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THE HEAVY METALS ABUNDANCE IN SOILS FROM FĂGĂRAŞ DEPRESSION (Cd, Co, Cr, Cu)

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Key words: Făgăraş Depression, soils, heavy metals, abundance.

Abstract. Făgăraş Depression soil cover consists of a mosaic of soils where are prevalent Protisols represented by Fluvisols (39.2%), Luvisols represented by Luvisols and Haplic Luvisols (38.8%), Cambisols represented by Districc Cambisols and Eutric Cambisols (12.5%) and Hydrisols represented by Gleysols, Stagnosols and Histosols (7.5%). Făgăraş Depression soils, predominantly acid, with low content of humus and total nitrogen and a poor supply with phosphorus and potassium mobile forms present total contents of Cd, Co, Cr and Cu lower than normal contents values in soils. This are around the average values existing in the world soils, except Cr, that has a lower average value with 34mg•kg^{-1} than the world soils average. Distribution of heavy metal content values is normal. Frequency histograms are symmetrical and platicurtice, except copper, that has a strong asymmetry of left and leptocurtice shaped curve. Abundance of Cd in Făgăraş Depression is upper the lithospheric and pedospheric abundance of this element, while Cr and Cu abundance is below of the two geogenic types of abundance. Co abundance is below the lithospheric abundance, but higher than pedospheric once. Area distribution of the four heavy metals showed zones with concentrations higher than normal background of the whole territory, but without exceeding normal levels. Inverse proportionally relationships between pH and heavy metal content, statistically assured for Cd and Co; direct proportionality relationship, statistically assured between Cd and humus on the one hand, and between total N and mobile potassium, on the other hand; direct proportionality, statistically assured between Cu and N total and finally, close relationships of direct proportionality, statistically assured between Cd, Co and Cr were established. Copper has shown direct

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correlation, statistically assured, only with chromium. Origin of heavy metals is predominantly geogenic, namely, the minerals and the crystalline rocks of the northern versant of the Făgăraş Mountains. Industrial units present in this area did not influence the content of heavy metals in soils of Făgăraş Depression territory.

Introduction

The stability of the heavy metals abundance in soils is of real practical importance, both in terms of determining whether pollutant risk and in terms of assessing the nutritive potential for plants, as some of these chemicals as micronutrients, have a significant role in plant nutrition.

Evaluation of heavy metal abundance has experienced a large development in countries like Russia, USA, Germany, United Kingdom, Norway, Finland, Poland, etc. As a result of research carried out, atlases presenting the abundance of heavy metals in soils have been drawn up (Fauth et al., 1985; Bølviken et al., 1986, McGrath and Loveland, 1992, Lis and Pasieczna, 1995; Solminen et al., 2005).

In our country, although was not published yet an atlas at national level, have been made studies on specific areas, followed by sequential publication of maps on heavy metals distribution in soils (Răuţă et al., 1981; Lăcătuşu et al., 1981, 1989, 1993, 1994, 1995,1997, Mihăilescu et al., 1986, 1987 a and b, 1988).

Large scale researches were conducted for urban soils, as a result being published two atlases with the distribution of heavy metals in urban soils from Iassy (Iancu et al., 2008) and Bucharest municipalities(Lăcătuşu et al., 2008).

This paper is refers to the distribution of heavy metals in soils predominantly acid, on an area by approximately 1000 km^2, located in the central part of the country, namely the Făgăraş Depression.

1.Material and methods

The entire surface of the depression was investigated, being performed 50 soil profiles located relatively uniform, so as to cover majority of soil types and subtypes (Table 1). Have been collected soil samples, on the geometric horizon, namely 0-20cm, 20-40cm and 40-60cm. Sampling points locations can be seen in Figure 1.

