

## THE SOILS QUALITY FROM THE SOUTHERN – EASTERN PART OF BAIA MARE ZONE AFFECTED BY METALLURGICAL INDUSTRY

**Gheorghe DAMIAN, Floarea DAMIAN, Daniel NĂSUI, Corina POP & Cornel PRICOP**

*North University of Baia Mare, 62A Dr. Victor Babeş Street, 430083 Baia Mare, Romania,  
e-mail: damgeo@ubm.ro*

**Abstract:** The studied area is placed south of the copper plant Baia Mare and was under the impact of gaseous emissions for many years. Heavy metals pollution and landslide caused by pedogenetic conditions were the factors that led to the limitation of land use for agriculture. Based on the physical and chemical characteristics and soil composition profile there were identified these types of soil: eutricambosol, typical luvisol, stagnic, gleyic luvisol and aluviosols. Lead concentration value is between 32-165ppm with high frequency values exceeding 100 ppm, the allowable maximum limit (100 ppm) for lead is exceeded by 1.14 – 1.65 times. Lead concentrations increase in close proximity of copper plant (425-995ppm). Copper concentrations are between 28-118ppm and at present the maximum frequency is below the allowable maximum values within the territory of Groşi locality for all the soils type. Copper concentrations between 54-750ppm, with the value frequency of over 100 ppm, are representative of soils in the area adjacent to the metallurgical plant. On the Groşi territory locality the Cu show concentration elevated above background. Zinc concentrations within the Groşi locality are between 89-308ppm. Zinc concentrations between 252-1325ppm are greater in the region adjacent to Baia Mare metallurgical plant. Cadmium concentrations do not exceed the allowable maximum value in the area of Groşi locality (0.7ppm) but in the area nearby metallurgical plant the excessive concentrations (1.20-9.44ppm) exceed the alert and intervention thresholds. Determinations by X-ray diffraction have emphasized the existence of montmorillonite and allophane in the clay fraction associated with hidromuscovite, feldspar and quartz. Low note of bonitate are due to gleyization and pseudogleyization and pollution from human activities and in particular the metallurgical industry. If used as grassland, the grades levels are between 58-81 and fall within the class III of bonitate and I class.

Key words: heavy metal, soil pollution, bonitation of soil, type of soil.

### 1. INTRODUCTION

High levels of heavy metals caused by atmospheric deposition, around Baia Mare metallurgical plants are found in soil and plants, (Lăcătuşu et al., 1996, Culicov et al., 2000, Lăcătuşu & Lăcătuşu 2008, Lăcătuşu et al., 2008, Damian et al., 2008a, Suciuc et al., 2008, Lăcătuşu et al., 2009).

Soil contamination with heavy metals is expanding both in surface as well as in depth up to 1.20 meters in the industrial areas of Baia Mare, (Damian et al., 2008b). Heavy metal pollution that affects urban soils, directly and indirectly affects human health, (Grzebisz et al., 2002; Gazdag & Sipter 2008). The heavy metal concentration in soil depends of their migration forms and the

contribution of these forms to the total concentration, (Shubina & Kolesov 2002). Also the potential mobility of heavy metal in soil depends of soil and the metal specie, (Šichorová et al., 2004).

This paper aims to achieve a fair presentation of the current soil quality affected by the metallurgical industry, from the southern part of Baia Mare Cuprom zone, in the Groşi locality.

### 2. MATERIALS AND METHODS

#### 2.1. Area studied location

The area selected for the study of heavy metal pollution of soils corresponds with the land used for agriculture, orchards and pasture and was compared

with the industrial area near the copper metallurgical plant. Differences of concentration of heavy metals in soils between urban and rural zone have been reported by Secu et al., (2008).

Central zone of the studied area is located 5 km south of Baia Mare, and overrides locality Groși territory, (Fig. 1), the total area of the village is 2,353 ha. The geographic coordinates of the village are 47°36'lat. N and 23°36'long. E.

