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A study on cadmium, lead, zinc and cobalt concentration in Baia Mare County, N-W Romania soils

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Abstract. Heavy metals are naturally present in soils, and are predominantly inherited from the soil's parent material but are significantly altered by anthropogenic activity which can pose both environmentals and human health risks. The main soil pollution source is: mine entrances (mine openings) ore processing factories, metallurgical factories. Naturally around 0.9 million ha of soils are affected by chemical pollution and 0.2 million of them by excessive pollution. Between all the soil pollution contaminants heavy metals Cu, Pb, Zn and Cd or acid precipitation, this represent aggressive effects on soil and usually can be found in areas like Baia Mare, Copșa Mică and Zlatna. This paper aims to investigate the contents of heavy metals (Cd, Pb, Zn and Co) in soils from Baia Mare county from N-W Romania. Samples of soil was collected with the pedological drill, the sampling depths were 0-20 cm, 20-40 cm, 40-60 cm and 60-80 cm, disaggregated in the Berghof MWS2 microwave and the chemical compositions of soil was determined through AAS method. Regarding the heavy metals from areas investigated, the highest concentration of this heavy metals was found in Ferneziu (F) area in 20-40 cm horizon (25.18 ± 1.53 mg/kg Cd; 3290.18 ± 177.42 mg/kg Pb; 1941.55 ± 28.51 mg/kg Zn). These values decrease with the soil depth, the soil profile at the depth of 60-80 cm being 16.86 ± 1.45 mg/kg Cd; 2531.20 ± 193.62 mg/kg Pb; 1364.74 ± 38.18 mg/kg Zn. These concentration values for heavy metals exceeded the maximum allowable limits (MAL) covered by law. The lowest concentrations of heavy metals was found in Tăuții Mărgherăuș (TM) area the concentratin in this case are below MAL (0.22 ± 0.03 mg/kg Cd [1 mg/kg MAL]); 13.49 ± 0.75 mg/kg Pb [20 mg/kg MAL]; 55.68 ± 1.57 mg/kg [100 mg/kg-MAL] and 9.19 ± 0.26 mg/kg-Co [15 mg/kg MAL]. These high concentrations could be an effect of pollution from tow metallurgical factories in Baia Mare, Romplumb and Cuprom which are the main sources of pollution from the city and also for the neighboring agricultural areas. The areas studied near the source of pollution have greatest heavy metals concentrations compared with Seini (S) and Tăuții Mărgherăuș (TM) areas which are at considerable distance from the source of pollution. Based on the data presented we also can say that in this area the effect of heavy metal pollution is lower than in other areas we studied. One possible explication for reducing heavy metals concentration is distance from the source of polution.
Key Words: Contaminated soil, FAAS, heavy metals, metal availability, trace element contents.

Rezumat. Metalele grele au fost identificate în diferite componente ale mediul înconjurător, în sol, aer, apă, dar și în plantele din întregul univers. Metalele grele când sunt în concentrați mari sunt considerațe a fi poluante ai mediul înconjurător din cauza diverselor efecte negative asupra sănătății umane. În ultimii 15 ani, emisiile atmosferice de praf din județul Maramureș a scăzut semnificativ. În anii 2001, 2008 și 2013 semisiile totale au fost de: 25.0, 2.13 și 2.05 tone. Acestă tendință este rezultatul inchiderii mai multor instalații industriale sau scăderea producției acestora, precum și punerea înaplicare au unor măsuri de protecție a mediul înconjurător în industrie și transporturi. Pentru această cercetare au fost selectate următoarele areale: cartierul Ferneziu (F), cartierul Săsar (S), zona centrală a municipiului Baia Mare (Centru (C)), ca și zone poluate, dar și cartierul Seini (S) și Tăuții Măgherăuș (TM) ca și zone de referință. Această cercetare îți propune să investigheze conținutul de metale grele (Cd, Pb, Zn și Co) în solurile din municipiul Baia Mare N-V României. Probele de sol au fost recoltate cu ajutorul sondei pedologice pe adâncimea profilului de sol: 0-20 cm, 20-40 cm, 40-60 cm, 60-80 cm. În ceea ce privește concentrația metalelor grele din arealele luate în studiu, cea mai mare concentrație a acestor metale grele au fost identificate în arealul Ferneziu (F) la adâncimea de 20-40 cm horizon (25.18 ± 1.53 mg/kg Cd; 3290.18 ± 177.42 mg/kg Pb; 1941.55 ± 28.51 mg/kg Zn). Aceste concentrații depășesc conentrația maxima adimisă de lege (MAL). Cele mai mici concentrații de metale grele au fost identificate în arealul Tăuții Măgherăuș (TM), concetrația metalelor grele investigate în acest caz sunt sub limita maximă

