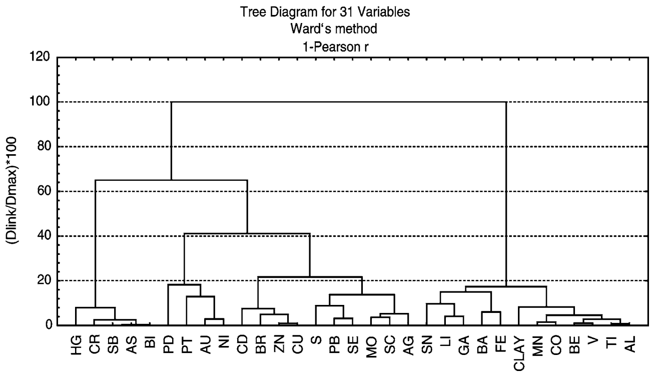

Fig. 3 Hierarchical clustering results (dendrogram) of the heavy metals, S, Br and clay concentrations in topsoil samples of the Xuzhou city

Conclusions

The sources of the different elements in urban topsoils are typically common to most urban environments (traffic, industry, natural substrate, etc.), but their distribution patterns vary according to the peculiarities of each city. In the case of the city of Xuzhou (China), the following conclusions can be drawn: (1) the variability of Al, Ti, Ga, Li, V, Co, Pt, Mn and Be is controlled by soil parent material; (2) Ag, Se, Sc, Pb, Cu, Zn, Cd, Au, Ni, S and Mo contents are controlled by a long-term anthropogenic activity connected with traffic, resulting in large-scale chemical anomalies; (3) Bi, Cr, As, Hg and Sb exhibited large-scale anomalies, spatially related to the location of anthropogenic activities such as coal-burning industry or other industries; (4) the patterns of distribution of Fe, Ba, Sn, Pd and Br are affected by mixed sources.

Table 5 Pearson correlation coefficients between metals of group 1 and clay

	Al	Fe	Ti	Ga	Ba
Clay	**0.635**[b]	0.469[a]	**0.631**[b]	**0.452**[a]	**0.563**[b]
	Li	V	Co	Mn	Be
Clay	**0.601**[b]	**0.714**[b]	**0.589**[b]	**0.756**[b]	**0.630**[b]

[a] Correlation coefficient is significant at the 0.05 level (1-tailed)

[b] Correlation coefficient is significant at the 0.01 level (2-tailed)

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