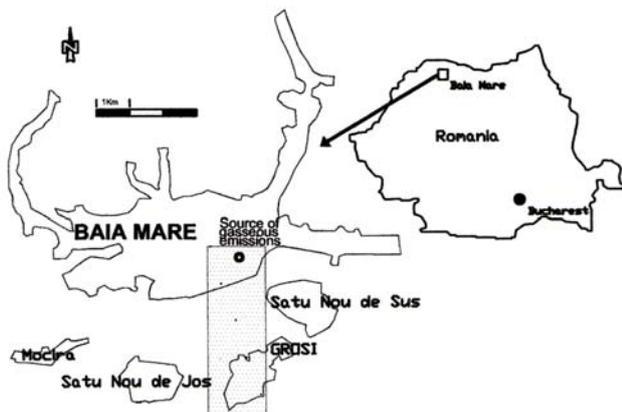


Figure 1. Location of the study area

Many agricultural areas can not be used due to severe soil erosion. Orchards represent 7% of total agricultural land. The land use in the studied area is shown in figure 2.

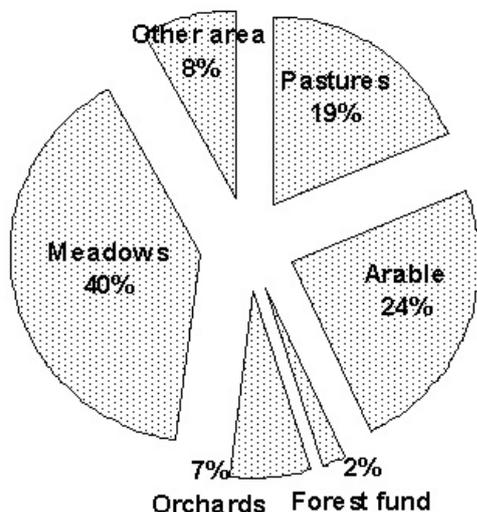


Figure 2. Land use distribution areas.

Landslide areas from Groși locality are distributed in the south-west area. They are the result of exogenous modelling, the occurrence of which is conditioned by several factors: gravity, land constitution, land slope, the weight of the masses, wetness. Landslides fall in land movements caused by the water saturation of rock and soil mass.

## 2.2. Description of the soil types

According to the Romanian Soil Taxonomic Classification, (Florea & Munteanu 2003) in the investigated area there were found these types of soils: eutricambosol, typical luvisol, stagnic luvisol, gleyic luvisol and aluviosols.

**Eutricambosols.** Vegetation characteristic of these soils is represented by forests partly replaced by pastures and meadows. Eutricambosols are moderately acidic with a slightly difference on profile. Humus content is relatively high in the organic horizon (2.76 – 4.44%).

**Luvisols** are represented by subtypes: typical, stagnic and gleyic luvisol types. They appear on small areas near metallurgical plant and are prevalent in the area investigated in the southern extension. These soils are developed on the low plains and poorly drained terrains.

**Typical luvisol** is present on large areas in the studied region, being covered by orchards and grasslands. The Ao horizon has grey colour. The colours of the Bt horizon vary from brown to red. Soil profile is as follows: Ao-Bt-C.

**Stagnic and gleyic luvisol types** are poor in humus and nutrients and has low natural fertility being covered by natural grasslands. Soil profile is as follows: Ao-Ea(EI)-E\B-Bt-C. The Ao horizon is 15 cm thick, the brown-grey colour indicating a low content of humus. The structure is granular and the texture ranging from clay loamy to clay.

**Aluviosols** are present only in the western proximity of metallurgical plant. It consists of an Ao horizon 40cm thick, which stay on top of C horizon of alluvial deposits.

Soil sampling took into account the distance from pollution sources, the position in relation with the dominant wind direction and separate representative soil types. Soil sampling was done in the first 0-20 cm, distributed over the whole surface area of Groși locality extending both to Cuprom metallurgical smelter as well as in areas where the soil has different usage: agricultural land, grassland, orchards. Soil samples were dried at a temperature of 100–105°C, sieved through a stainless sieve with diameter of 2 mm and homogenized. Analysis of soil samples was done using the analysis method of flame atomic absorption spectrometry (AAS) with the Sollar S4 type.

X-ray diffraction analyses were used to determine the species of clay minerals from soil samples that were separated by decantation method. X-ray diffraction analyses were performed with Philips diffractometer, Muller Type, PW 1352/10 registration system at the North University Baia Mare.