The soil samples were been analyzed in the laboratory, in terms of general chemical properties and total heavy metal content. Thus, the pH was potentiometric measured in aqueous suspension using a glass-calomel double electrode. Humus was measured by the Walkley - Black method, in Gogoaşă modification, total nitrogen was measured by Kjeldhal method and mobile forms of phosphorus and potassium were been extracted with a solution of ammonium acetate-lactate at pH 3.7 (AL), by Egner-Rhiem-Domingo. Phosphorus was measured by spectrophotometry and potassium by flame photometry.

The total content of heavy metals (Cd, Co, Cr and Cu) was measured with flame atomic absorption spectrometer in air-acetylene flame in hydrochloric solution resulted by digestion of soil samples in $HClO_4$-HNO_3 mixture.

Majority of the methods used are standardized by STAS systems or ISO.

Analytical data were statistical processed, computing the values of the spreading and grouping centre parameters Also, correlative relationships between some of the chemical properties were established. The maps of heavy metals distribution trend have been carried out by using Surfer program. Values of geochemical and pedogeochemical abundance indexes were calculated by Lăcătuşu and Ghelase method (1992).

Table 1. Făgăraş Depression Soils

Soil class	Soil types and subtypes	Percent of Depression surface (according to soils map)	Degree of representativeness of collected samples (%)
Protisols	Eutric Leptosols	0,3	
	Haplic Fluvisols		
	Eutric Fluvisols	38,9	
	Total	**39,2**	**34**
Cernisols	Haplic Phaeozems	0,3	
Umbrisols	Skeletic Umbrisols	1,1	**4**
Cambisols	Distric Cambisols	10,7	
	Eutric Cambisols	1,8	
	Total	**12,5**	**28**
Luvisols	Haplic Luvisols	4,8	
	Albic Luvisols	14,7	
	Stagnic Luvisols	19,3	
	Total	**38,8**	**26**
Hydrisols	Mollic Gleysols	2,7	
	Haplic Gleysols	4,5	
	Haplic Stagnosols	0,1	
	Eutric Histosols	0,2	
	Total	**7,5**	**8**
Anthrosols	Erodic Anthrosols	0,6	

2.Results and discussions

2.1.Soil cover of Făgăraş Depression. Făgăraş Depression is located in the middle part of the country, being demarcated to the north by the Hârtibaciu Plateau, to the east by Perşani Mountains, to the west by Sibiu Depression and to the south by Făgăraş Mountains. It has a surface about 1066 Km^2 and a medium altitude by 500m. In fact, this Depression is an alluvial - proluvial plane consisting of a mix of piedmont glacises, developed at contact of the mountain from existing

terraces and floodplains along the Olt Valley and its tributaries. The soils cover is composed from a diversity of types and subtypes, among which prevails Fluvisols and Luvisols up to 78% (Table 1, Figure 1).

The diversity of soil types and subtypes generates a wide range of values for each of analyzed chemical characteristics (Table 2) as evidenced by the values of variation coefficient.

Given the fact that most values of the variation coefficient (VC) except pH values were been higher than 30, for results interpretation were taken into account the median values (Me). Thus, in general, soils are moderately acid, with a low content of humus and total nitrogen, small-medium mobile phosphorus and low mobile potassium supply. Therefore, the soils having a limited trophicity

