ABUNDANCE OF HEAVY METALS IN URBAN SOILS AS CONCERNS GENESIS AND POLLUTING IMPACT

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Data referring to the abundance, contamination or pollution level and content of heavy metals in labile and stable fractions of urban soils in two important municipalities of Romania – Bucharest, the country's capital and Baia Mare the most important center of extracting and processing the polymetallic sulphides are presented. Urban soils in both cities were grouped in three categories, namely, park soils, soils in areas along the streets and soils in vegetable and fruit gardens. Six heavy metals were analyzed: Cd, Cr, Cu, Ni, Pb and Zn. Urban soils in the Bucharest parks are not polluted with heavy metals. They present only a contamination degree, while the soils in the Baia Mare parks have a an average content 11 times higher than those in Bucharest, being strongly polluted with Cd, Cu and Pb. Urban soils located along the streets in Bucharest present slight (Cu, Pb, Zn) and moderate (Cd) pollution, while those located along the streets in Baia Mare are characterized by very strong (Pb), strong (Cd, Cu) and moderate (Zn) pollution. Soils along the streets in Baia Mare are almost five times more polluted than those in Bucharest. Urban soils in vegetable and fruit gardens in Bucharest are characterized by strong (Cd), moderate (Zn) and slight (Cu), while the similar soils in Baia Mare present strong (Cd, Cu, Zn) and very strong (Pb) pollution. Pollution with heavy metals of urban soils in the vegetable and fruit gardens in Baia Mare is over four times more intensive than that in soils with similar use in Bucharest.

***Keywords**: heavy metals, urban soil, genesis, pollution, impact.*

The wide extension research recently carried out in the urban soils may be explained by the complexity of their functions. These soils represent both the foundation of settlement infrastructure and the nutrition medium for plants, usually horticultural crops, grown in parks or around the houses in the areas farther the down town.

The urban soils are under a permanent impact of emissions and imissions from the industrial units, motor transport and home activity. As a result, the pollution elements and chemical substances will enter the general circuit of soil-plant system, affecting sometimes the health of people consuming the products obtained on such soils.

Among the frequently identified pollutants in the urban areas, the heavy metals have a significant place. They come, according to the circumstances, from both the extractive industry of non-ferrous ores and their processing up to obtaining the material goods. Also the heavy metals can enter the soil as a result of applying some chemical substances used as pesticides or waste water for irrigation.

Such reasons are serious motivations to study the heavy metals in urban soils of many urban settlements, which started even from the apparition of the first concerns about this category of soils as a distinct field of soil science. This issue was largely debated at the International Symposium on Urban Soils, held in Western Berlin (1981) and publication of its works (1982) regarding the soils in several cities, such as Hamburg [9], Kiel [8], and Tokyo [10]. Similar studies followed. As a result, in the last time, data on this subject were published regarding the heavy metal pollution of soils in many other urban agglomerations, like Moscow [12], Trondheim [11], Lods [3], Barcelona [1], Nanjing [13], Torino [2] etc.

The importance of knowing the contamination or pollution level of urban soils in Romania determined the authors to present the research results obtained on soils in two cities of this country, which are different from the viewpoint of natural and socio-economic conditions. These cities are Bucharest municipality, the country's capital, and Baia Mare municipality, an important center for mining and processing the non- ferrous ores.

MATERIAL AND METHOD

The Bucharest municipality is located in the south-eastern part of the country, in the Romanian Plain, at an average absolute altitude of 80 m. It covers a land area of 228 km^2 and has a population of about 2 million inhabitants. The main industrial activities are represented by energy industry, construction of machines and light industry. In addition there is a very intensive urban motor transport.

The Baia Mare municipality is located in the north-western part of the country, in the depression with the same name, at an average absolute altitude of 228 m. It covers a land area of 23.5 km^2 and has a population of about 150000 inhabitants. The mining of complex sulphides and their industrial processing represent the main industrial activities in this city.

Brown Reddish soils and Argilo-Illuvial Chernozems represent the major natural soil cover in the Bucharest area, and Albic Luvisols and Brown Luvic soils in the Baia Mare area. The soils in both areas were strongly changed by the urban infrastructure works and even by the mechanical operations. The high rates of emissions and imissions have been added to them, these being higher in Baia Mare than in Bucharest.

Topsoil (0-20 cm) samples were collected from different points of cities representing soils in parks, areas along to streets, and vegetable and fruit gardens.