Table 1. Physical and chemical properties of the soils

Type of soil/sample	pH	Humus %	N total %	CEC meq/100gsoil	Sand	Clay		Soil texture
						%		
BM1Gleyic Luvisol	4.16	4.14	0.18	18.14	22.9	45.3	31.8	Clay loam
BM2 Aluvisol	4.98	4.08	0.21	15.67	33.1	38.7	28.2	Clay loam
BM3 Typical Luvisol	5.49	4.80	0.23	15.87	26.2	43.2	30.6	Clay loam
BM4StagnicLuvisol	4.20	3.90	0.21	25.03	25.4	41.8	32.9	Clay loam
BM5 Typical Luvisol	3.54	3.66	0.11	17.78	28.8	44.9	26.3	Loam
BM6 Typical Luvisol	4.50	3.43	0.12	21.27	24.8	43.1	32.1	Clay loam
BM7 Typical Luvisol	4.40	5.44	0.19	19.33	34.9	42.1	33.0	Loam
BM8 Typical Luvisol	4.70	3.14	0.22	16.73	35.0	42.1	22.9	Loam
BM9 Typical Luvisol	4.35	4.26	0.18	20.29	29.1	43.5	28.4	Clay loam
BM10 Typical Luvisol	4.50	4.97	0.12	15.99	29.7	45.3	25.0	Loam
G Eutricambosol P10	5.80	3.31	0.16	22,01	32.0	34.0	34.0	Clay loam
G Eutricambosol P14	8.10	4.30	0.21	99.00	34.0	21.0	45.0	Clay
G Typical Luvisol P7	5.20	3.10	0.15	19.70	21.0	44.0	35.0	Clay loam
G Typical Luvisol P3	6.30	4.90	0.24	49.03	27.0	34.0	39.0	Clay loam
G Stagnic Luvisol P8	5.30	4.30	0.21	20.02	26.0	30.0	44.0	Clay
G Gleyic Luvisol P11	4.15	4.02	0.19	18.16	22.0	40.0	38.0	Clay loam

BM1-Baia Mare soils; G Luvisol -Grosi soils; P3-Number of soil sample; N total- nitrogen content; CEC – cation exchange capacity Develop methodology of pedological studies, Ecopedological Parameters, third part 1987, Research Institute for Soil Science and Agrochemistry, Bucharest, Romania

### 3. RESULTS AND DISCUSSION

#### 3.1. Physical and chemical properties of soils

Physical and chemical properties of studied soils, with a particular importance in retaining heavy metals pollutants are presented in table 1. Comparative analyses of land close to the metallurgical plant area and the land from the southern part resulted in significant differences in physical-chemical properties. The pH value ranges from strongly acidic (3.54) to moderate acid (5.49) for luvisol subtypes from the vicinity of Copper plant. The pH of luvisol type from the extended area of Grosi locality correspond to the reaction class of moderately acid to weak acid, (Develop methodology of pedological studies, Ecopedological Parameters, third part 1987).

Eutricambosols from the studied area presents pH values falling within the classes of reaction moderately acid – weak alkaline. Texture varies from clayey for Eutricambosols to loamy clayey for luvisol types. Humus content varies between 3.1–4.9% for surface soil layers. Total nitrogen content is low in the studied soils and is representative for small and medium content classes (Develop methodology of pedological studies, Ecopedological Parameters, third part 1987).

Ecopedological Parameters, third part 1987). Total cation exchange capacity of the studied soils varies with the type of soil (Tab. 1).

Fine clay fraction of luvisols, stagnic luvisols contains big quantities of clay. Clay minerals influence the amount of soil cation exchange capacity along other components, (Stewart & Hossner 2001). Clay minerals, oxides and organic matter are the most important components for the sorption of heavy metals, (Plesničar & Zupančič 2005). Clay is most represented by montmorillonite. Retention of heavy metals from contaminated soils by clay minerals is due to their exchange surface, (Li & Li 2000). The presence of clay minerals in the soil, along with other components, influences retention of heavy metals by soil, (Holm et al., 2003).

X-ray diffraction spectrum (Fig. 3) indicates the presence of poorly crystallized clays smectites. In addition to montmorillonite there is a considerable amount of amorphous smectite represented by allophane.

