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Heavy metal contents of urban soils in parks of Iaşi, Romania

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Abstract

The determination of the heavy metal content of the soils of parks in major urban areas has become vital for the estimation of potential threats to public health. In order to assess the quality of the soils in the Copou and Expoziţiei parks of Iaşi, as well as to determine the likely sources of the heavy metals, 85 samples were collected, and their Fe, Mn, Cr, Co, Ni, Cu, Zn, Pb and As contents were measured. The statistical parameters indicate a great variability of the heavy metal contents of the soils of Copou Park, compared to those of Expoziţiei Park. Multivariate analysis has highlighted the presence of 3 factors describing heavy metal variability which account for nearly 86% of the total variance. The Igeo and EF values, which can be correlated with the location and age of the parks, suggest the presence of an anthropogenic source for Cr, Cu, Zn, Pb and As.

Keywords: pollution, urban parks, heavy metals, Iaşi, Romania.

1. Introduction

The modification of the natural geochemical background of heavy metals in the biosphere due to anthropogenic activity is one of the most serious issues that humanity is facing at the moment. Within urban ecosystems, the pronounced increase in population, estimated to reach 6.3 billion people by 2050 (United Nations, 2012), will undoubtedly lead to an intensification in the action of anthropogenic factors, whose cumulated influence has a negative effect upon the health of urban inhabitants. The role played by public parks and gardens becomes essential under such circumstances, given the multiple positive functions that they fulfill in relation to urban society, among which we could stress the environmental, economic and social functions (Chiesura, 2004). The improvement of public health (Groenewegen et al., 2006; European Environment Agency, 2010) through the reduction in the level of stress experienced (Hartig et al., 2003; Fuller et al., 2007; Abkar et al., 2010), the lowering of the risk of cardiovascular diseases (Sundquist et al., 1999; Mitchell and Popham, 2008; Maas et al., 2009), the decrease in obesity rates (Ellaway

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et al., 2005), greater social cohesion (Dunett et al., 2002; Maas et al., 2006) due to the facilitation of human interaction (Coley et al., 1997), the preservation of biodiversity and of the cultural heritage, the decrease in pollution levels through the absorption of carbon-dioxide, the moderation of the urban micro-climate, as well as the increase in the price of land adjacent to parks and public gardens (Dunett et al., 2002) are just a few of the positive effects of the presence of parks and public gardens in large urban areas. Nevertheless, the direct or indirect contact between humans and the soil of the parks in large urban areas may negatively influence human health, since anthropogenic activities can have a detrimental effect upon the soils of urban parks (Kelly and Thornton, 1996; Li et al., 2001; Hursthouse et al., 2004; Lăcătuşu et al., 2004; Chenet al., 2005; Lee et al., 2006; Onder et al., 2007; Qingjie et al., 2008; Marjanović et al., 2009; Tume et al., 2008; Fiera, 2009; Kochubovsky, 2010; Prazeres et al., 2010; Jien et al., 2011) through the generation of heavy metals which can be inhaled, ingested or, rarely, absorbed by human skin (Diaz, 2006).

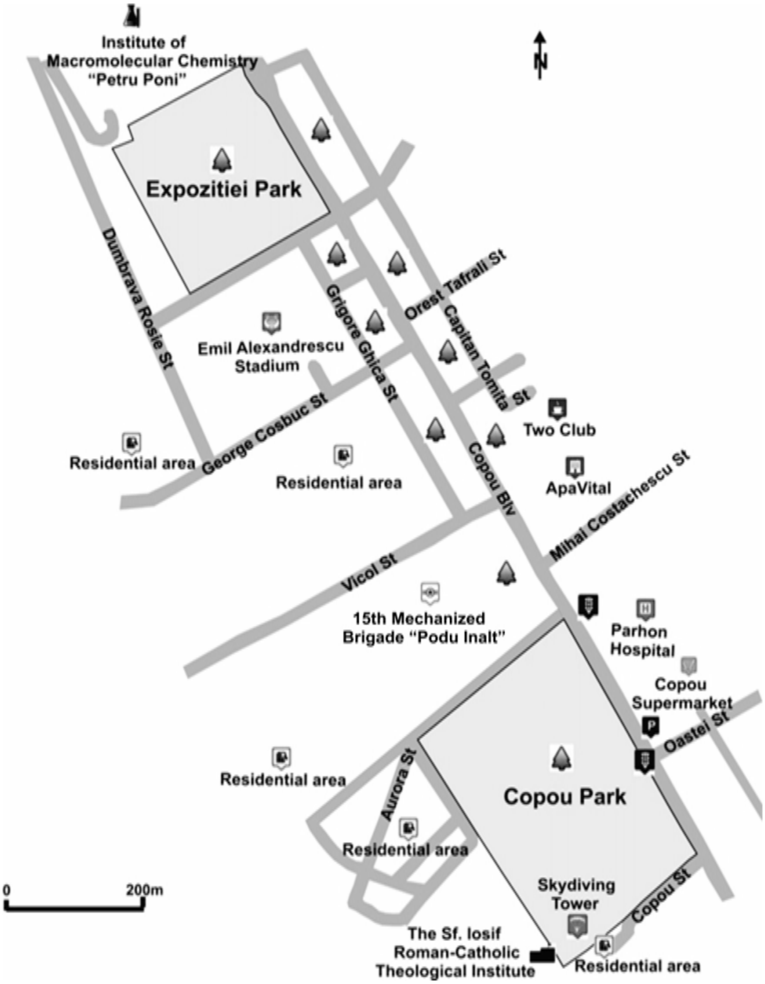

Fig. 1 Location map of study area.

2. Materials and methods

2.1. Study area

The city of Iaşi, mentioned for the first time in a document in 1408, is the main urban centre of NE Romania. With its 313,994 inhabitants, Iaşi is the second largest city in Romania. From a geological viewpoint, the city of Iaşi is located in the central-eastern region of the Moldavian Platform. The latter is comprised of a basement containing plagioclase paragneisses and leucocratic gneisses, as intercepted by a borehole extending 1,121 m into the ground, and a nappe made up of Upper Vendian – Meotian sedimentary deposits, including the Bessarabian clays at the Cryptomactra outcrop (Ionesi, 1994). According to the Agency for Environmental Protection (2011), despite the great density of its population, the city of Iaşi does not offer more than 5.91 m^2 of public park, garden or square per inhabitant. According to Law 24/2007, the Copou and Expoziţiei parks are public. Devised between 1753 and 1923, they are located on Copou Hill and cover surface areas of 10 ha and 7.6 ha, respectively (Fig. 1).