adimisă (MAL) (0.22 ± 0.03 mg/kg Cd [1 mg/kg MAL]; 13.49 ± 0.75 mg/kg Pb [20 mg/kg MAL]; 55.68 ± 1.57 mg/kg [100 mg/kg-MAL] and (9.19 ± 0.26 mg/kg-Co [15 mg/kg MAL]). Aceste concentrații mari reprezintă efectul poluări care provine de la cele două mari inteprinderii din Baia Mare, Romplumb și Cuprom care de altfel sunt principalele surse de poluare din municipiu dar și pentru suprafețe agricole învecinate. Zonele studiate din apropierea sursei de poluare prezintă cele mai mari concentrații de metale grele în comparație cu arealul Seini (S) și Tăuții Măgherăuș (TM) zone care sunt la distanță considerabilă față de sursa de poluare. Pe baza datelor prezentate, putem afirma de asemnea că aceste zone prezintă concentrați de metale grele mai scăzute decât în alte areale luate în acest studiu. O posibilă explicație pentru acestă reducere a concentrației de metale grele este distanța față de sursa de poluare.
Cuvinte cheie: Contaminarea solului, FAAS, metale grele, disponibilitatea metalelor, trasabilitatea conținutului de metale.

Introduction. Heavy metals have been reported in different components of the enviroment in soil, air, water and plants throughout the universe. Heavy metals contaminants when present at high concencentration are considered as environmental pollutants because of their adverse effects on human health (Yang et al 2007; Nawab et al 2015; Ok et al 2011; Oroian et al 2012; Peter al 2012).

The social and economic development from the last century requires the implementation of some measures to prevent the unsustainable exploitation of soil and subsoil and to protect the flora, fauna, air, water, both at local and global level. Contamination of the soil is more and more frequent, negatively affecting the life cycles, and therefore the quality of the environment is necessary, including the determination of the pollution levels (Burtică et al 2000; Roman 1994; Murgoi et al 2016a; Butean et al 2014; Damodaran et al 2011; Hasan 2015).

At the national level, chemical pollution of soil affect approximate 0.9 million ha, of which 0.2 ha are affected by excessive pollution. Pollution by heavy metals (Cu, Pb, Zn, Cd) and acid precipitation, due to SO_2 have aggressive effects upon soil and are present especially in areas like Baia Mare, Copșa Mică and Zlatna. Thus, it is very helpful to understand the content of heavy metals in urban soil to determine with precision the degree of soil pollution and take the possible measures to decrease the effects of pollution and rehabilitate the affected areas (Bora et al 2013).

Over the last 15 years, atmospheric emissions of dust in Maramureș County significantly decreased. In 2001, 2008 and 2013 years total emissions of heavy metals were: 25.0, 2.13 and 2.05 tons. This trend is a result of the closure of many industrial facilities or the decrease of their production, as well as the implementation in industry and transport activities of a lot of measures to protect the environment (Butean et al 2014).

In modern ecology soil pollution is any action which results in disruption of the normal operation of the soil. The soil represents a support and a living environment in different natural or anthropogenic ecosystems. Disturbances are manifested by chemical, physical or biological degradation.

Food chain contamination with these heavy metals is one of the most important sources of entry into the humans and other living organism (Khan et al 2013; Nawab et al 2015). Different antropogenic (minig, fuel production smelting, energy, intensive agriculture, wastewater irrigation and sewage sludge application) and geological (weatheringg of rocks and deposits and dumping) are the innitial source of toxic metals (Ok et al 2007; Chanpiwat et al 2010; Shah et al 2010; Wei & Yang 2010; Kong et al 2011; Nawab et al 2015; Peter et al 2011). Numerous research papers have been conducted to understand the soil contamination with heavy metals derived from antropogenic soucers such as mining process and smelting activities, cement productions, waste incineration, vehicle exhaust, fertilizers and using pesticides (Ahmad at al 2012; Moon et al 2011; Murgoi et al 2016b; Mohmoudabadi et al 2015; Zauro et al 2015; Roba et al 2015; Nica 2015).

Previous studies have established the deleterious effects of heavy metals and other elements on human health and environmental quality. In fact many governments in Europe and worldwide regulate the limits of elements in soils and sediment, both in industrial and residential areas in an effort to protect the health of its citizens (Paulette et al 2015; Brevik 2013). Other studies in Eastern Europe have reported on heavy metal pollution using traditional lab-based methods, or soil spatial variability as a natural topographic variant, and a few studies have investigated the tow simultaneously, let alone in critical rural/urban interfaces.

In Romania element limits in soils were established by the Romanian Ministry of the Forest, Waters, and Environment (1997) establishing normal values, alert limits, and intervention (action) limits; the latter two of which are further defined by sensitive soil and less sensitive soils. For illustrate the similarities and differences between naturally occurring soil elemental concentrations and those artificially imposed by anthropogenic impacts, we will assess elemental levels with comparison to the more sensitive intervention limits, commonly used for agricultural and residential soil quality assessment along with common soil elements using a proximal sensing system *in-situ* (Paulette et al 2015;). In Romania government mandated action limits for soil elemental concentrations include the followings: Cd (1 mg/Kg); Pb (20 mg/kg); Zn (100 mg/kg) and Co (15 mg/kg) (Romanian Ministry of the Forest, Waters, and Environment [1997]).