The muscovite was determined associated with these components. Low intensity 9.905 line reveals the presence of degraded micas represented by hidromuscovite or illite. There also appear small amounts of feldspars. The smectites are formed mostly from exogenous alteration of feldspar or of muscovite. Quartz grains appear in the clay fraction.

#### 3.2. Soil pollution by heavy metals

The analyzed soils have textured features (fine clay fraction prevalence and silts) and compositional (in terms of high humus content) that favours the retention of heavy metals.

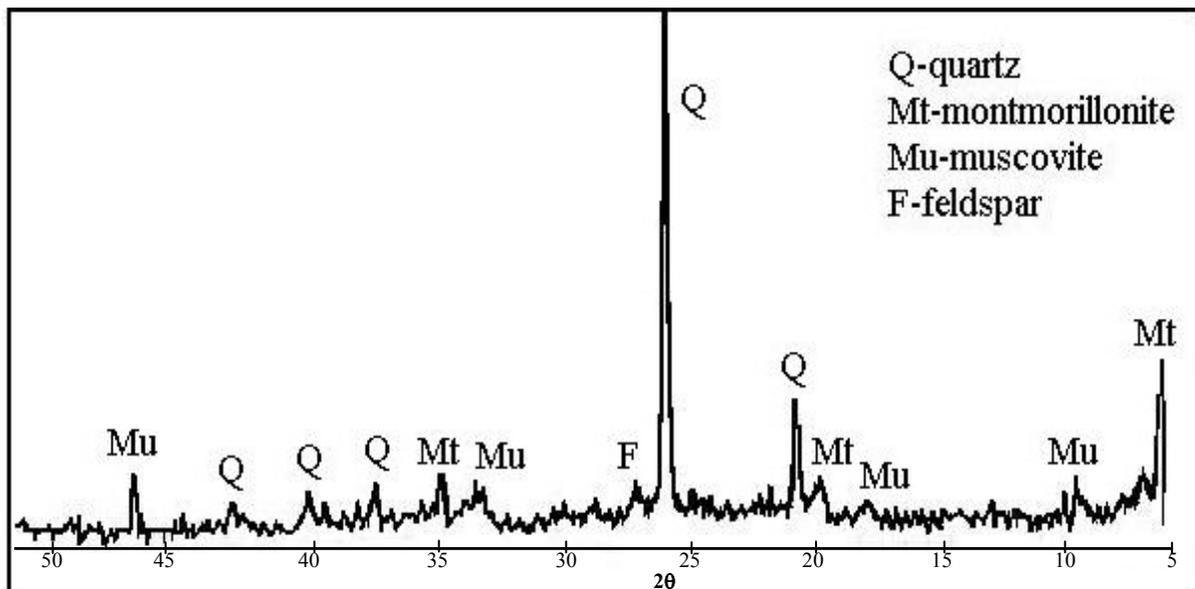


Figure 3. X-ray diffraction spectrum of clay fraction in the eutricambosol type.

Table 2. The geographical position of soil samples and the total contents of the heavy metals

Samples	X	Y	Zn ppm	Cu ppm	Pb ppm	Cd ppm
BM1 Gleyic Luvisol	695699	5280700	366	187	939	1.20
BM2 Aluvisol	695078	5281689	434	90.9	952	9.44
BM3 Typic Luvisol	695554	5280729	536	199	904	5.85
BM4 Stagnic Luvisol	696034	5280955	252	190	995	4.40
BM5 Typic Luvisol	696010	5281248	275	149	977	1.55
BM6 Typic Luvisol	695972	5280813	1325	105	630	8.01
BM7 Typic Luvisol	695956	5280828	402	90	690	6.05
BM8 Typic Luvisol	695992	5280775	375	750	596	4.80
BM9 Typic Luvisol	695980	5280859	425	67	635	5.20
BM10 Typic Luvisol	696026	5280753	390	54	425	7.50
G Eutricambosol P1	696492	5277945	201	50	123	<0,7
G Eutricambosol P2	696753	5278109	131	46	125	<0,7
G Typic Luvisol P3	697026	5277700	308	53	143	<0,7
G Typic Luvisol P4	697221	5277250	185	51	114	<0,7
G Typic Luvisol P5	697123	5277236	89	28	32	<0,7
G-Gleyic Luvisol P6	695937	5278420	237	83	123	<0,7
G Typic Luvisol P7	695502	5278724	196	82	154	<0,7
G Stagnic Luvisol P8	695577	5278756	155	46	132	<0,7
G Typic Luvisol P9	695662	5278235	124	54	132	<0,7
G Eutricambosol P10	695433	5277621	284	118	165	<0,7
G Gleyic Luvisol P11	695733	5277177	126	43	68	<0,7
G Eutricambosol P12	696496	5276428	115	38	40	<0,7
G Stagnic Luvisol 13	696236	5276517	125	33	63	<0,7
G Eutricambosol P14	695834	5276466	124	35	77	<0,7