The soils in the Copou (CS) and Expoziţiei (ES) parks are cambic chernozems (Lăcătuşu et al., 2005).Previous research on heavy metal pollution in the soils of Iaşi has been conducted by Lăcătuşu et al. (2005), Lăcătuşu et al. (2008), Iancu and Buzgar (2008), and Lăcătuşu and Lăcătuşu (2009).

2.2. Soil sampling

The samples were collected between April 1st and May 15th 2012 so as to avoid the impact of the urban microclimate (extreme temperatures, heavy rain or snowfall) upon the representativity of the heavy metal contents of the soils. A total of 85 soil samples (CS – 49 samples; ES – 36 samples) were collected at the nodes of a virtual square grid having a side with a N60ºE orientation, perpendicular to the Carol I Boulevard. The size of the basic sampling grid cell was 50×50 m, the sampling depth interval was between 0.0 m and 0.3 m, while the mass of the samples ranged between 1.5 and 2.0 kg. The samples were collected manually, using a stainless steel spade shovel, and then placed in plastic bags.

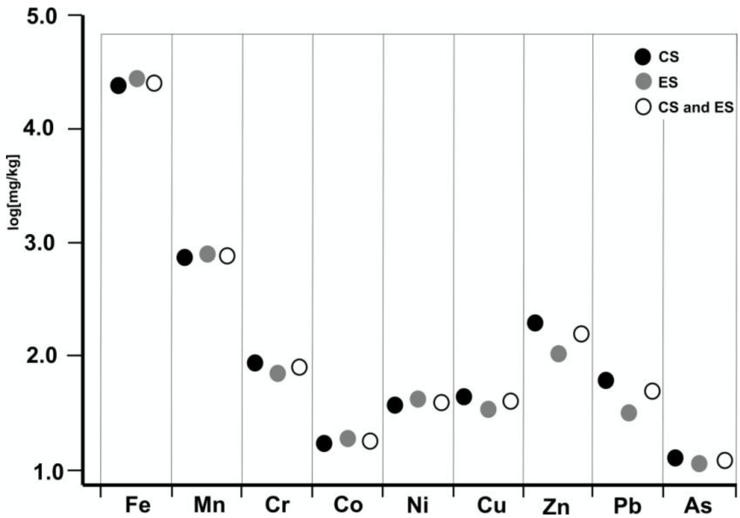

Fig. 2 Mean values of the heavy metal contents.

Tab. 1 Descriptive statistics parameters of soil heavy metals

Element	Range (mg/kg)	Mean (mg/kg)	Geom. Mean	Median (mg/kg)	Standard Deviation	Coef. of Variation
CS and ES (N = 85)						
Fe	18604.5÷36719.4	30453.4	30163.7	31473.7	3909.2	0.128
Mn	557.6÷944.8	831.6	826.9	851.8	83.7	0.101
Cr	50.3÷184.3	87.2	84.8	81.9	22.4	0.258
Co	9.6÷24.9	19.4	19.1	20.3	3.3	0.171
Ni	22.2÷53.4	42.9	42.3	45.0	6.8	0.159
Cu	27.3÷135.1	43.3	40.2	35.8	21.6	0.500
Zn	75.8÷3526.0	170.1	127.6	118.0	372.2	2.188
Pb	20.7÷897.3	53.1	40.3	35.4	96.4	1.815
As	9.9÷54.0	13.2	12.8	12.6	4.6	0.353
CS (N = 49)						
Fe	18604.5÷36719.4	29431.2	29079.7	30634.4	4287.9	0.146
Mn	557.6÷921.5	810.1	804.4	836.4	91.9	0.113
Cr	60.8÷184.3	94.5	91.6	86.8	25.7	0.272
Co	9.6÷24.9	18.6	18.2	19.9	3.6	0.196
Ni	22.2÷51.9	41.1	40.3	43.6	7.4	0.182
Cu	28.3÷135.1	47.8	43.0	35.4	27.0	0.567
Zn	75.8÷3526.0	211.7	143.5	123.5	487.1	2.300
Pb	20.7÷897.3	66.7	46.7	40.5	125.4	1.879
As	9.9÷54.0	13.7	13.2	12.8	6.1	0.441
ES (N = 36)						
Fe	21192.3÷35460.4	31844.8	31704.2	32243.1	2829.5	0.089
Mn	635.0÷944.8	860.9	858.6	867.3	60.8	0.071
Cr	50.3÷106.7	77.2	76.4	76.4	11.4	0.148
Co	12.1÷23.7	20.5	20.3	20.9	2.4	0.119
Ni	29.7÷53.4	45.4	45.2	46.4	4.8	0.107
Cu	27.3÷61.4	37.3	36.7	35.9	7.3	0.197
Zn	77.6÷220.4	113.3	108.8	99.1	36.1	0.319
Pb	21.6÷79.1	34.6	33.1	31.8	11.8	0.343
As	10.1÷13.7	12.4	12.4	12.5	0.7	0.061
Artificial areas for leisure: parks (N = 4)[1]						
Fe	19848.8÷22961.2	20983.3	20951.0			
Mn	570.0÷705.7	629.6	626.8			
Cr	38.3÷67.3	51.6	50.5			
Co	6.9÷9.0	8.0	8.1	[2]	[2]	[2]
Ni	33.4÷37.9	34.6	34.6			
Cu	26.6÷161.3	69.2	52.4			
Zn	64.2÷86.1	70.1	69.5			
Pb	9.6÷33.9	19.2	17.0			

[1] contents selected from the results of the analyses carried out on samples collected from the soils of the city of Iași, by Iancu and Buzgar (2008); [2] number of analyses too small for the values of the parameters to be significant

2.3. Analytical procedure

In order to determine the Fe, Mn, Cr, Co, Ni, Cu, Zn, Pb and As contents of the overall quantity of sampled soil, an EDXRF spectrometer (Epsilon5 PANalytical) was used. The samples were oven dried at a regulated temperature of 50ºC for 48 hours, then mixed with a binder at a ratio of 5:1 and homogenized for 20 minutes in an agate mortar using a mechanical mill. Approximately 15 g of the mixture were pressed into aluminium pellets with a diameter of 40 mm, using a hydraulic press operating at a pressure of 20 tons. The standardization was performed by using 23 Certified Reference Materials (SO1-4, RT, RTH, GSD and LKSD). The exposure time was 50 seconds, with the exception of As, for which the exposure interval was 90 seconds. Thus, the total concentration represents the mean value of both counts. The accuracy of the results was checked with the help of some of the CRMs.

3. Statistical analysis

The main statistical parameters of the heavy metal contents of CS and ES were determined using SPSS 19 software.