The hazard represented by high heavy metals concentration in soils from Baia Mare area of North-West Romania, has been widely publicized and studied extensively since 1995 in several articles (Lăcătușu et al 1995, 1996, 2002; Ciobanu et al 1999; Culicov et al 2002; Damian et al 2010, 2008; Levei et al 2009; Bora et al 2013, 2015a). Soil pollution is caused by the heavy metals bearing dust emissions and dispersion from the pollutant sources during over 100 years, with a strong increase in the last 50 years (Mihali et al 2013).

An increase in metal concentration also influence the soil microbial communities, especially respiration and enzymatic activity by blocking essential functional groups, displacing essential metal or modifying the ative conforming of biological molecules that serve as good indicators of metal pollution (Doelman et al 1994; Hasan 2015). Several studies have shown the negative relations between heavy metals concentrations and microbial activity. However at relatively low concentrations some heavy metals ions (Pb, and Cd) are essential for microorganisms growth since they provide vital cofactors for metallo-proteins (Doelman et al 1994).

Material and Method

Description ot he sampling area. The municipality of Baia Mare is located in Baia Mare depression, on the middle of the river Săsar in north-west of Romania ($47^0$39'-$47^0$48' North and $23^0$10'-$23^0$30' East) (Figure 1). Baia Mare has an average altitude of 228 m abobe de sea level, totaling an area of about 675 km^2 (Vaum-Ivascu 2011). Mostly this depression is dominated bay meadows and terraces. The relief in this depression lends itself more to the vertical zoning. The structure of this municipality is consists from Blidari, Firiza, Valea Neagră (Valley) and Valea Borcutului (Valley). To the north it bordering whit Igniș and Gutâi mountains, in south whit localities Recea and Groși and east with localities Baia Sprie and Tăuții Măgherăuș (Miloiu 2008). The dominant winds in this region are the west-northwest (V-NV) and north-east (N-E). The climate is moderately continental with anual average temperature of 9.4^{0}C, and the annual average rainfall of 857.5 mm/m^2. High temperature oscillations and rapid changes of wether are the characteristics of this region (Bora et al 2014).

The main sources of pollution in this area are two major factories Cuprom SA and S.C. Romplumb S.A. The mines were closed on January 2007, but their waste facilities (tailing ponds and dams) still have a negative impact on the environment. In this area no closure or remediation works were done on these facilities (Butean et al 2014).

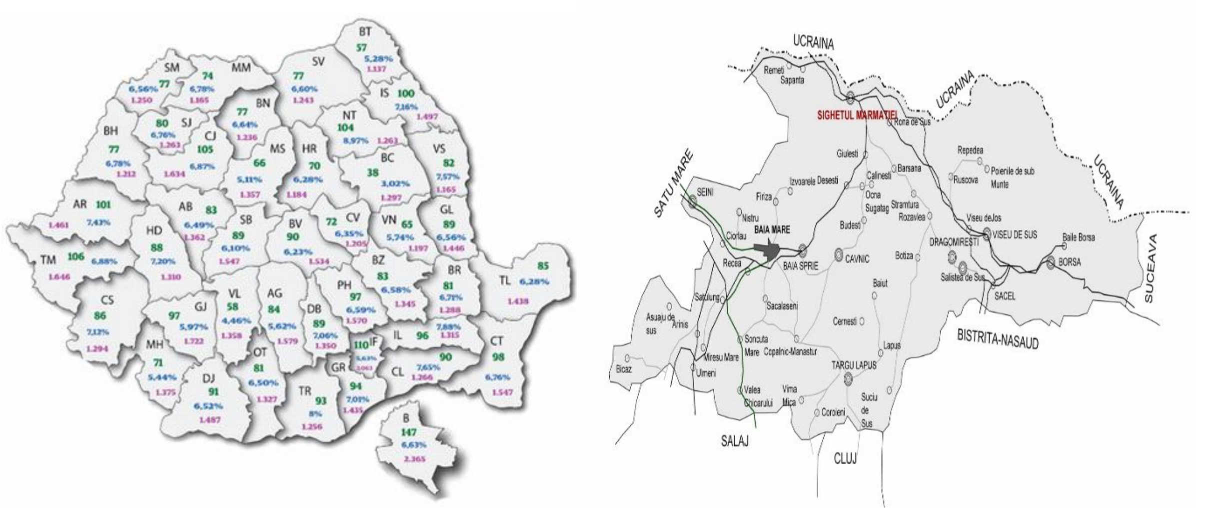

Figure 1. The studied area and the sampling locations.

Soil sampling. For study speific areas have been selected: Ferneziu district, Săsar district the central zone of the municipality (Centru) as polluted areas, Seini and Tăuții Măgherăuș district as reference area. The main sol types from the studies areas: aluviosols and eutricambosols in Ferneziu area aluviosols and luvosols in Săsar, luvisols in Center aluviosols and entricambosols in Seini (Florea & Munteanu 2003; Damian et al 2008; Mihali et al 2013; Bora et al 2013).