Order no. 756/1997 on the assessment of environmental pollution, by the Ministry of Waters, Forests and Environmental Protection, published in the Official Monitor in 1997.

In the investigated area soils located on slopes are less polluted than those on flat terrain and micro valleys. Retention of heavy metals in soils is due to both organic and inorganic soil components and its physical and chemical properties. Heavy metals are mobilized in soil by physical, chemical and biological processes, (Bradl et al., 2005).

Accumulation of metals in soil depends on the pollutant source position in relation to prevailing winds in the area. It is noted that the prevailing wind direction is E-W and NW-SE corresponding to the central geographical position in which Groși locality is placed (Fig. 4). However, the average annual rate of wind movements from west and north-east is 30%

and can explain the relatively low intake of gaseous emissions and so a low level of pollution for the investigated soils.

Soil samples were chemically analyzed for the content of heavy metals: Zn, Cu, Pb, Cd. The geographical location and the content in heavy metals of the studied area are presented in table 2. The analysis results of heavy metal concentrations (Pb, Zn, Cu, Cd) were compared with reference values of the Order no. 756/1997.

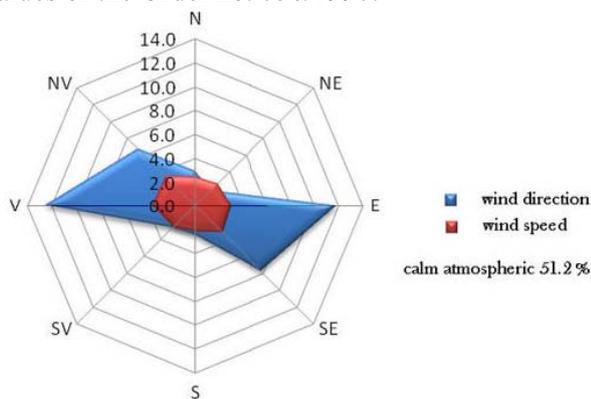


Figure 4. Annual compass rose of frequency and wind speed.

High concentrations of lead in the humus-rich horizons correlate with moderately acid pH. When pH is neutral or slightly alkaline the high concentrations of lead could be explained by complexation of lead by organic matter, (Bradl et al., 2005) from samples rich in humus, (Tab. 1). All examined samples have higher content than normal values and exceed allowable maximum level for lead. Allowable maximum values (100 ppm) for lead is exceeded by 1.14 – 1.65 times.

The concentrations of zinc exceed the alert threshold only once, the rest being above normal values but within allowable maximum limit. Copper is polluting especially luvisol type and for eutricambosol it appears within normal limits. Cadmium concentrations do not exceed the allowable maximum values.

### 3.3 Spatial distribution of pollution

A GIS interpolation method was used to draw the distribution of heavy metal concentrations using our soil samples. The measured values were correlated with each of the field measured coordinates, recorded in Microsoft Excel and saved in a .dbf format. The software used is ArcGIS 9.2, and points were introduced in a 'Shapefile' file by placing the coordinates for each point. Then, each point received its associated values in tabular data (.dbf) by import. The chosen interpolation method is

IDW (inverse distance weighted). The principle of this technique is based on the theory that close points are more similar (homogeneous) than the ones further apart. For this purpose the IDW method doubles the importance assigned to closed point values. The interpolation results were superimposed on a SRTM map (Shuttle Radar Topography Mission – 2002) that covers the land topography including the height of ground objects and the Lăpuș catchment river network map.

Higher concentrations of the heavy metals analyzed are directly related with adjacent areas of metallurgical plant Cuprom. In general, heavy metal concentration decreases when distance from the main source of pollution increases (Hasselbacha et al., 2005).