3.1.Mean concentration

The order of mean concentrations of the heavy metals present in the soils from the two studied parks (Tab. 1; Fig. 2) allows the following observations:

a) CS: Fe>Mn>Zn>Cr>Pb>Cu>Ni>Co>As;
b) ES: Fe>Mn>Zn>Cr>Ni>Cu>Pb>Co>As;
c) CS displays higher mean concentration values for Cr, Cu, Zn, Pb and As compared to ES;
d) ES displays higher mean concentration values for Fe, Mn, Co and Ni compared to CS.

The hypothetical mean section obtained by calculating the mean heavy metal values of CS and ES for the alignments parallel to Copou Boulevard and projecting the mean on a line perpendicular to the boulevard (Figs. 3 and 4) have revealed a series of interesting aspects. Thus, CS displays the highest Fe, Mn, Cr, Co, Ni, Cu, Pb and As values at a distance of 5 m from Copou Boulevard. Zn represents a particular case, since its maximum value is displayed at a distance of 255 m from Copou Boulevard. This is due to the presence of an exceptional sample (3526 mg/kg), which, once eliminated, decreases the distance at which the maxim value of Zn is recorded to 5 m from Copou Boulevard. Throughout the mean section, the lowest heavy metal values are recorded at varying distances from Copou Boulevard: 155 m (Fe, Cr, Co, Ni), 255 m (Mn, Cu, Zn), and 305 m (Pb, As). In the case of ES, the highest values are displayed at distances of 5 m (Fe, Mn, Co, Ni, Cu, Zn, Pb), 55 m (As) and 255 m (Cr) from Copou Boulevard, while the lowest values were determined at distances of 5 m (As), 55 m (Cr), 205 m (Co, Ni) and 255 m (Fe, Mn, Cu, Zn, Pb). By comparing the mean heavy metal contents of CS and ES to those of other parks in Romania and across the Globe (Tab. 2), one notices similitudes between them and those of parks in Turin (Fe – CS; Mn – CS and ES; Zn – CS), Sevilla (Zn – ES), Glasgow (Ni – CS and ES; Cr – ES), Belgrade (Co – CS and ES; Cu – CS and ES), Beijing (Pb – CS), Taipei (Pb – ES), and Yilan (As – CS and ES).

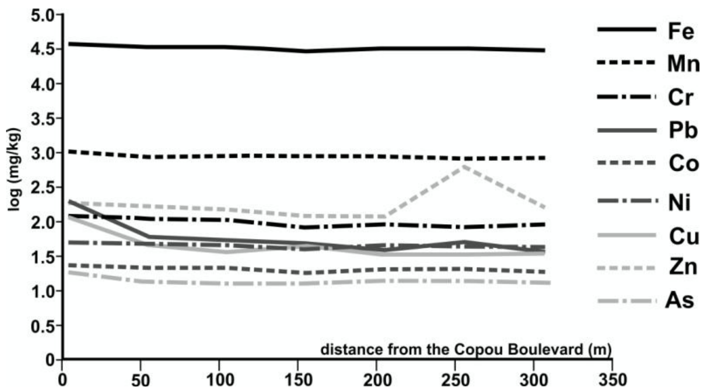

Fig. 3 Copou Park – the variation of the heavy metal content mean values as a function of the distance from Copou Boulevard.

Tab. 2 The mean heavy metal contents of soils found in various parks in Romania and across the Globe versus those of CS and ES (mg/kg)

Fe	Mn	Cr	Co	Ni	Cu	Zn	Pb	As	Location
-	-	-	-	-	24.8	168	93.4	-	Hong Kong, China[1]
20700	323	40	-	22	73	138	150	-	Sevilla, Spain[2]
32800	947	229	-	193	111	242	158	-	Torino, Italy[3]
35300	-	93	-	58	140	364	971	-	Glasgow, England[4]
11125	182	18.3	9.5	20.5	14.3	59.2	11.2	-	Bucureşti, Romania[5]
-	-		-	22.2	71.2	87.6	66.2	-	Beijing, China[6]
-	-	18	-	28	-	91.7	30	-	Talcahuano, Chile[7]
-	417.6		16.5	-	46.3	174.2	298.6	-	Belgrade, Serbia[8]
-	-	27.1	-	28.7	20.8	92.7	25	12.8	Yilan, Taiwan[9]
-	-	27.5	-	23.4	17.5	90	37	5.26	Taipei, Taiwan[10]
-	-	14.2	-	17.6	91.1	126	21.8	6.56	Kaohsiung, Taiwan[11]
20983.3	629.6	51.6	8.0	34.7	69.2	70.1	19.3	-	Iaşi, Romania[12]
29431.2	810.1	94.5	18.6	41.1	47.8	211.7	66.8	13.8	CS
31844.9	860.9	77.3	20.6	45.5	37.3	113.4	34.6	12.4	ES

[1] Li et al. (2001); [2, 3, 4] Hursthouse et al. (2004); [5] Lăcătuşu et al. (2004); [6] Chen et al. (2005); [7] Tume et al. (2008); [8] Marjanović et al. (2009); [9, 10, 11] Jien et al. (2011); [12] the mean contents obtained through the selection of 4 samples collected from soils in the city of Iași and discussed by Iancu and Buzgar (2008)

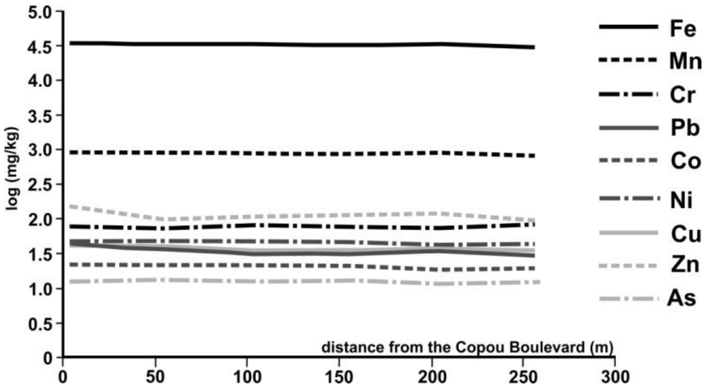

Fig. 4 Expoziției Park – the variation of the heavy metal content mean values as a function of the distance from Copou Boulevard.

3.2. Coefficients of variation (CV)

The CV calculated for the heavy metals of CS and ES (Tab. 1) are within the [0.101÷2.188] range, their order of concentration being the following: Mn < Fe < Ni < Co < Cr < As < Cu < Pb < Zn. It is worth noting that the coefficients of variation calculated for the heavy metal contents of ES display markedly lower values than those calculated for CS. According to Yongming et al. (2006), the order of magnitude of CV values may point to an anthropogenic contribution to the heavy metal content of the soils studied. Thus, based on this criterion, CS may have undergone a significant input of Pb and Zn (CV > 1), while, in the case of Mn, Fe, Ni, Co, Cr (CS and ES) and As, Cu, Zn, Pb (ES), such an input is either absent or greatly reduced (CV < 0.4). Moreover, CS seems to have been subjected to low levels of anthropogenic stress, since the CV values of As and Cu are within the (0.4÷1) range.