The sampling was carried out from July-November 2014 on four depths (3 samples/depth) (0-20 cm; 20-40 cm; 40-60 cm; 60-80 cm) aimed to cover the entire surface of selected areas. Sampling was performed with a handle steel soil sampler. Agrochemical sampling depth of 0-20 cm is performed after removing dust, roots, leaves or other residues from the surface. The amount of collected sample is between 0.5-1 kg. Each sample was placed in properly labeled plastic bags and closed according ISO 11464/2006.

The sampling locations include residential areas and public parks. They have been chosen in each studied area by drawing an imaginary area of 1 m^2 on whose diagonal 3 samples of approximately 0.5-1 kg have been extracted from a depth between all four depths. In Ferneziu and Seini district the sampling points were established primarily in private grounds/areas.

The soil sampler were crushed and dried at room temperature for 48 hours, after this operation, they have been dried at 105^0C using oven model Sanyo/Sterilizer MOV-112 S. Subsequently samples pulverization and homogenization was performed using automatic mojar Resh 110 Germany, 50 g of homogenized samples were prelevated for further analysis. For disaggregation soil samples the working protocol ISO 11466/2006 was used. An amount of 0.2-0.5 g dried and milled material was put into 12 mL aqua regia (9 mL HCl + 3 mL HNO_3) and after 15 minutes the mineralization was performed bay using a microwave Berghof MWS-2 set in 2 steps (Tabel 1). For dilution of soil samples was used 50 mL ultrapure water using (Milli-Q Integral ultrapure water-Type 1) (Bora et al 2015b).

Table 1

Working parameters in the microwave for decomposition of soil samples

Working Steps	*1*	*2*
Temperature (^{0}C)	180	100
Power (%)	99	98
Time (minutes)	25	10

Berghoff MWS-2 devices user manual.

Chemical analysis. The soil samples were analyzed by FAAS (Perkin Elmer AAnalyst 800, Shelton USA). Flame AAS is the official method of analysis for the determination of trance elements with relatively high concentrations according to EU regulations. The analysis precision was usually very good, being on average above 1% for all the elements considered at the mg/L or mg/kg concentration level.

All reagents used were of analytical grade (Merk). Stock standard solutions were prepared weekly. There were used only standard solutions with commercially distilled water (Merck) at a concentration of 1000 mg/L for mineral elements which will be determined. The intermediate solutions were stored in polyethylene bottles and glassware was cleaned by soaking in 10% v/v HNO_3 for 24 hours and rinsing at least seven times with ultrapure water. For quality control purpose blacks and triplicates samples (n=3) were analyzed during the procedure. The variation coefficients were under 10% and detection limits (mg/L) were determined by the calibration curve method. LOD (limit of detections) and LOQ (limit of quantification) limits were calculated according to the next mathematical formulas: LOD=3 SD/s and LOQ=10 SD/s (SD=the estimation of the standard deviation of the regression line, and s=slope of the calibration curve) (Table 2).

Table 2

Instrumental conditions for the determination of each element (FAAS technique)

Element	*Wavelength*	*Slit*	*Correlation coefficient*	*Flame (2300^0C)*	*Background correction*	*LOD* (mg/L)*	*LOQ** (mg/L)*
Cd	228.8	0.7	0.999999	Air-Acetylene	Deuterium	0.028	0.093
Pb	283.3	0.2	0.999853	Air-Acetylene	Deuterium	0.051	0.166
Zn	213.9	0.7	0.999999	Air-Acetylene	Deuterium	0.018	0.059
Co	240.7	0.2	0.999900	Air-Acetylene	Halogen	0.120	0.399

*Detection limit.
**Quantification limit.

Statistical analysis. Statistical interpretation of results was done with the Duncan test using version 22 of SPSS (SPPS Inc. Chicago, IL, USA). The statistical processing of results was done primarily to calculate the following statistical parameters: arithmetic mean, standard deviation, mean error by using the statistical package SPSS. Data was interpreted by analysis of variance (ANOVA), media separation was done using the Duncan test at $P \leq 0.05$. Person's correlation was done using version 22 of SPSS (SPPS Inc. Chicago, IL, USA).

Results and Discussion

The content of heavy metals in Ferneziu (F) area. The characteristic type of soil from Ferneziu (F) is aluviosols (AS) and eutricambosols (EC) class. The soil is characterized by a pH from 5.7-7.5, a content of 3.48-6.40% humus, degree of base saturation from 61.7-92.5%, 0.23% N total; 745.62 ($mg \cdot kg^{-1}$) P mobile and 515 ($mg \cdot kg^{-1}$) k mobile (Mihali et al 2013). Redox potential between 326-358 (mV) and electronic conductivity between 173.8-196.0 (µS/cm) (Bora el al 2013).