High concentrations of Pb, Zn, Cd, and Cu are associated with Ao soil horizon. The influence of atmospheric conditions on the variation of heavy metals concentrations is highlighted particularly in the south area of the Groși locality. The heavy metals distribution in upper soil horizon of the investigated area is influenced by prevailing air currents, which disperse the air emissions from the copper and lead metallurgical activity.

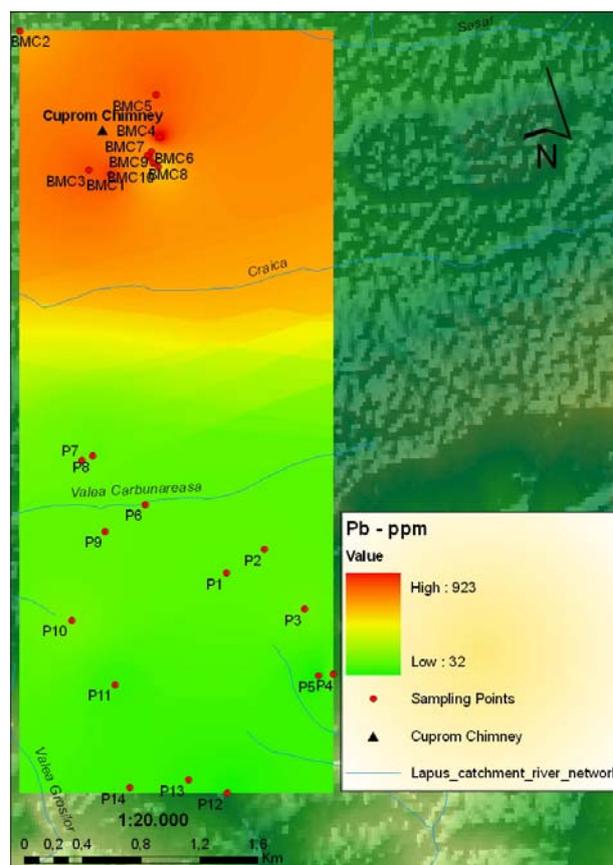


Figure 5. Lead distribution in the upper soils horizon

Lead distribution in the upper soils horizon (Fig. 5) is characterized by decreasing

concentrations with distance from the main pollution sources.

Total Cu concentration values (Fig. 6) exceed normal limits and allowable maximum values on areas that are expanding in Groși locality. Overall we can state a reduced copper pollution of the soil.