Soil samples were analyzed in the laboratory on the chemical and physical properties, as well as the total content of heavy metals (Cd, Cr, Cu, Ni, Pb and Zn). The heavy metals were determined using the atomic absorption spectrometry in HCl solution obtained after soil $HClO_4$ and HNO_3 digestion by atomization in the air acetylene flame. The speciation of heavy metals in the total content was determined by applying the Lăcătuşu and Kovacsovics method [6].

The statistical processing of analytical data was carried out by computing the variation parameters ($X_{min.}$, $X_{max.}$, X, σ, CV), and the arithmetic mean-X was determined from the grouping parameters. The establishment of soil contamination/pollution level of soils was carried out according to the Lăcătuşu method [8].

RESULTS AND DISCUSSIONS

Abundance of heavy metals in the Bucharest and Baia Mare urban topsoils

Tables 1, 2 and 3 show the statistical parameters of the total content of heavy metals in the urban topsoils, grouped according to their land use. With few exceptions, their analysis reveals high and very high quantities of heavy metals in the Baia Mare urban soils as compared to the Bucharest area. Thus, contents up to 11.3 $mg.kg^{-1}$ Cd, 449 $mg.kg^{-1}$ Cu, 967 $mg.kg^{-1}$ Pb and 914 $mg.kg^{-1}$ Zn were detected in the soils of Baia Mare parks, while the maximum values of these elements in urban soils of Bucharest park areas were within the normal abundance range of these chemical elements in soils (*tab. 1*).

Table 1

Statistical parameters of heavy metals content ($mg·kg^{-1}$) in topsoil (0-20 cm) of urban soils in park areas of Bucharest (B) and Baia Mare (BM) municipalities as compared to normal values (NV), maximum allowable limits (MAL), alert threshold (AT) and triggering values (TV) for a less sensitive soil use

	Cd		Cr		Cu		Ni		Pb		Zn	
	B	BM	*B*	BM	*B*	BM	*B*	BM	*B*	BM	*B*	BM
x_{min}	0.25	1.70	11	15	8	59	11	20	5	235	36	290
x_{max}	0.63	11.31	29	40	24	449	27	31	20	967	110	914
average	0.43	4.10	18	25	14	165	21	25	11	627	59	508
σ	0.13	2.72	4	7	4	114	4	4	3	221	20	257
CV (%)	30.00	67.00	24	27	25	68	19	17	30	35	34	51
NV[†]	0.3		30		20		20		15		50	
MAL[‡]	3.0		100		100		50		100		300	
AT_{SPS}[§]	5.0		300		250		200		250		700	
TV_{SPS}[§]	10.0		600		500		500		1000		1500	

[†] from Fiedler and Rösler (1988); [‡] after Kloke (1980); [§] Order 756 of MAPPM (1997)

The highest contents of Pb and Zn up to 5385 $mg.Kg^{-1}$ Pb and 1486 $mg.Kg^{-1}$ Zn, respectively, were detected in the soils along the streets of Baia Mare, while the maximum values of the other analyzed chemical elements were at the level of those determined in soils of city's parks. Increased contents of heavy metals were detected in soils along the streets in Bucharest, the highest contents being those of Pb and Zn, but at lower levels as compared to those detected in soils with similar use in Baia Mare (*tab. 2*).

Table 2

Statistical parameters of heavy metals content (mg·kg^{-1}) in topsoil (0-20 cm) of urban soils in street areas of Bucharest (B) and Baia Mare (BM) municipalities as compared to normal values (NV), maximum allowable limits (MAL), alert threshold (AT) and triggering values (TV) for a less sensitive soil use

	Cd		Cr		Cu		Ni		Pb		Zn	
	B	BM	*B*	BM	*B*	BM	*B*	BM	*B*	BM	*B*	BM
x_{min}	1.10	0.70	19	11	28	13	19	19	39	85	84	92
x_{max}	4.60	10.90	39	36	199	426	49	42	284	5385	387	1486
average	2.20	3.90	28	23	63	210	37	29	128	1192	163	530
σ	0.70	3.10	6	5	47	179	8	6	108	1271	86	397
CV (%)	37.00	79.00	22	24	65	85	19	21	85	106	52	75
NV†	0.3		30		20		20		15		50	
MAL‡	3.0		100		100		50		100		300	
AT_{SPS}§	5.0		300		250		200		250		700	
TV_{SPS}§	10.0		600		500		500		1000		1500	

† from Fiedler and Rösler (1988); ‡ after Kloke (1980); § Order 756 of MAPPM (1997)

Finally, the highest contents of heavy metals were also detected in the vegetable and fruit gardens in Baia Mare, reaching up to 3261 mg.Kg^{-1} Pb, 2685 mg.Kg^{-1} Zn and 404 mg.Kg^{-1} Cu. As concerns the other chemical elements in the two municipalities, the contents of heavy metals are comparable. In the Cd case, the maximum and minimum values exceed the maximum allowable limits and the alert threshold for a sensitive use of soil (*tab. 3*).