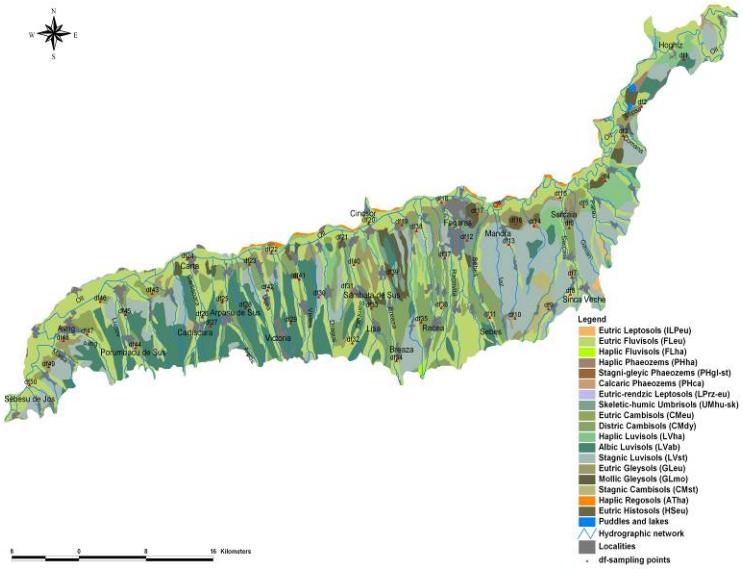

Fig. 1 - The soils map of Făgăraş Depression (according to Romanian Soil Map scale 1:200000 - Braşov and Sibiu sheets)

2.2.General abundance of heavy metals Statistical parameters of the four heavy metals contents shows that individual values extend over large intervals, as indicated by the coefficients of variation values that are greater than 30 (Table 3 and 4). Also it is noted the high standard deviation values (σ). Is highlighted near values of the arithmetic mean (X) and median (Me), as order of magnitude. Also, except for Co, the contents of the first analyzed horizon are higher than those in subjacent horizons.

Separated on classes, types and subtypes of soils, the average values of heavy metals contents (Table 5) shows some differences. Thus, Leptic subtypes of Fluvisols have higher levels of Cd and Cu. At the level of soil classes appear some differences in content, which shows that the highest concentrations of Cd, Co and Cr were registered in Cambisols and of Cu in Hydrosols. Generally, the lower contents of these heavy metals have been recorded in Luvisols. The phenomenon is due to physical and chemical processes that occur in such soils, respectively mobilization and concentration, under reducing conditions in the first case, and eluviation and leaching in the second.

Frequency histograms (Figure 2) show a normal distribution of values, with a strongly left asymmetry only in the case of copper. Excess specific values, show that only copper distribution could be considered high (leptocurtice), other distributions being flat (platicurtice).

Tab. 2 - Statistical parameters of the main chemical properties of soils from Făgăraş Depression (n=135)

Statistical parameter	pH	Humus	Ntotal	C/N	PAL	KAL
		%			$mgKg^{-1}$	
Xmin	4,32	0,84	0,040	4,2	1	23
Xmax	8,18	22,80	0,613	52,4	128	356
X	5,84	3,19	0,140	15,5	24	89
σ	0,94	2,36	0,076	4,6	27	58
VC (%)	16	74	54	30,0	113	65
Xg	5,77	2,70	0,125	15,0	13	74
Me	5,58	2,82	0,133	15,1	14	77
Mo	5,45	2,46	0,130	13,7	11	61

If we compare the average values of total content of Cd, Co, Cr and Cu in soils of Făgăraş Depression with average values of the contents of these chemicals in the world's soils as follows: Cd = 0,48±0,27 $mg \cdot Kg^{-1}$; Co = 9,6±6,1 $mg \cdot Kg^{-1}$; Cr) 57±39 $mg \cdot Kg^{-1}$ and Cu = 22±12,4 $mg \cdot Kg^{-1}$ (values calculated by us after the analytical data of Kabata Pendias and Pendias, 2001) we find that our soils have more Cd, with 0,05 $mg \cdot Kg^{-1}$, more Co, with 3,0 $mg \cdot Kg^{-1}$, but less Cr and copper, with 34,3 $mg \cdot Kg^{-1}$, and 2,6 $mg \cdot Kg^{-1}$ respectively. We believe that the only value of Cr can be considered significant, other values are practically equal to the average values of the world's soils. We note that the mean content values of chemical elements in the world's soils are below normal contents.