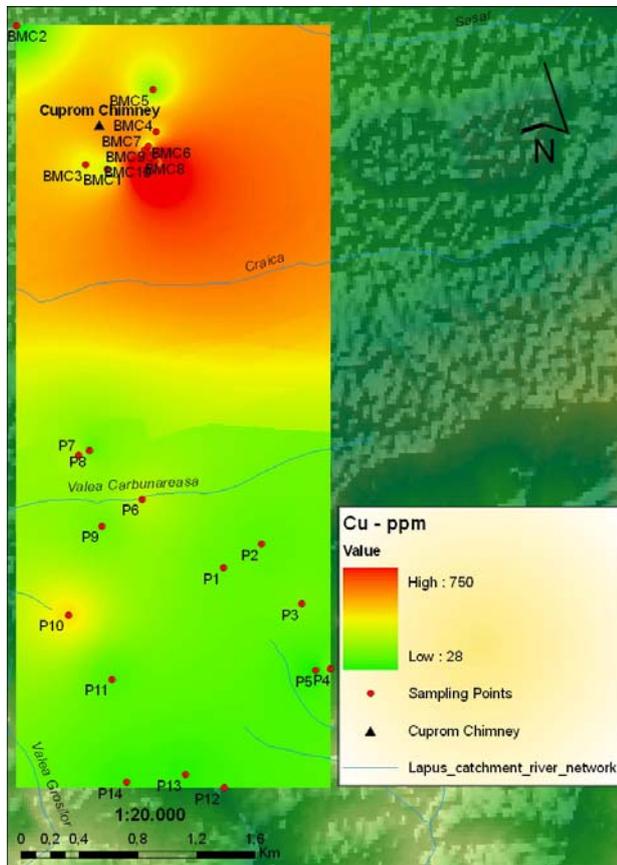


Figure 6. Copper distribution in the upper soils horizon

The Zn contamination level (Fig. 7) is high in adjacent areas to Cuprom plant, Zn pollution decreases when the distance from the source increases. The image of zinc distribution in the top horizon of soil emphasizes two points with small surfaces where the total contents exceed the intervention threshold. This area overlaps with areas where the movement of currents come in contact with the natural barrier (the forest), which is correlated with frequency of 20% of wind direction to SW. Relatively small concentrations of Zn in the upper soil horizon is due to dispersion ability of this metal, confirmed by concentrations that exceed the intervention threshold for the lower horizons of soil profiles in the industrial area, (Damian et al., 2008b).

Total concentrations ranging between alert and intervention threshold for Cd (Fig. 8) are distributed over wider areas in the industrial region. It can be appreciated that heavy metal pollution is moderate compared with the adjacent area of Cuprom plant (with an excessive pollution).