Table 3

Statistical parameters of heavy metals content (mg·kg^{-1}) in topsoil (0-20 cm) of urban soils in vegetable and fruit gardens in Bucharest (B) and Baia Mare (BM) municipalities as compared to normal values (NV), maximum allowable limits (MAL), alert threshold (AT) and triggering values (TV) for a less sensitive soil use

	Cd		Cr		Cu		Ni		Pb		Zn	
	B	BM	*B*	BM	*B*	BM	*B*	BM	*B*	BM	*B*	BM
x_{min}	1.90	0.30	25	12	21	23	28	18	33	151	126	180
x_{max}	4.90	16.60	40	31	70	404	54	50	127	3261	212	2695
average	3.40	3.30	31	21	39	166	42	25	77	834	65	585
σ	1.20	3.70	6	6	18	193	9	6	39	857	31	622
CV (%)	3.40	113.00	20	29	47	116	23	30	51	103		106
NV†	0.3		30		20		20		15		50	
MAL‡	3.0		100		100		50		100		300	
AT_{SPS}§	3.0		100		100		75		50		300	
TV_{SPS}§	5.0		300		200		150		100		600	

† from Fiedler and Rösler (1988); ‡ after Kloke (1980); § Order 756 of MAPPM (1997)

Comparing the maximum and mean values with values of alert threshold and triggering for a less sensitive land use shows that, in the case of soils in parks and along the streets (*tab. 1, 2*), the threshold values are exceeded in the case of urban soils in Baia Mare. Thus, in the case of soils in parks, Cd maximum value is higher than the trigger value. In exchange, in the case of Cu, Pb and Zn, the maximum values exceed only the value of alert threshold. Unlike the soils in parks, the Pb maximum value in soils along the streets is 5.4 times higher than the trigger value, while the mean value is 1.2 times higher than the trigger value.

As concerns the soils in the vegetable and fruit gardens (*tab. 3*), Cd maximum and mean values exceed the level of alert threshold and triggering for the sensitive soils in both locations, while, in Baia Mare, maximum and mean values are exceed only in the case of Cu, Pb and Zn. There are also mentioned the Pb maximum and mean values in soils of vegetable and fruit gardens in Bucharest exceeding the level of triggering an alert threshold.

The values of enriching coefficients regarding the urban soils in Baia Mare (*tab. 4*) clearly show the high level of contamination of urban soils in Baia Mare with heavy metals, especially Cd, Cu, Pb and Zn as compared to the urban soils in Bucharest. Thus, the Pb content in the Baia Mare parks is 59 times higher than in the Bucharest parks. Likewise, increased Cd, Cu, Pb and Zn contents were detected in urban soils of Baia Mare, both in soils along the streets and in the vegetable and fruit gardens.

Table 4

Riching coefficients of urban soils in Baia mare municipality as compared to urban soils in Bucharest municipality

Area nature	Cd	Cr	Cu	Ni	Pb	Zn
Park area	9.5	1.4	12	1.2	57.0	8.6
Street area	1.8	0.8	3.3	0.8	9.3	3.2
Area of vegetable gardens	1.0	0.7	4.2	0.9	26.0	8.4

Placing in a decreasing order of the abundance value of heavy metals in urban soils from Baia Mare leads to the following succession: Pb-Zn-Cd-Cu, as compared to Bucharest (Cd-Zn-Cu-Pb). The Cr and Ni abundance in urban soils of the two municipalities is at the normal level.

Assessment regarding the heavy metals contamination/pollution level of urban soils in the two municipalities

According to Lăcătuşu (1998), the ***"soil contamination"*** term defines the content interval within which any measured value of heavy metals, bound to the texture and organic matter content, has not or will not have immediate negative effects on plant growth and development or on another environmental components, and the ***"soil pollution"*** term defines the content interval within which any measured value induces negative effects on some or all of the environmental components.

A distinction between soil contamination range and soil pollution range is established by means of ***contamination/pollution index*** (C/P I). This index represents the ratio between the heavy metal content effectively measured in soil by chemical analysis and the reference value. ***Reference value represents*** the A value obtained by calculation for each sample using the formulae of the Dutch system [8].