Regarding the heavy metals concentrations from Ferneziu (F) area, it can observed (Tabel 3), that the highest Cd, Pb, Zn and Pb concentrations were found in 20-40 cm horizon (25.18 ± 1.53 mg/kg Cd; 3290.18 ± 177.42 mg/kg Pb; 1941.55 ± 28.51 mg/kg Zn). These values decrease with the soil depth profile sol in 60-80 cm (16.86 ± 1.45 mg/kg Cd; 2531.20 ± 193.62 mg/kg Pb; 1364.74 ± 38.18 mg/kg Zn). The lowest concentrations of these heavy metals were fond in: 40-60 cm (16.86 ± 1.45 mg/kg Cd), 60-80 cm (2531.20 ± 193.68 Pb), 0-20 cm (1364.74 ± 38.18 mg/kg Zn). For Co the situation is different, the highest values were found in 0-20 cm interval (20.48 ± 0.28 mg/kg) and the lowest in 20-40 (13.53 ± 0.48 mg/kg).

Concentration values for heavy metals exceeded the maximum allowable limit required by law (25.18 ± 1.53 mg/kg Cd [1 mg/kg-MAL]; 3290.18 ± 177.42 mg/kg Pb

[20 mg/kg MAL]; 1941.55 ± 28.51 mg/kg [100 mg/kg-MAL] and 20.48 ± 0.28 mg/kg Co [15 mg/kg-MAL]).

The differences between variants were statistically assured (F = 5.453; p = 0.025 for Cd); (F = 17.117; p = 0.001 for Pb); (F = 18.918; p = 0.001 for Zn) and (F = 14.673; p = 0.001 for Co) (Tabel 3).

This high values could be an effect of pollution from tow metallurgical factories in Baia Mare, Romplumb and Cuprom which are the main sources of pollution from the city and also for the neighboring agricultural areas.

The average content in soil of each studied heavy metal was compared with content of the same metal with the MAL (maximum allowable limit) according to Romanian legislation (Order no 756/3 November 1997). In the case of Cd, Pb, Zn and Co for this area, the concentration of this heavy metals are much higher than the MAL (21.14 ± 3.05 mg/kg Cd [MAL 1 mg/kg]; 2854.76 ± 174.45 mg/kg Pb [MAL 20 mg/kg]; 1561.15 ± 35.69 mg/kg Zn [MAL 100 mg/kg] and 17.43 ± 0.42 mg/kg [MAL 15 mg/kg]).

The content of heavy metals in Săsar (S) area. The characteristic type of soil from Săsar (S) is aluviosols (AS) and luvosols (LV) class. The soil is characterized by a pH of 4.5-7.2, a content of 3.84-10.05% humus, degree of base saturation of 22.91-97.25% (Mihali et al 2013). Redox potential between 242.37-355.33 (mV) and electronic conductivity between 112.00-147.62 (μS/cm) (Bora el al 2013).

Heavy metals concentrations from Săsar (S) area, exhibit (Tabel 4), a differentially distributed heavy metals throughout the soil profile. In case of Cd the highest concentrations were found in 0-20 cm horizon (5.64 ± 0.30 mg/kg Cd), and the lowest was found in 20-80 cm (1.02±0.07 mg/kg). On the other hand, the highest concentration of Pb, Zn was recorded on the depth of soil profile between 60-80 cm (789.40±1.74 mg/kg Pb) and 40-80 cm (1773.73 ± 36.78 mg/kg Pb; 1688.73±69.81 mg/kg Zn) these values are equal in terms of statistics, and the lowest concentration was found in the surface of the soil (0-20 cm 640.02 ± 13.80 Pb and 20-40 cm 1271.51 ± 21.24 mg/kg Zn). For Co, concentration values of this heavy metal are equal in terms of statistics.

Concentration values for heavy metals exceeded the MAL by law (5.64 ± 0.32 mg/kg Cd [1 mg/kg-MAL]; 789.40 ± 1.74 mg/kg Pb [20 mg/kg MAL]; 1688.73 ± 69.81 mg/kg Zn [100 mg/kg-MAL]). In case of Co concentration (6.63 ± 0.14 mg/kg) is less than the MAL (15 mg/kg).

The differences between variants were statistically assured (F = 24.714; p ≤ 0.000 for Cd); (F = 3.725; p = 0.061 for Pb); (F = 12.758; p = 0.002 for Zn) and (F = 2.023; p = 0.189 for Co) (Table 4).