3.3. Correlation coefficients

The Pearson (r) correlation coefficients of the heavy metals present in the soils of CS and ES (Tab. 3) indicate a significant positive association between Fe-Mn-Co-Ni both in the case of CS and in that of ES, which suggests a common source for these elements. If one considers Mn the conservative lithogenic reference element, as described by Loska et al. (1997), one could assume that the Fe, Mn, Co and Ni contents represent the natural geochemical background of the studied soils. The strong positive correlations among Cu, Zn, Pb and As of CS, coupled with their antagonistic behaviour towards Mn, suggest, apart from a common source, an anthropogenic input to the natural geochemical background. As far as ES is concerned, the significant positive correlations between Cu-Zn-Pb, Zn-Pb-As and Pb-As point to an anthropogenic contribution to the natural geochemical background.

3.4. Multivariate analysis

3.4.1. Principal component analysis (PCA)

The application of PCA to group chemical elements according to their statistical behaviour has revealed that the variability of heavy metal contents of the soils in CS and ES can in fact be reduced to three components, which account for 85.7% of the total variance (Tab. 4). The communalities (Jackson, 2003) between the heavy metal contents vary between 0.545 (Zn) and 0.970 (Fe). The initial component matrix (Tab. 4) indicates the association of Fe, Mn, Co and Ni within the F1 component. The elevated values of As set it apart within the F2 component, while the F3 component is characterized by high Cr values.

Tab. 3 Pearson's correlation matrix for the metal concentrations

	Fe	Mn	Cr	Co	Ni	Cu	Zn	Pb	As
CS and ES (N = 85)									
Fe	1								
Mn	0.905**	1							
Cr	-0.151	-0.209	1						
Co	0.993**	0.907**	-0.152	1					
Ni	0.968**	0.891**	-0.147	0.969**	1				
Cu	-0.597**	-0.548**	0.328**	-0.605**	-0.666**	1			
Zn	-0.568**	-0.410**	0.061	-0.549**	-0.533**	0.276*	1		
Pb	-0.698**	-0.603**	0.179	-0.700**	-0.705**	0.480**	0.668**	1	
As	-0.256*	-0.229*	0.044	-0.277*	-0.300**	0.103	0.327**	0.775**	1
CS (N = 49)									
Fe	1								
Mn	0.939**	1							
Cr	-0.113	-0.181	1						
Co	0.995**	0.942**	-0.119	1					
Ni	0.968**	0.928**	-0.109	0.971**	1				
Cu	-0.639**	-0.639**	0.306*	-0.643**	-0.715**	1			
Zn	-0.599**	-0.474**	-0.118	-0.584**	-0.558**	0.184	1		
Pb	-0.705**	-0.668**	0.042	-0.709**	-0.707**	0.416**	0.609**	1	
As	-0.291*	-0.299*	-0.039	-0.317*	-0.332*	0.045	0.273	0.811**	1
ES (N = 36)									
Fe	1								
Mn	0.737**	1							
Cr	0.224	0.161	1						
Co	0.985**	0.736**	0.208	1					
Ni	0.953**	0.695**	0.269	0.947**	1				
Cu	-0.226	0.107	0.078	-0.259	-0.250	1			
Zn	-0.213	0.178	0.203	-0.175	-0.148	0.508**	1		
Pb	-0.500**	-0.059	0.137	-0.498**	-0.505**	0.609**	0.797**	1	
As	0.339*	0.688**	0.043	0.342*	0.290	0.325	0.556**	0.427**	1

** correlation is significant at the 0.01 level (2-tailed)
* correlation is significant at the 0.05 level (2-tailed)

Tab. 4 Total variance explained and component matrices for the heavy metals

Total Variance Explained

Component	Initial Eigenvalues			Extraction Sums of Squared Loadings			Rotation Sums of Squared Loadings		
	Total	% of Variance	Cumulative %	Total	% of Variance	Cumulative %	Total	% of Variance	Cumulative %
1	5.386	59.841	59.841	5.386	59.841	59.841	4.438	49.313	49.313
2	1.310	14.559	74.401	1.310	14.559	74.401	2.113	23.480	72.793
3	1.017	11.304	85.705	1.017	11.304	85.705	1.162	12.911	85.705
4	0.608	6.753	92.458						
5	0.483	5.365	97.823						
6	0.105	1.165	98.988						
7	0.057	0.638	99.626						
8	0.028	0.311	99.937						
9	0.006	0.063	100.000						

Component Matrix

Element	Component matrix			Rotated Component Matrix		
	1	2	3	1	2	3
Fe	0.956	0.155	0.178	0.951	-0.253	-0.040
Mn	0.886	0.234	0.119	0.902	-0.168	-0.110
Co	0.958	0.150	0.168	0.947	-0.260	-0.047
Ni	0.959	0.148	0.144	0.941	-0.268	-0.070
Cr	-0.239	-0.315	0.874	-0.077	0.043	0.955
Cu	-0.680	-0.366	0.233	-0.669	0.045	0.449
Zn	-0.651	0.340	-0.081	-0.467	0.565	-0.086
Pb	-0.843	0.469	0.169	-0.515	0.822	0.133
As	-0.444	0.786	0.263	-0.017	0.939	0.049

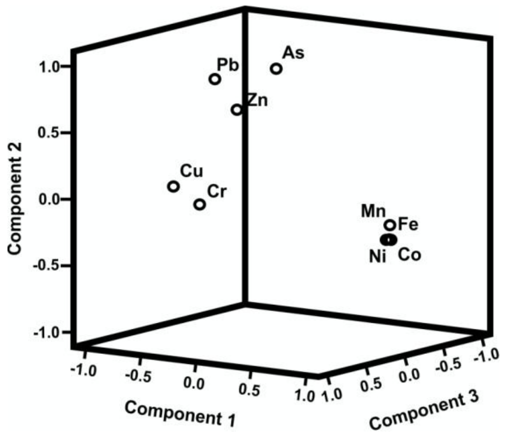

Fig. 5 The component plot in rotated space for the heavy metals.