The mean values of contamination/loading indices (*tab. 5*) clearly separate the Cr and Ni chemical elements reflecting only a contamination of analyzed urban soils, which is a slight to moderate contamination in the Cr case and a strong to very strong contamination in Ni case, as compared to the other analyzed heavy metals which are predominant polluting elements.

In the Bucharest park areas, the soils are only contaminated with heavy metals from a slight (Cr, Pb) to moderate (Cu, Zn) up to strong (Cd, Ni) contamination. Unlikely, a strong (Cd, Cu and Pb) and moderate (Zn) pollution, and a slight (Cu) and strong (Ni) contamination were recorded in the Baia Mare parks.

By totalizing the individual values of index which signifies pollution of each analyzed chemical element (singular pollution), another index is obtained defining the ***multiple pollution***. In the case of urban soils in the Baia Mare parks, the multiple pollution index value shows an excessive pollution with heavy metals, while the urban soils in the Bucharest parks are not polluted with heavy metals. The soils along streets in the two municipalities are polluted with Cd, Cu, Pb and Zn, but the pollution level is different, namely: a slight-moderate pollution in Bucharest and a strong-very strong pollution in Baia Mare. The multiple pollution of soils in the Bucharest park areas is at a very high level, while that in Baia Mare is at an excessive level. Soils in the Baia Mare park areas are almost four times more polluted with heavy metals than those in Bucharest municipality.

In the areas of vegetable and fruit gardens, the pollution level is similar with that in the soils along the streets, in many situations. Thus, in the case of Bucharest, a Cu slight, Cr and Zn moderate, Cd strong and Ni and Pb very strong contamination were recorded. Also, a Cd, Cu and Zn strong pollution was recorded in the garden soils from Baia Mare, while the Pb pollution was very strong.

The multiple pollution of garden soils in the two localities was very strong in the Bucharest urban soils and excessive in the Baia Mare urban soils. In latter ones, the pollution was three times more intensive than in the Bucharest urban soils.

Fractionating of total content of heavy metals

Table 5

Mean values of contamination/pollution index (Ic/p) with heavy metals of urban soils in Bucharest (upper cell value) and Baia Mare (lower cell value) and the signification of these indices

Area nature	Singular contamination/pollution						Multiple pollution
	Cd	Cr	Cu	Ni	Pb	Zn	
Park area	0.62 (s.c.)	0.16 (s.c.)	0..8 (m.c.)	0.62 (s.c.)	0.13 (s.c.)	0.39 (m.c.)	-
	6.12 (s.p.)	0.24 (s.c.)	4.92 (s.p.)	0.67 (s.c.)	7.75 (s.p.)	3.71 (m.p.)	22.5 (e.p.)
Street area	3.3 (m.p.)	0.27 (m.c.)	1.85 (s.p.)	0.97 (s.c.)	1.57 (s.p.)	1.17 (s.p.)	7.92 (v.s.p.)
	6.09 (s.p.)	0.22 (s.c.)	6.38 (s.p.)	0.78 (v.s.c.)	14.9 (v.s.p.)	3.91 (m.p.)	31.28 (e.p.)
Area of vegetable and fruit gardens	4.62 (s.p.)	0.31 (m.c.)	1.15 (s.p.)	0.90 (v.s.c.)	0.82 (v.s.c.)	2.07 (m.p.)	7.84 (v.s.p.)
	4.85 (s.p.)	0.19 (s.c.)	4.75 (s.p.)	0.63 (s.c.)	10.02 (v.s.p.)	4.06 (s.p.)	23.68 (e.p.)

s.c. - slight contamiantion; m.c. - moderate contamiantion; s.c. – strong contmaiantion; v.s.c. – very strong contamination; s.p. – slight pollution; m.p. – moderate pollution; s.p. – strong pollution; v.s.p. – very strong pollution; e.p. – excessive pollution.

Determination of fractions of heavy metals bound to the soil components and the mean percentage distribution of these fractions revealed differences between soils in the two locations regarding both the percentage value to the mean value of total content. Thus, the percentage value in the Bucharest urban soils increases from labile fractions (fractions soluble in soil solution, exchangeable and bound to organic matter) to stable fractions (bound to Fe and Mn oxides, and to crystalline net of minerals or residual fraction) is observed.

On the contrary, in the case of Baia Mare urban soils, the highest percentage values were recorded in the category of labile fractions (*tab. 6*).