Table 3

The content of heavy metals in soil from the Ferneziu (F) area (mg/kg)

Depth (cm)	*Location*	*Cd*	*Pb*	*Zn*	*Co*
		*MAL**	*MAL**	*MAL**	*MAL**
		1 mg/kg	*20 mg/kg*	*100 mg/kg*	*15 mg/kg*
0-20	Ferneziu (F)	20.82±4.15 ab	2645.12±94.74 c	1364.74±38.18 c	20.48±0.28 a
20-40		25.18±1.53 a	3290.18±177.42 a	1941.55±28.51 a	13.53±0.48 c
40-60		16.86±1.45 c	2952.57±57.52 b	1610.81±53.56 b	17.74±0.55 b
60-80		21.69±2.00 ab	2531.20±193.68 c	1327.51±22.54 c	17.43±0.36 b
Average		21.14±3.05	2854.76±174.45	1561.15±35.69	17.43±0.42
Minimum values		16.86±1.45	2531.20±193.68	1327.51±22.54	13.53±0.48
Maximum values		25.18±1.53	3290.18±177.42	1941.55±28.51	20.48±0.28
F (Fisher Factor)		5.453	17.117	18.918	14.673
Sig.		p = 0.025	p = 0.001	p = 0.001	p = 0.001

Average value ± standard deviation (n=3). Different letters are significantly different for P ≤ 0.05 between varieties. The difference between any two values, followed by at least one common letter, is insignificant. MAL = Maximum allowable limit.

Table 4

The content of heavy metals in soil from the Săsar (S) area (mg/kg)

Depth (cm)	*Location*	*Cd*	*Pb*	*Zn*	*Co*
		*MAL**	*MAL**	*MAL**	*MAL**
		1 mg/kg	*20 mg/kg*	*100 mg/kg*	*15 mg/kg*
0-20	Săsar (S)	5.64±0.32 a	640.02±13.80 c	1305.34±17.28 b	5.83±0.16 a
20-40		2.71±0.23 b	744.65±33.10 ab	1271.51±21.24 b	6.63±0.14 a
40-60		2.15±0.23 bc	732.53±10.90 ab	1773.73±36.78 a	5.33±0.30 a
60-80		1.02±0.07 c	789.40±1.74 a	1688.73±69.81 a	5.37±0.33 a
Average		2.88±0.21	726.65±14.88	1509.83±36.28	5.79±0.23
Minimum values		1.02±0.07	640.02±13.80	1271.51±21.24	5.33±0.30
Maximum values		5.64±0.32	789.40±1.74	1688.73±69.81	6.63±0.14
F (Fisher Factor)		24.714	3.725	12.758	2.023
Sig.		p ≤ 0.000	p = 0.061	p = 0.002	p = 0.189

Average value ± standard deviation (n=3). Different letters are significantly different for P ≤ 0.05 between varieties. The difference between any two values, followed by at least one common letter, is insignificant. MAL = Maximum allowable limit.

The content of heavy metals in Center (C) area. In the Center (C) area the characteristic type of soil is luvosols (LV) class. The soil is characterized by a pH of 6.3-7.1, the humus content varies between 1-4%, base saturation level of 80-100% (Mihali et al 2013). Redox potential between 326-358 (mV) and electronic conductivity between 173.4-196.0 (μS/cm) (Bora el al 2013).

In this area the highest concentration of Cd was found at 40-60 cm (0.89 ± 0.01 mg/kg) and 60-80 cm (0.91 ± 0.02 mg/kg), these values are equal in terms of statistics, the lowest concentrations was found in the surface of the soil 0-20 cm (0.36 ± 0.09 mg/kg) and 20-40 cm (0.35 ± 0.03 mg/kg). Pb has reached high values of concentration in the surface of the soil 0-20 cm (130.51±6.13 mg/kg), but also in depth of the soil profile 40-60 m (136.57 ± 8.56 mg/kg) and 60-80 cm (135.89 ± 8.02 mg/kg), as well these values are equal in terms of statistics. For Zn concentration we found statistically equal values. Co has the highest concentration at surface of the soil 0-20 cm (1.49 ± 0.04 mg/kg) and the lowest concentrations was found in depth of soil profile 20-40 cm (0.59 ± 0.06 mg/kg), 40-60 cm (0.55 ± 0.01 mg/kg) and 60-80 cm (0.62 ± 0.06 mg/kg) these values are equal in terms of statistics (Table 5).

Concentration values of heavy metals exceeded the MAL imposed by law 0.91±0.02 mg/kg Cd (1 mg/kg MAL); 135.89±8.02 mg/kg Pb (20 mg/kg MAL); 167.05±1.05 mg/kg Zn (100 mg/kg-MAL). In case of Co concentration (1.49±0.04 mg/kg) the value is below the MAL (15 mg/kg).

The differences between variants were statistically assured ($F = 10.206$; $p = 0.004$ for Cd); ($F = 12.334$; $p = 0.002$ for Pb); ($F = 1.217$; $p = 0.365$ for Zn) and ($F = 33.441$; $p \leq 0.000$ for Co) (Table 5).

The content of heavy metals in Seini (S) area. In the Seini (S) area the characteristic type of soil is luvosols (LV) class. The soil is characterized by a pH from 5.91-6.33, the humus content varies between 2-3%, base saturation level of 80-100%. Redox potential between 326-358 (mV) and electronic conductivity between 173.4-196.0 (μS/cm) (Bora el al 2013). Due to shallow groundwater, yellowish-brown spots appear frequently after the oxidation of iron compounds (Vaum-Ivascu 2011).