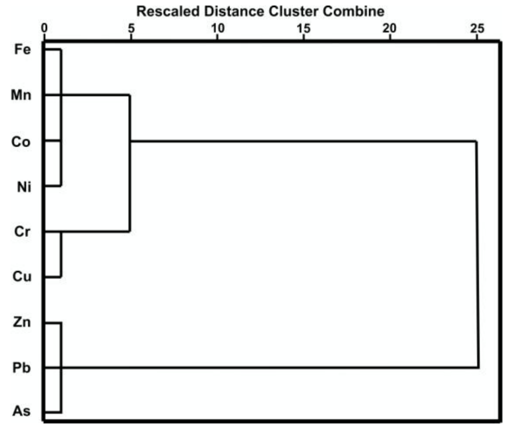

Fig. 6 Dendrogram of HCA of heavy metal concentrations in urban park soils (CS and ES).

The rotated component matrix (Tab. 4; Fig. 5) indicates high positive factor loadings of Fe, Mn, Co and Ni, and moderately negative loadings of Cu, Zn and Pb. The similar values of the loadings of Fe (0.951), Co (0.947), Ni(0.941) and Mn (0.902) suggest a common source which, in our opinion, is represented by the parent materials of CS and ES. F2 is dominated by As (0.939), Pb (0.822) and Zn (0.565). In this case, the Zn loading, which is reduced compared to the other elements of F2, suggests a source that is quasi-independent in relation to the other elements of the group. F3 is dominated by Cr (0.955) and Cu (0.449), the major difference between the loadings of the two chemical elements likely indicating different origins. If one considers Mn the conservative lithogenic reference element, as described by Loska et al. (1997), and admits that metal precipitation in soil is greatly influenced by the presence of Mn oxides and hydroxides (Kabata-Pendias and Pendias, 2001; Kabata-Pendias and Mukherjee, 2007), one notices that the distance between Mn and the other heavy metals varies largely (Fig. 6). Therefore, unlike Fe, Mn, Co and Ni, the elements As, Pb, Zn, Cr and Cu have different sources.

3.4.2. Hierarchical cluster analysis (HCA)

Based on HCA, which was performed using the Ward method and Euclidian distances as criteria for cluster formation, the chemical elements (CS and ES) were divided into the following three clusters: a) Fe-Mn-Co-Ni; b) Cr-Cu; c) Zn-Pb-As (Fig. 6), thereby confirming the grouping identified by means of PCA.

4. The distribution of heavy metals in relation with NVS, SAT and SIT

The distribution of heavy metals in soils, in relation with the normal values in soils (NVS), sensible alert thresholds (SAT) and sensible intervention thresholds (SIT) (Law 756/1997), and the average concentration of Fe in the upper crust (Reimann et al., 2008), respectively, are illustrated in Table 5 and Figure 7. For most of the chemical elements, the NVS are exceeded both in the case of CS and ES. Moreover, CS is characterized by heavy metal values which exceed both SAT (Cr, Cu, Zn, Pb, As) and SIT (Zn, Pb, As), while in the case of ES contents within the [SAT÷SIT) range have been recorded only for Pb. Based on the distance from Copou Boulevard, as projected onto a hypothetical mean section (Figs. 8 and 9), the distribution of the mean content values, as compared to NVS (or the average concentration in the upper crust in the case of Fe), indicate a much greater variance range for CS than for ES. If Fe and Mn display ratio values roughly equal to 1 for both CS and ES, the other studied elements exhibit high values in the sampling points situated in the immediate vicinity of Carol I Boulevard: CS and ES – Co, Ni, Cu, Pb; CS – Cr; ES – Zn.

Tab. 5 The contents of the heavy metals reported to NVS, SAT and SIT

Element	Park	[0÷NVS) (%)	[NVS÷SAT) (%)	[SAT÷SIT) (%)	≥SIT (%)	NVS[1]/SAT[1]/SIT[1] (mg/kg)
	CS and ES	38.8	61.2	0	0	
Fe	CS	31.8	25.9	0	0	30890[2]
	ES	7.0	35.3	0	0	
	CS and ES	84.7	15.3	0	0	
Mn	CS	54.1	3.5	0	0	900/1500/2500
	ES	30.6	11.8	0	0	
	CS and ES	0	81.2	18.8	0	
Cr	CS	0	40.0	17.6	0	30/100/300
	ES	0	41.2	1.2	0	
	CS and ES	12.9	87.0	0	0	
Co	CS	10.5	47.0	0	0	15/30/50
	ES	2.3	40.0	0	0	
	CS and ES	0	100	0	0	
Ni	CS	0	57.6	0	0	20/75/150
	ES	0	424	0	0	
	CS and ES	0	94.1	5.9	0	
Cu	CS	0	51.8	5.9	0	20/100/200
	ES	0	42.3	0	0	
	CS and ES	28.235	69.4	1.2	1.2	
Zn	CS	5.882	49.4	1.2	1.2	100/300/600
	ES	22.353	20.0	0	0	
	CS and ES	0	80.0	16.5	3.5	
Pb	CS	0	41.2	12.9	3.5	20/50/100
	ES	0	38.8	3.5	0	
	CS and ES	0	96.5	2.3	1.2	
As	CS	0	54.1	2.3	1.2	2/15/25
	ES	0	42.4	0	0	

[1] Law 756/1997
[2] average concentration of Fe in the upper crust (Reimann et al., 2008)

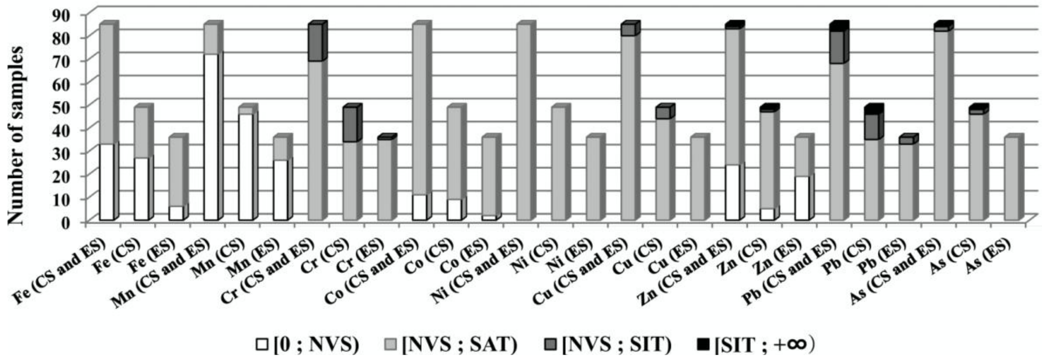

Fig. 7 The contents of the heavy metals reported to NVS, SAT and SIT (the content of Fe reported to average concentration of Fe in the upper crust, from Reimann et al., 2008)

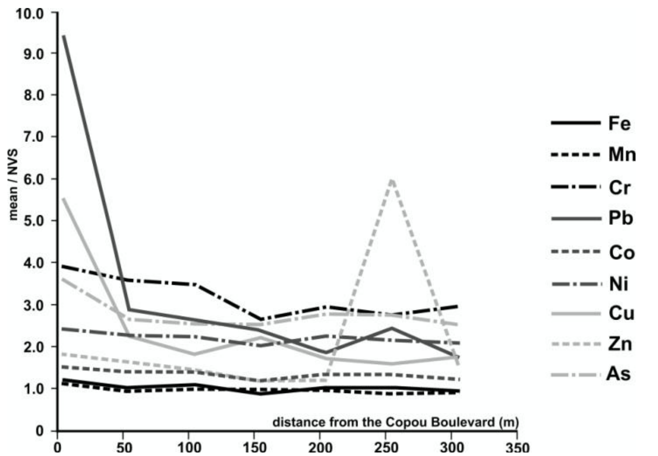

Fig. 8 Copou Park – mean content/NVS ratios as a function of distance from Copou Boulevard.