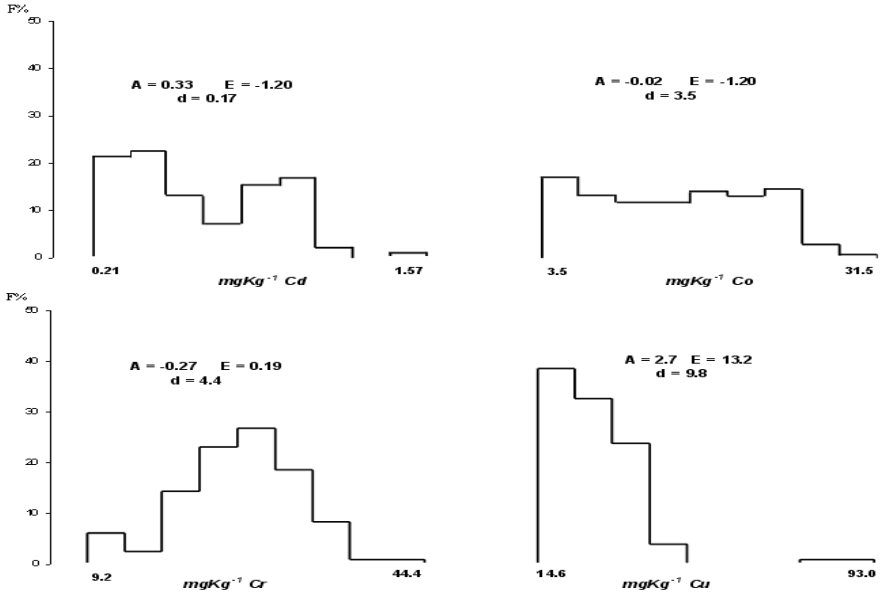

Fig. 2 - Histograms of distribution frequency of heavy metals from Făgăraş Depression soils

Tab. 3 - Statistical parameters of Cd and Co contents in Făgăraş Depression soils

Depth, cm	Cd				Co			
	0-20	0-40	40-60	**0-60**	0-20	20-40	40-60	**0-60**
Statistical parameters								
n	50	49	36	**135**	50	49	36	**135**
Xmin	0,15	0,08	0,04	**0,04**	1,15	0,00	0,45	**0,00**
Xmax	1,40	1,07	1,01	**1,40**	23,5	25,2	28,5	**28,5**
X	0,62	0,54	0,40	**0,53**	13,4	14,2	11,7	**12,6**
σ	0,32	0,32	0,29	**0,32**	7,0	7,8	7,5	**7,8**
VC (%)	51	60	72	**60**	52	55	64	**62**
Me	0,59	0,52	0,39	**0,50**	13,1	13,7	11,0	**12,2**

If calculating the values of accumulation index (AI), as the ratio of median value and the value for normal content in soils, it appears that they are subunit. Therefore, in the analyzed soils, it carried out a small accumulation of heavy metals, not reaching normal values from unpolluted soils (Table 6). Among all the analyzed heavy metals, copper is most closely to normal.

Although in the past, at the Făgăraş Depression have been operated at high intensity (lately much lesser) three industrial units, two of them with chemical

profile, and the third with mechanical profile, at Făgăraş, Victoria and Mârşa, soils are not polluted with heavy metals.

On the other hand, despite the lower abundance of heavy metals in the analyzed soils, had not been reported aspects of nutrition deficiency in plants and animals with any of the four chemical elements. The phenomenon is due to acid soil reaction which contributes to the mobilization in the soil solution of sufficient quantities of chemical elements for plant nutrition.

Tab. 4 - Statistical parameters of Cr and Cu contents in Făgăraş Depression soils

Depth, cm	Cr				Cu			
	0-20	20-40	40-60	**0-60**	0-20	20-40	40-60	**0-60**
Statistical parameters								
n	50	49	36	**135**	50	49	36	**135**
Xmin	6,93	7,91	4,8	**4,80**	5,36	4,81	5,14	**4,81**
Xmax	34,0	33,8	38,7	**38,7**	84,2	80,0	37,3	**84,2**
X	22,6	22,9	21,9	**22,7**	20,6	19,1	18,1	**19,4**
σ	6,9	6,7	7,4	**6,8**	12,1	11,9	8,4	**11,2**
VC (%)	31	21	34	**30**	59	62	47	**58**
Me	22,8	22,8	23,7	**23,6**	19,1	18,1	17,5	**18,0**