Concentration values of heavy metals in this case are below of the MAL (0.19±0.02 mg/kg Cd [1 mg/kg MAL]); 73.42±1.26 Zn mg/kg [100 mg/kg-MAL] and 4.04±0.07 mg/kg-Co [15 mg/kg MAL], in case of Pb this heavy metal exceeds MAL (20.19±0.72 mg/mg [20 mg/kg MAL]). Regarding heavy metals from Seini (S) area, these concentrations are statistically equal for all analyzed heavy metal. Variants were statistically assured ($F = 2.794$; $p = 0.109$ for Cd); ($F = 1.680$; $p = 0.248$ for Pb); ($F = 0.967$; $p = 0.454$ for Zn) and ($F = 0.336$; $p = 0.800$ for Co) (Table 6).

Based on the data presented we can say that in this area the effect of heavy metal pollution is lower than in other areas we studied. One possible explication for reduced heavy metals concentration is the distance from the source of pollution.

The content of heavy metals in Tăuții Măgherăuș (TM) area. In the Tăuții Măgherăuș (TM) area the characteristic type of soil is preluvosoil (EL) class. Preluvosoil typical profile has a slightly shorter than other soil found in the complex because it meets at the southern slopes, better warm or on a microrelief with good drainage and parent material rich in alkaline elements (Bora et al 2015a).

Regarding the heavy metals concentrations from Tăuții Mărgherăuș (TM) the concentration in this case are below the MAL (0.22±0.03 mg/kg Cd [1 mg/kg MAL], 13.49±0.75 mg/kg Pb [20 mg/kg MAL], 55.68±1.57 mg/kg Zn (100 mg/kg-MAL) and 9.19±0.26 mg/kg-Co [15 mg/kg MAL]). Regarding heavy metals from Tăuții Măgherăuș (TM) area, th concentrations are statistically equal ($F = 0.436$; $p = 0.733$ for Cd), ($F = 0.757$; $p = 0.647$ for Pb), ($F = 3.479$; $p = 0.070$ for Zn) and ($F = 2.119$; $p = 0.176$ for Co).

Based on the data presented we also can say that in this area the effect of heavy metal pollution is lower than in other areas we studied. One possible explication for lower heavy metal concentration is distance from the source of pollution.

Table 5

The content of heavy metals in soil from the Center (C) area (mg/kg)

Depth (cm)	*Location*	*Cd*	*Pb*	*Zn*	*Co*
		*MAL**	*MAL**	*MAL**	*MAL**
		1 mg/kg	*20 mg/kg*	*100 mg/kg*	*15 mg/kg*
0-20	Center (C)	0.36±0.09 b	130.51±6.13 a	156.19±2.78 a	1.49±0.04 a
20-40		0.35±0.03 b	104.52±7.01 b	156.76±3.19 a	0.59±0.06 b
40-60		0.89±0.01 a	136.75±8.56 a	167.05±1.05 a	0.55±0.01 b
60-80		0.91±0.02 a	135.89±8.02 a	166.30±4.38 a	0.62±0.06 b
Average		0.69±0.04	126.92±7.43	161.57±2.85	0.81±0.04
Minimum values		0.35±0.03	104.52±7.01	156.19±2.78	0.59±0.06
Maximum values		0.91±0.02	136.75±8.56	167.05±1.05	1.49±0.04
F (Fisher Factor)		10.206	12.334	1.217	33.441
Sig.		p = 0.004	p = 0.002	p = 0.365	p ≤ 0.000

Average value ± standard deviation (n=3). Different letters are significantly different for P ≤ 0.05 between varieties. The difference between any two values, followed by at least one common letter, is insignificant. MAL = Maximum limit allowable limit.

Table 6

The content of heavy metals in soil from the Seini (S) area (mg/kg)

Depth (cm)	*Location*	*Cd*	*Pb*	*Zn*	*Co*
		*MAL**	*MAL**	*MAL**	*MAL**
		1 mg/kg	*20 mg/kg*	*100 mg/kg*	*15 mg/kg*
0-20	Seini (S)	0.24±0.03 a	17.96±0.50 a	70.82±1.25 a	4.05±0.15 a
20-40		0.16±0.01 a	21.70±0.94a	76.53±1.14 a	4.00±0.09 a
40-60		0.25±0.03 a	21.26±0.94 a	73.92±0.40 a	4.16±0.01 a
60-80		0.13±0.01a	19.82±0.50 a	72.41±2.26 a	3.95±0.02 a
Average		0.19±0.02	20.19±0.72	73.42±1.26	4.04±0.07
Minimum values		0.13±0.01	17.96±0.50	70.82±1.25	3.95±0.02
Maximum values		0.25±0.03	21.70±0.94	76.53±1.14	4.16±0.01
F (Fisher Factor)		2.794	1.680	0.967	0.336
Sig.		p = 0.109	p = 0.248	p = 0.454	p = 0.800

Average value ± standard deviation (n=3). Different letters are significantly different for P ≤ 0.05 between varieties. The difference between any two values, followed by at least one common letter, is insignificant. MAL = Maximum allowable limit.