5. Assessment of metal contamination

5.1. The geochemical background (GB)

Certain methods used in the assessment of heavy metal pollution levels require the tracing of the GB. For this purpose, the background values determined by Apostoae and Iancu (2009) for Cr, Co, Ni, Cu, Zn and Pb (Tab. 6) were used. In the case of Fe, Mn and As (Tab. 6), the GB was determined through the method suggested by Reimann et al. (2005, 2008), after having previously tested and eliminated the outliers by means of the methods proposed by Wellmer (1998), Jackson and Chen (2004), and Sinclair and Blackwell (2006). Without going into details, it must be stated that the GB is higher than the NVS (or the average concentration of Fe in the upper crust) for Fe, Mn, As, Cr, Ni, Cu, Zn and Pb, but lower for Co.

5.2. The geo-accumulation index (Igeo)

In order to estimate the potential input of heavy metals into the soils of the Copou and

Expoziţiei parks, the method suggested by Müller (1969) was used:

$$I_{geo} = \log_2 [c_i \times (1.5 \times GB_i)^{-1}] \quad (1)$$

where: C_i is the measured concentration of heavy metal in soil; GB_i is the geochemical background value; and 1.5 is the background correction factor owing to lithogenic effects.

I_{geo} values can be used to classify the pollution levels, as follows: $I_{geo} \leq 0$: unpolluted soil; I_{geo} Є (0÷1]: unpolluted to moderately polluted soil; I_{geo} Є (1÷2]: moderately polluted soil; I_{geo} Є (2÷3]: moderately to strongly polluted soil; I_{geo} Є (3÷4]: strongly polluted soil; I_{geo} Є (4÷5]: strongly to very strongly polluted soil; $I_{geo} > 5$: very strongly polluted soil.

Fig. 9 Expoziţiei Park –mean content/NVS ratios as a function of distance from Copou Boulevard.

Tab. 6 The GB for heavy metals

Metal concentration (mg/kg)								
Element	Min.	Max.	Mean	St. Dev.	Geom. Mean	Geom. St. Dev.	GB	Distribution Type
Fe	27626.9	36719.4	31812.9	1914.2	31755.5	1.0	34875.7	Normal
Mn	890.6	944.8	858.1	45.5	856.9	1.0	931.	Normal
As	11.9	14.1	12.8	0.5	12.8	1.0	14.1	Normal
Cr							59.1[1]	
Co							11.8[1]	
Ni							46.9[1]	
Cu							48.6[1]	
Zn							139.3[1]	
Pb							43.7[1]	

[1] after Apostoae and Iancu (2009)

Although the mean values of I_{geo} indicate no or only moderate pollution (Fig. 10), individual values (Tab. 7) highlight certain interesting aspects. Thus, in the case of ES, I_{geo} values indicate either the absence of pollution (Fe, Mn, Ni, Cu, As), or a reduced pollution level (Cr, Co, Zn, Pb). CS displays a much wider variation range for I_{geo}: unpolluted (Fe, Mn, Ni), unpolluted to moderately polluted (Co, Cu, Cr, As), unpolluted to strongly polluted (Pb), and unpolluted to strongly/very strongly polluted (Zn).

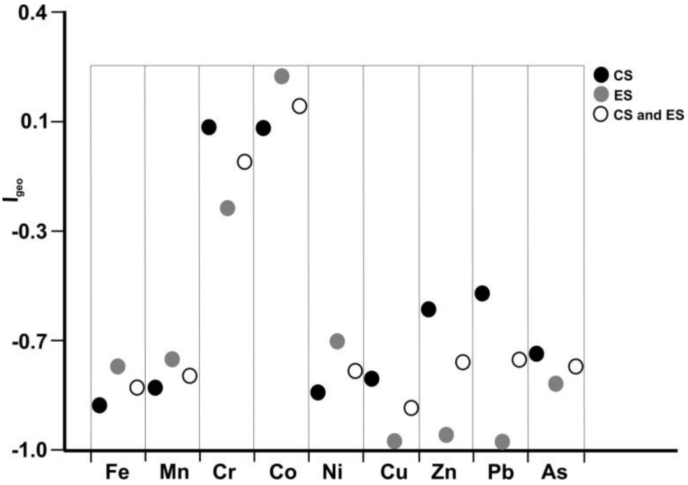

Fig. 10 Mean values of the geo-accumulation index (I_{geo}) for heavy metals.

5.3. The enrichment factor (EF)

The equation used to calculate EF, based on the standardization of a tested element against a reference one (Chester and Stoner, 1973; Zoller et al., 1974), was the following (Buat-Menard and Chesselet, 1979):

$$EF = [(C_{n(sample)} / C_{ref(sample)}]/[B_{n(background)} / C_{ref(background)}] \quad (2)$$

where: $C_{n(sample)}$ is the content of the examined element in the examined environment; $C_{ref\ (sample)}$ is the content of the reference element in the examined environment, in the present case, Mn, regarded by Loska et al. (1997) as conservative lithogenic reference element; $B_{n(background)}$ is the content of the examined element in the upper crust (Reimann et al., 2008); and $B_{ref\ (background)}$ is the content of the examined element in the upper crust, in the present case, Mn, regarded by Loska et al. (1997) as conservative lithogenic reference element).

Based on the EF value, Sutherland (2000) distinguishes the following three intervals: (0÷1]: background concentration; (1÷2]: depletion to minimal enrichment; (2÷5]: moderate enrichment; (5÷20): significant enrichment; (20÷40]: very high enrichment; > 40: extremely high enrichment. The mean values of EF (Fig. 11) suggest the presence of heavy metal contents that vary between the values characteristic for the geochemical background (CS – Fe; ES – Fe), minimal enrichment (CS – Cr, Co, Ni; ES – Cr, Co, Ni, Cu, Zn, Pb) and moderate enrichment (CS – Cu, Zn, Pb, As; ES – As). Individual EF values (Tab. 8) reveal wide variation ranges, which allow a much more precise characterization of the studied parks. Thus, ES displays both heavy metal contents which are characteristic for the geochemical background (Fe), and effects of an anthropogenic input which varies from minimal (Cr, Co, Ni) to moderate (Cu, Zn, Pb, As). CS, on the other hand, is characterized by a much wider range of EF values, suggesting a significant anthropogenic input in the case of Cr, Cu, Zn, Pb and As.