Table 6

Mean percentage distribution of heavy metals ($mg \cdot kg^{-1}$) in urban soils in Bucharest (upper cell) and Baia Mare (lower cell) as against the absolute values of mean contents of heavy metals in the two locations and three areas

Fraction nature	Areas		
	Parks	Streets	Gardens
Soluble in soil solution	8	14	17
	18	21	19
Exchangeable	15	19	24
	19	18	21
Bound to organic matter	16	13	18
	12	14	16
Bound to Mn and Fe oxides	23	29	21
	27	29	28
Residual	38	25	20
	24	18	16
$mg \cdot kg^{-1}$	20	70	67
	226	331	273

The phenomenon is caused by both the differences between the initial nature of natural soils and the present reaction of these soils. Thus, if the initial reaction of the Bucharest urban soils oscillates in a neutral – slightly alkaline range, the initial reaction of the Baia Mare urban soils frequently oscillates in an acid range, from strongly to slightly acid. The acidity of soils in Baia Mare is due both the genetic dowry and the acid rain impact. The acid reaction favors the mobility of these chemical elements, as well as others, being preferably accumulated in labile fraction. On the other hand, if the reference regards the mean total content of the six heavy metals studied in the urban soils of the two localities, it can be observed that, globally, the mean content of heavy metals in the Baia Mare urban soils is 11.3 times higher in parks and 4.7 times higher in vegetable and fruit gardens as compared to the soils with similar use in the Bucharest municipality. Therefore, the percentage value considerably differs from one location to the other. Thus, 13 per cent of the total content of heavy metals, as it is the organic material-bound fraction in urban soils along the streets in Bucharest, represent 9.1 $mg.Kg^{-1}$, while 14 per cent of the total content of heavy metals, as it is the organic material-bound fraction in urban soils along the streets in Baia Mare, represent over 5 times more, namely, 46.3 $mg.Kg^{1}$.

The calculation regarding the total content of heavy metals in the labile fraction of soils in the Bucharest park areas shows that this represents 7.8 $mg.Kg^{-1}$, while in the soils from the Baia Mare park areas, the labile fraction means 111 $mg.Kg^{-1}$. Therefore, the content of heavy metals in labile fraction of the Baia Mare soils is 14 times higher as compared to the soils in Bucharest. Likewise, the values in the soils along the streets areas will be 32 $mg.Kg^{-1}$ and 175 $mg.Kg^{-1}$, respectively, that is over 5 times higher in the soils along the streets in Baia Mare as compared to soils with similar use in Bucharest. The values in soils from the vegetable and fruit gardens particular to the two localities, are 39 $mg.Kg^{-1}$ and 153 $mg.Kg^{-1}$, respectively, resulting a content of heavy metals four times higher in the vegetable and fruit gardens from the Baia Mare municipality as compared to the Bucharest municipality.

The high quantities of heavy metals in the labile fraction, especially in the horticultural soils from the Baia Mare municipality, offer a high mobility to these chemical elements under the conditions of an acid reaction. As a result, the plants will take up high quantities of such chemical elements, some of them having toxicity characteristics. Thus, negative effects may occur for the consumers' health.

CONCLUSIONS

The natural soil cover of the two urban areas is different, namely, there are Brown Reddish soils and Argilo-Illuvial Chernozems in the Bucharest area, and Albic Luvisols with Luvic Brown soils in the Baia Mare area.

Major sources of pollution are the energy industry, industry for machine construction and motor transport in Bucharest, and industry for extraction of non-ferrous ores and their industrial processing in Baia Mare.

Urban soils in the Bucharest areas are not polluted with heavy metals, while the content of heavy metals in the Baia Mare park areas exceeds 11 times the mean content of the heavy metals in the Bucharest park areas, being characterized by a strong (Cd, Cu, Pb), and moderate (Zn) pollution.

Urban soils along the streets in Bucharest are characterized by a slight (Cu, Pb, Zn), and moderate (Cd) pollution, while those along the streets in Baia Mare are characterized by a very strong (Pb), strong (Cd, Cu) and moderate (Zn) pollution. Soils along the streets in Baia Mare are almost five times more polluted than those in Bucharest.

Urban soils in the Bucharest vegetable and fruit gardens are characterized by a strong (Cd), moderate (Zn), and slight (Cu) pollution, while soils with similar use in Baia Mare are characterized by a strong (Cd, Cu, Zn) and very strong (Pb) pollution. The pollution with heavy metals of urban soils in the Baia Mare vegetable and fruit gardens is over four times more than that of soils with similar use in Bucharest.

The mean content of heavy metals in the labile fraction of urban soils in Baia Mare is almost eight times higher than that of soils with similar use in the Bucharest urban soils.

The Cr and Ni contents in urban soils of the two municipalities show only a slight to very strong contamination.

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