Tab. 5 - The heavy metals content according to the class, type and subtype of soils from Făgăraş Depression

Soil class	**Soil type and subtype**	**Cd**	**Co**	**Cr**	**Cu**
		mg·Kg^{-1}			
PROTISOLS	Gleyc Fluvisols (FL gl)	0,243	5,02	14,9	17,0
	Calcaric Fluvisols (FL ca)	0,298	9,20	22,2	20,9
	Distric Fluvisols (FL dy)	0,627	14,75	25,0	17,2
	Eutric Fluvisols (FL eu)	0,517	11,80	21,1	17,8
	Litic Fluvisols (FL li)	1,000	23,4	24,4	44,9
	Total	**0,537**	**12,8**	**21,5**	**23,6**
UMBRISOLS	Umbrisols (UM)	**0,594**	**15,3**	**27,1**	**26,2**
CAMBISOLS	Distric Cambisols (CM dy)	0,686	15,5	24,4	20,8
	Fluvi-eutric Cambisols (CMeu-fv)	0,638	18,4	25,9	20,5
	Lepti-eutric Cambisols (CM eu-le)	0,691	14,9	27,0	24,1
	Total	**0,672**	**16,3**	**25,8**	**21,8**
LUVISOLS	Stagnic Luvisols (LV st)	0,433	9,0	20,4	12,0
	Albic Luvisols (LV ab)	0,548	14,3	23,2	8,5
	Haplic Luvisols (Lv ha)	0,459	8,8	20,8	12,9
	Total	**0,480**	**10,7**	**21,5**	**11,1**
HYDRISOLS	Mollic Gleysols cernic (GL mo)	0,232	8,6	13,7	26,4
	Histic Gleysols (GL hi)	0,653	11,2	27,4	39,4
	Total	**0,443**	**9,9**	**20,5**	**32,9**

Tab. 6 - Accumulation index values (AI) of Cd, Co, Cr and Cu in Făgăraş Depression soils

Chemical element	AI
Cd	0,50
Co	0,81
Cr	0,79
Cu	0,90

2.3.Geochemical and pedogeochemical abundance

The values of the geochemical and pedogeochemical abundance indicates the ratio between general content of heavy metals in soils studied and overall abundance of these heavy metals in the lithosphere and in the pedosphere, respectively.

Tab. 7 - Values of geochemical abundance index (GAI) and pedogeochemical abundance index (PgAI) for Cd, Co, Cr and Cu from Făgăraş Depression soils

	Cd	Co	Cr	Cu
GAI	4,07	0,70	0,27	0,41
PgAI	1,77	2,52	0,76	0,97

Subunit values indicate a content less than Clark's value (average content of a chemical element in the lithosphere) or general average content of soils, and values higher than one indicate a superior content (and Ghelase, 1992).

By analyzing data from table 7 we also see that in Făgăraş Depression soils, only Cd can be found at content level higher than Clark's value. On the other hand, Co, Cr and Cu can be found at relative values lower than 0, 30; 0, 73 and 0, 59 as compare with the lithospheric abundance.

If we relate to the general medium content of the soils, we could notice that it was exceed of 1.8 and 2.5 times, only for Cd and Co, while Cr and Co have registered relative values lower, with 0.24, 0.03 respectively.

Therefore, the abundance of Cd in Făgăraş Depression soils is upper the pedospherical and lithospherical abundance, while the abundance of Cr and Cu is lower than these two types of abundance. Co abundance in the analyzed soils is below lithospherical abundance, but higher than the pedospherical.