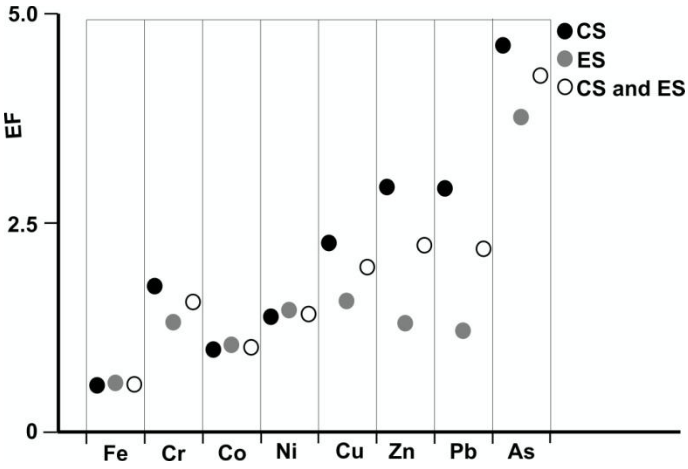

Fig. 11 Mean values of the enrichment factor (EF) for heavy metals.

6. Discussion

6.1. Analysis of the sources of pollutants

The heavy metals present in the soils of urban parks can be regarded as geological variables which could be expressed, in general form, through the following formula:

$$f(x) = (x, y, z, t) \quad (3)$$

where: x, y, z are the geographical coordinates of the park within the urban area; and t is the age of the parks.

As far as the geographical coordinates are concerned, apart from the parent rock of the soil and the process of pedogenesis, which can influence the heavy metal content (Zhang et al., 2008), the location of the parks within the urban area determines the intensity of the factors acting upon the soils (Li et al., 2001; Chen et al., 2005; Onder et al., 2007; Papafilippaki et al., 2008; Jien et al., 2011; Özbaş, 2011). In addition, the age of the parks is critical (Chen et al., 2005; Zhang et al., 2005), as anthropogenic factors act with varying intensity during different historical periods, characterized by highly variable socio-economic conditions.

6.1. 1. The location of the Copou and Expoziției parks within the city of Iaşi

The interdependence between the location of the parks and the heavy metal contents of their soils allows the estimation of the likely anthropogenic sources of these elements.

The industrial activities acknowledged as sources of Co, Mn, Cu, Zn, Cr, Ni, Pb (Lacătuşu et al., 2008; Al Momani, 2009; Cannon and Horton, 2009; Wei and Yang, 2010; Guo et al., 2012) play, in our opinion, a secondary role in the process of heavy metal accumulation in the case of CS and ES. Our view is supported, on one hand, by the great distance between the locations of the parks and that of the industrial area (approximately 8.5 km), and, on the other hand, by the significant reduction in industrial activity in

Tab. 7 Geo-accumulation index (I_{geo}) for the soil heavy metals

Element	Park	≤ 0	%	(0÷ 1]	%	(1÷ 2]	%	(2÷ 3]	%	(3÷ 4]	%	(4÷ 5]	%	≥ 5	%
	CS and ES	85	100	0	0	0	0	0	0	0	0	0	0	0	0
Fe[1]	CS	49	57.6	0	0	0	0	0	0	0	0	0	0	0	0
	ES	36	42.4	0	0	0	0	0	0	0	0	0	0	0	0
	CS and ES	85	100	0	0	0	0	0	0	0	0	0	0	0	0
Mn	CS	49	57.6	0	0	0	0	0	0	0	0	0	0	0	0
	ES	36	42.34	0	0	0	0	0	0	0	0	0	0	0	0
	CS and ES	59	69.4	25	29.4	1	1.2	0	0	0	0	0	0	0	0
Cr	CS	27	31.8	21	24.7	1	1.2	0	0	0	0	0	0	0	0
	ES	32	37.6	4	4.7	0	0	0	0	0	0	0	0	0	0
	CS and ES	16	18.8	69	81.1	0	0	0	0	0	0	0	0	0	0
Co	CS	12	14.1	37	43.5	0	0	0	0	0	0	0	0	0	0
	ES	4	4.7	32	37.6	0	0	0	0	0	0	0	0	0	0
	CS and ES	85	100	0	0	0	0	0	0	0	0	0	0	0	0
Ni	CS	49	57.6	0	0	0	0	0	0	0	0	0	0	0	0
	ES	36	42.4	0	0	0	0	0	0	0	0	0	0	0	0
	CS and ES	78	91.8	7	8.2	0	0	0	0	0	0	0	0	0	0
Cu	CS	42	49.4	7	8.2	0	0	0	0	0	0	0	0	0	0
	ES	36	42.4	0	0	0	0	0	0	0	0	0	0	0	0

Element	Park	≤ 0	%	(0÷ 1]	%	(1÷ 2]	%	(2÷ 3]	%	(3÷ 4]	%	(4÷ 5]	%	≥ 5	%
Zn	CS and ES	78	91.8	5	5.9	1	1.2	0	0	0	0	1	1.2	0	0
	CS	44	51.8	3	3.5	1	1.2	0	0	0	0	1	1.2	0	0
	ES	34	40.0	2	2.4	0	0	0	0	0	0	0	0	0	0
Pb	CS and ES	75	88.2	7	8.2	2	2.4	0	0	1	1.2	0	0	0	0
	CS	41	48.2	5	5.9	2	2.4	0	0	1	1.2	0	0	0	0
	ES	34	40.000	2	2.3	0	0	0	0	0	0	0	0	0	0
As	CS and ES	84	98.8	0	0	1	1.2	0	0	0	0	0	0	0	0
	CS	48	56.5	0	0	1	1.2	0	0	0	0	0	0	0	0
	ES	36	42.3	0	0	0	0	0	0	0	0	0	0	0	0

[1] in relation to the average concentration of Fe in the upper crust (Reimann et al., 2008)

Tab. 8 Enrichment factors (EF) for the soil heavy metals

Element	Park	(0÷1]	%	(1÷2]	%	(2÷5]	%	(5÷20]	%	(20÷40]	%	≥ 40	%
Fe	CS and ES	85	100	0	0	0	0	0	0	0	0	0	0
	CS	49	57.6	0	0	0	0	0	0	0	0	0	0
	ES	36	42.4	0	0	0	0	0	0	0	0	0	0
Mn	reference element												
Cr	CS and ES	2	2.4	69	81.2	14	16.5	0	0	0	0	0	0
	CS	0	0	35	41.2	14	16.5	0	0	0	0	0	0
	ES	2	2.4	34	40.0	0	0	0	0	0	0	0	0