Table 7

The content of heavy metals in soil from the Tăuții Măgherăuș (TM) area (mg/kg)

Depth (cm)	*Location*	*Cd*	*Pb*	*Zn*	*Co*
		*MAL**	*MAL**	*MAL**	*MAL**
		1 mg/kg	*20 mg/kg*	*100 mg/kg*	*15 mg/kg*
0-20	Tăuții Măgherăuș (TM)	0.19±0.03 a	12.12±0.63 a	57.90±2.95 ab	8.69±0.30 a
20-40		0.24±0.03 a	13.97±0.69 a	62.48±1.16 a	9.79±0.04 a
40-60		0.20±0.02 a	14.43±0.66 a	52.78±1.01 ab	9.85±0.41 a
60-80		0.25±0.02 a	13.42±1.01 a	49.58±1.14 b	8.43±0.29 a
Average		0.22±0.03	13.49±0.75	55.68±1.57	9.19±0.26
Minimum values		0.19±0.03	12.12±0.63	57.90±2.95	8.43±0.29
Maximum values		0.25±0.02	14.43±0.66	62.48±1.16	9.79±0.04
F (Fisher Factor)		0.436	0.757	3.479	2.119
Sig.		p = 0.733	p = 0.647	p = 0.070	p = 0.176

Average value ± standard deviation (n=3). Different letters are significantly different for $P \leq 0.05$ between varieties. The difference between any two values, followed by at least one common letter, is insignificant.
MAL = Maximum allowable limit.

Pearson correlation coefficients of heavy metals in soil. Person's correlation analysis was conducted in a way that the relationships between heavy metals from soil to be revealed. For this study, the four elements with discreet values cross all samples evaluated were utilized for correlation analysis (Table 8). In evaluation of the soils in Zlatna, Romania, were found many strong relationships between heavy metals, but they stopped short of considering interactions of pollutant metals and natural soil elements (Weindorf et al 2013). Paulette et al (2015) in her research has found many strong relations between metals from Zlatna soil Mn & V (0.988), Fe & Mn (0.982), Fe & V (0.970), Cu & Zn (0.964), K & Rb (0.925), Sr & V (0.925), Mn & Sr (0.919) and Cu & Fe (0.905).

From the data presented in Table 8 is observed that also in this case was obtained many strong relationships between heavy metals from soil (Pb & Cd 0.977, Zn & Cd 0.693, Zn & Pb 0.787, Co & Cd 0.809, Co & Pb 0.806, Co & Zn 0.539). These relationships are important when considering how pollutant fate, uptake and biochemical reactions are impacted by local pedological conditions.

For example consider that many pollutants exist as cationic species which may compete for and in many instances occupy, electrostatically charged exchanges sites on colloidal clays (Paulette et al 2015).

Table 8

Pearson correlation coefficients of heavy metals in soil*

	Cd	*Pb*	*Zn*	*Co*
Cd	*1.000*	0.977**	0.693**	0.809**
	p = ...	p ≤ 0.000	p ≤ 0.000	p ≤ 0.000
Pb	0.977**	*1.000*	p = ...	0.806**
	p ≤ 0.000	*p = ...*	0.787**	p ≤ 0.000
Zn	0.693**	0.787**	*1.000*	0.539**
	p ≤ 0.000	p ≤ 0.000	*p = ...*	p ≤ 0.000
Co	0.809**	0.806**	0.539**	*1.000*
	p ≤ 0.000	p ≤ 0.000	p ≤ 0.000	*p = ...*

*significant correlation for $p < 0.05$, at the 95% confidence level; N = 60.

Based on Person's correlation presented in Table 8 we can also observe that the concentration of a heavy metal can be affected by an another heavy metal concentration (Pb & Cd 0.977, Zn & Cd 0.693, Zn & Pb 0.787, Co & Cd 0.809, Co & Pb 0.806, Co & Zn 0.539).

Conclusions. In accordance with the specified limits, the content of heavy metals (Cd, Pb, Zn, Co) in total forms proving their high concentrations in soils in all the areas studied. The exception is Seini (S) and Tăuții Mărgherăuș (TM), areas were the concentrations of heavy metals was below of the LMA. These high concentrations could be an effect of pollution from tow metallurgical factories in Baia Mare, Romplumb and Cuprom which are the main sources of pollution from the city and also for the neighboring agricultural areas. The areas studied near the source of pollution have greatest heavy metals concentrations compared with Seini (S) and Tăuții Măgherăuș (TM) areas which are at considerable distance from the source of pollution. The highest concentration was found throughout the soil profile in all areas studied. Person's correlation analysis revealed a number of strong inter-elemental relationships. These strong relationships offer critical insight into fate, transport and biogeochemical cycling of contaminants with respect to natural soil elements which often have similar electrostatic attraction to soil colloids.

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