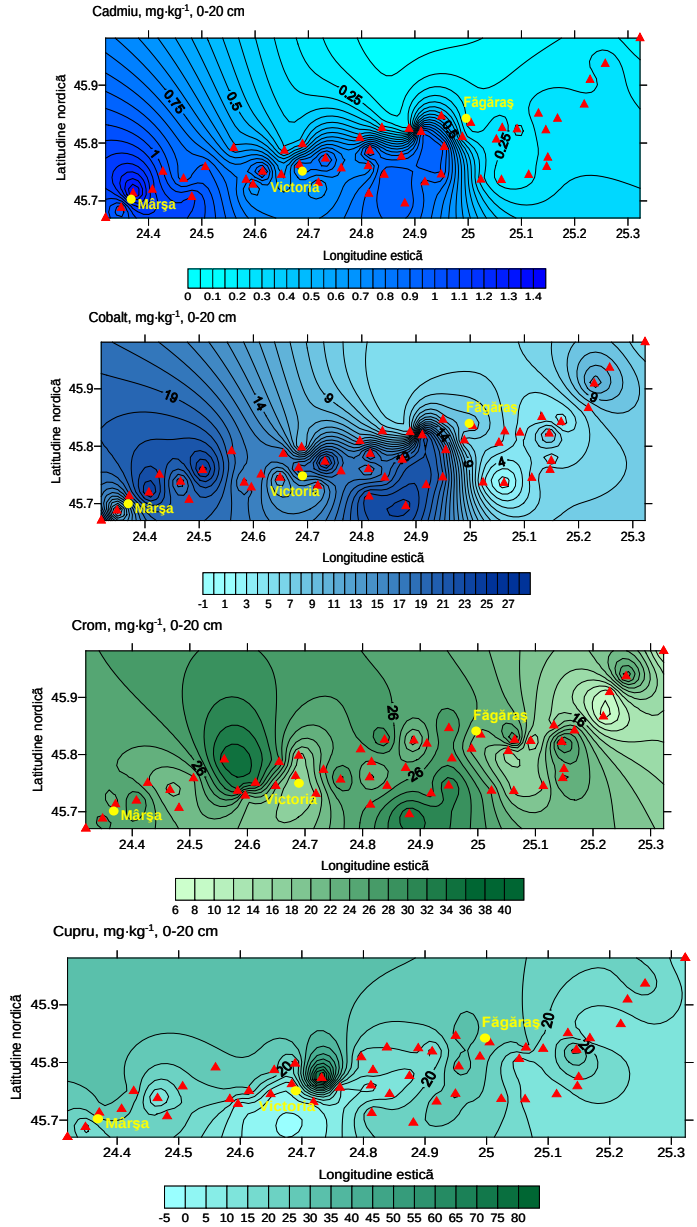

Fig. 3 - Area distribution of heavy metals in Făgăraş Depression

2.4.Area distribution of heavy metals Although majority of the ranges of values of the four heavy metals concentrations fall within a contents domain below

the normal contents, however, within these ranges are separates two areas with higher contents levels (Fig. 3).

Thus, for cadmium, isoconcentrations curves with values from 0, 5 to 0, 75mg·Kg^{-1} are agglomerate, on the direction, almost central, from NE to SW, namely from Făgăraş to Mârşa. Cobalt isoconcentrations curves, with values ranged from 14 to 23 mg·Kg^{-1} are overlapping in the same area.

On the map of Cr distribution trend can be separate multiple areas with contents that reach to over 30 mg·Kg^{-1}. They occur in the northern side of Victoria town, in the southeastern side of Făgăraş town, and isolated in the eastern part of a perimeter located along an alignment Şercaia-Şinca.

Copper distribution is monotonous, with background values around 20 mg·Kg^{-1}, except an area located between localities Viştea de Sus and Viştea de Jos, where the Cu concentration in the upper horizon reaching 70-80 mg·Kg^{-1}.