Element	Park	(0÷1]	%	(1÷2]	%	(2÷5]	%	(5÷20]	%	(20÷40]	%	≥ 40	%
Co	CS and ES	16	18.8	69	81.2	0	0	0	0	0	0	0	0
	CS	11	12.9	38	44.7	0	0	0	0	0	0	0	0
	ES	5	5.9	31	36.5	0	0	0	0	0	0	0	0
Ni	CS and ES	0	0	85	100.0	0	0	0	0	0	0	0	0
	CS	0	0	49	57.6	0	0	0	0	0	0	0	0
	ES	0	0	36	42.4	0	0	0	0	0	0	0	0
Cu	CS and ES	0	0	67	78.8	11	12.9	7	8.2	0	0	0	0
	CS	0	0	34	40.0	8	9.4	7	8.2	0	0	0	0
	ES	0	0	33	38.8	3	3.5	0	0	0	0	0	0
Zn	CS and ES	7	8.2	61	71.8	15	17.6	1	1.2	0	0	1	12
	CS	1	1.2	34	40.0	12	14.1	1	1.2	0	0	1	1.2
	ES	6	7.0	27	31.8	3	3.5	0	0	0	0	0	0
Pb	CS and ES	14	16.5	55	64.7	13	15.3	2	2.4	0	0	1	1.2
	CS	4	4.7	31	36.5	11	12.9	2	2.4	0	0	1	1.2
	ES	10	11.8	24	28.2	2	2.4	0	0	0	0	0	0
As	CS and ES	0	0	0	0	82	96.5	2	2.4	1	1.2	0	0
	CS	0	0	0	0	46	54.1	2	2.4	1	1.2	0	0
	ES	0	0	0	0	36	42.4	0	0	0	0	0	0

the city of Iaşi since 1990. The urbanization process in general (Olade, 1987; Lin et al., 2002; Cannon and Horton, 2009; Simon et al., 2013), carried out in the city of Iaşi by means of ongoing large development and rehabilitation works (REGIO, 2012), may generate airborne heavy metals that are difficult to estimate from a quantitative point of view. Commercial and residential complexes in the vicinity of the Copou and Expoziţiei parks, as well as facilities for cultural, educational and recreational activities, may generate heavy metals both through the process of physical decay of the buildings themselves and of the house paints (Chen et al., 2005; Gasana et al., 2006; Yongming et al., 2006; Sutherland et al., 2012), and through the production of solid waste (Pattnaik and Reddy, 2011). In this respect, it is worth noting that in the SW area of Copou Park, across a 50 m radius around the skydiving tower, high levels of Cr (71.5–110.5 mg/kg) and Zn (106.9–3526 mg/kg) have been measured.

The heavy traffic on the streets adjacent to the parks (on Copou Boulevard, in particular) influences the quantity of heavy metals present in the soil through the cumulated effects of vehicle emissions (Pb, As) (Olade, 1987; Chen et al., 1997; Trinidade et al., 2006; Onder et al.,2007; Papafilippaki et al., 2008; Enayatzamir et al., 2008; Jien et al., 2011; Özbaş, 2011; Deocampo et al., 2012) and of the mechanical degradation of the vehicles (Cu, Zn, Ni, Cr) (Chen et al., 2005; Amusan et al., 2003; Akbar et al., 2006; Adelekan and Abegunde, 2011; Fanget al., 2011; Tamrakar et al., 2011). The seven fold increase in the number of vehicles in Iaşi County which occurred between 1998 and 2012 (Address 104005/31.10.2012, Ministry of Administration and the Interior), as well as the use of leaded gasoline prior to 2005 (Government Resolution 689/2004), have contributed to the increase of heavy metals in the soils. An aspect which sets Copou Park apart from Expoziţiei Park is represented by the presence of parking lots adjacent to the commercial complexes nearby, as well as pedestrian crossings, which may lead to additional vehicle emissions. The maintenance works typical for parks in urban areas involve the use of chemical fertilizers, fungicides and insecticides, thus contributing to the increase in Cr, Cu, As and Pb contents of the soils (Semu and Singh, 1996; Hugues et al., 2009; Reinprecht, 2010). Although only approximately 1.2 km apart, from each other ES and CS differ in their morphology. Thus, upon studying the types of soil present on the territory of the city of Iaşi across the 0.0–1.20 m depth range, Lăcătuşu et al. (2005) identified a soil in its natural state, more precisely a cambic chernozem (the Amp, Am, A/B, Bv, C1k, C2k horizons), in ES, and a covered cambic chernozem, highly anthropogenic (the U1I1Am, U2I2Am, Am, Bvk, C1k, C2k horizons), in CS. The presence of a mixture of natural soil horizons and anthropogenic material across the 0.0–0.40 m depth range (the U1I1Am and U2I2Am urban soil horizons) in CS suggests a possible heavy metal source.

6.1.2. The age of the Copou and Expoziţiei parks

The age of the parks is significant, since their historical evolution has favored the accumulation of heavy metals in soils. Although, as far as their age is concerned (Chen et. al., 2005), Copou and Expoziţiei parks are part of classes B ($t > 100$ years) and A ($t < 100$ years) respectively, the anthropogenic factors that have acted upon their soils throughout time are difficult to estimate due to a lack of information in this respect. One could note, however, that the presence of the highly anthropogenic cambic soil mentioned by Lăcătuşu et al. (2005) in Copou Park results from a series of anthropogenic variables which have been affecting the area for a long period of time.

7. Conclusions

The study of heavy metal distribution in the soils of Copou and Expoziţiei parks has highlighted elevated Cr, Cu, Zn, Pb and As contents, whose origin is undoubtedly anthropogenic. At the same time, anthropogenic factors have acted with greater intensity upon the soil of Copou park, compared to that of Expoziţiei park, as reflected by the I_{geo} and EF values and by the placement of the heavy metal contents within the NVS, SAT and SIT limits. Both in the case of Copou park and in that of Expoziţiei park, the Fe, Mn, Co and Ni contents seem to belong to the natural geochemical background. The anthropogenic sources of the heavy metals that have affected the soils of Copou and Expoziţiei parks can be correlated with the location of the parks within the city of Iaşi, as well as with the age of the parks. Overall, the heavy metal contents of the soils of Copou and Expoziţiei parks are within the ranges determined for the heavy metal contents present in the soils of other urban parks across the Globe.

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