Tab. 8 - The values of correlation coefficients between contents of Cd, Co, Cr, Cu and the main agro-chemical characteristics of soils (n = 135) from Făgăraş Depression

Agro-chemical characteristics	pH	Humus	Ntotal	P_{AL}	K_{AL}
Chemical element					
Cd	-0,307**	0,359**	0,296**	0,087	0,214*
Co	-0,235**	0,143	0,078	0,077	0,249
Cr	-0,155	0,122	0,086	0,077	0,079
Cu	-0,121	0,192	0,224*	0,066	0,186

Tab. 9 - The values of correlation coefficients (the first number) and of correlation ratios (the second number) between the heavy metals contents from soils (n = 135) from Făgăraş Depression

Cd	**Cd**			
Co	0,858** 0,869**	**Co**		
Cr	0,436** 0,523**	0,445** 0,447**	**Cr**	
Cu	0,170 0,245	0,182 0,245	0,440** 0,465**	**Cu**

2.5.Heavy metals correlations. Between the total content of Cd, Co, Cr and Cu, on the one hand and pH H_2O values have been set inverse proportionality relationship, statistically assured for Cd and Co. Acid reaction is favoring accumulation of heavy metals and especially their mobilization in the soil solution.

As can be seen in Table 8, between contents of heavy metals and others agro-chemical characteristics have been set directly proportional relationship, statistically assured for Cd, except for mobile P, soluble in AL. Also, the relationship between copper and nitrogen is statistically assured. In fact, the copper affinity for nitrogen is known. Consequently, the latter was introduced in the calculation formula for determining the copper deficiency index (Lăcătuşu, 2006).

We notice chromium correlation with none of the considered agrochemical characteristics, and correlation of mobile phosphorus soluble in AL with any analyzed heavy metal.

Instead, it highlights the inter-correlations of heavy metals (Table 9) with values of correlation coefficients and correlation ratios statistically assured, except those of copper with cadmium and cobalt. The closest correlative links are those of Co with Cd. Such direct proportionality relationship, statistically assured, demonstrate the common origin of heavy metals in these soils. They are coming from minerals of the altered crystalline rocks carried from the northern slope of the Făgăraş Mountains and deposited in the depression area to the north.

Conclusions

The soil cover of the Făgăraş Depression consists of a complex of soils, composed by Protisols represented by Fluvisols (39.2%), Luvisols (38.8%), Cambisols represented by Distric Cambisols and Eutric Cambisols (12.5%), and Hydrosols represented by Gleysols, Stagnosols (5.5%) and Histosols (2%).

The Făgăraş Depression soils are predominantly acidic, with low contents of humus and total nitrogen and a reduced supply of mobile forms of phosphorus and potassium.

Total contents of Cd, Co, Cr and Cu in the Făgăraş Depression soils are lower than the normal content in soils. They are around the average values of the world soils, except Cr which has an average value lower than average of the world soils with 34 $mg \cdot Kg^{-1}$.

The distribution of heavy metal content values is normal. The frequency histograms are symmetrical and platicurtice, except copper, which has a strong left asymmetry and a leptocurtice shaped curve.

The abundance of Cd in the Făgăraş Depression soils is higher than its lithospherical and pedospherical abundance, while Cr and Cu abundance is below of the two types of geogenic abundance. The abundance of Co is lower than lithospherical abundance, but higher than the pedospherical.

The area distribution of the four heavy metals revealed zones with concentrations higher than normal background of the whole territory, without exceeding, however, the normal levels. Thus, on a NE-SW direction, from Făgăraş to Mârşa have been highlighted higher concentrations of Cd and Co. As islands, zones with higher concentrations of Cr and Cu have been outlined over the entire surface.

Inverse proportionality relationship between pH and heavy metal content, statistically assured for Cd and Co and direct proportionality relationship, statistically assured between Cd and humus on the one hand, and between total N and mobile potassium, on the other hand, have been established. Also, a direct relationship, statistically assured, has been established between Cu and N total.

Close direct proportionality relationships, statistically assured, have been calculated between Cd, Co and Cr. Copper is directly correlated, statistically assured, only with chromium.

The heavy metals source is predominantly geogenic, originating in minerals and crystalline rocks from the northern slope of the Făgăraş Mountains.

The existing industrial units on the Făgăraş Depression territory did not influenced the content of heavy metals in soils.

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