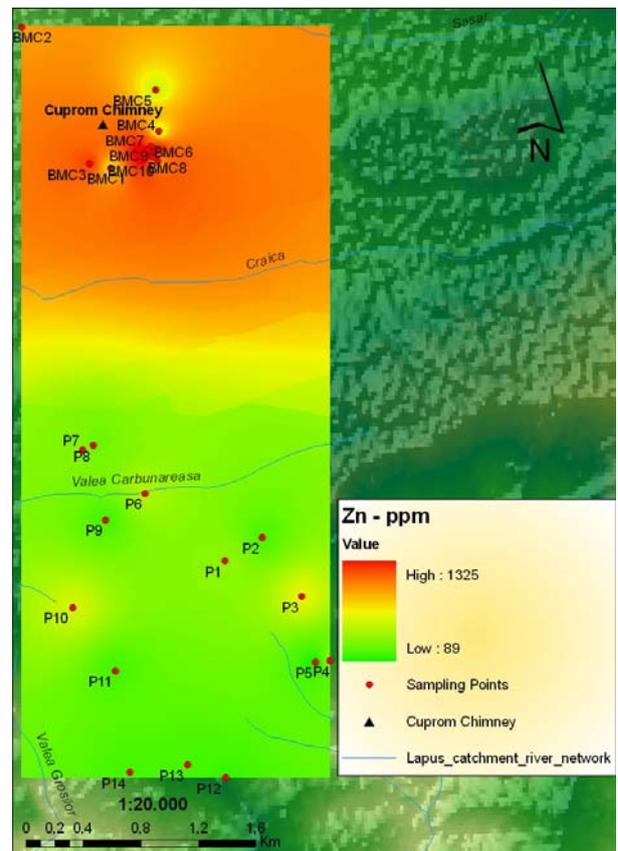


Figure 7. Zinc distribution in the upper soils horizon

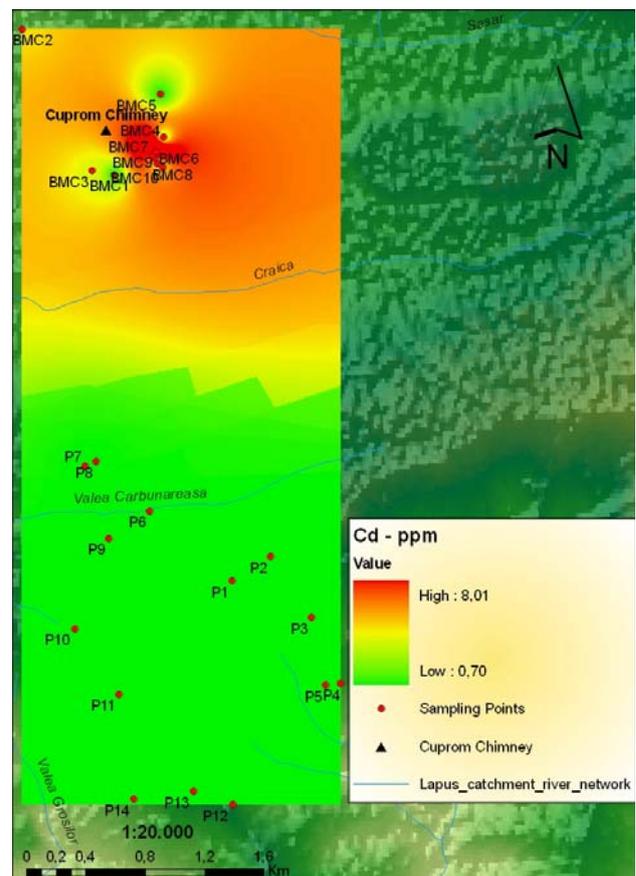


Figure 8. Cadmium distribution in the upper soils horizon

These results are contrary to our expectations with regard to geographical location and proximity to the location of the gas dispersion source. We appreciate that the specific atmospheric conditions from Baia Mare Depression and in particular the reduced frequency of winds around 30% to Groși area associated with the effect of higher dispersion of the Cuprom's gas chimney helped to avoid depositing the heavy metals on soil surface.

#### 4. THE BONITATION OF THE GROȘI SOILS

The soil quality is influenced by the soil properties, (Kiryushin 2007). For the notes of bonitation calculation, there have been used the next indicators: annual medium temperature, annual medium precipitations, soils agrochemical properties, the slope and landslides, heavy metals pollution degree, the depth of the underground water, (about Develop methodology of pedological studies, Interpreting soil surveys for different purposes, second part 1987).

Most soils fall within typical luvisol, stagnic and gleyic luvisol, subordinately there are present eutricambosols. Each of these soils was quality assessed. These soils are used mostly as arable land or grassland. Only few plots are used for orchards.

To calculate the soil bonitate class, an important role has the landslides from central and south-west of the studied area. These slides are of subsidence type and occurred due to the slope base erosion when there are breaking and falling vertical layers. Their weight creates a thrust on gleyic luvisols and encourages formation of downstream sliding in steps. In these soils slippage occurs because of a strongly wetted soil layer. These slides are extended on large areas forming a micro relief in

steps and transverse cracks.

The notes of bonitate for stagnic and gleyic luvisols range from 8-30, which fall in class IV and V of quality (Tab. 3). Small notes of bonitate are due to gleyization and pseudogleyization and pollution from human activities and in particular the steel industry. If they are used as grassland, the notes of bonitate range from 58-81 as they fall within the class III and first class of bonitate. From this point of view, soils should be used only for grassland.

Typically luvisol is used mainly as grassland. For grassland the note of bonitate is 81, which falls in the first class, indicating a good degree of use of these soils, (Tab. 3). For land used as arable land the note of bonitate is 34, comprising soils in the fourth grade. If they had been used to grow wheat the note of bonitate would be 45 which correspond to class III. Small notes of bonitate are primarily due to pollution with heavy metals.

Eutricambosols are used for growing fruit trees (apple, pear, plum, cherry). For use as orchards the note of bonitate is 17 and that corresponds to the bonitate class V, (Tab. 3). For use as grassland the note of bonitate is 36, which includes it in the fourth grade. Inclusion in IV and V class is due to pollution and land sliding affecting these lands. Using these soils as grassland and orchards seems to be good.

#### 5. CONCLUZIONS

The main soil types identified in the area based on physical-chemical characteristics and the composition of the soil profile are: eutricambosol, typical luvisol, stagnic luvisol and gleyic luvisol. Analyses of heavy metal concentrations in these soils showed a wide variation, and from their spatial distribution there is observed that the highest values are in close relation with proximity of copper plant.

Table 3. The soils the notes of bonitate

Use and agriculture	Stagnic luvisol		Typical luvisol		Gleyic luvisol		Eutricambosol		
	Note	Class	Note	Class	Note	Class	Use	Note	Class
Wheat	16	V	45	III	28	IV	Apple	15	V
Barley	16	V	40	IV	28	IV	Pear tree	13	V
Maize	14	V	33	IV	30	IV	Plum tree	26	IV
Sunflower	12	V	29	IV	26	IV	Cherry tree	12	V
Potato plant	8	V	26	IV	20	V	-	-	-
White beet	10	V	29	IV	26	IV	-	-	-
Soia bean	12	V	36	IV	28	IV	-	-	-
Pea -bean	19	V	35	IV	29	IV	-	-	-
Hayfield	52	III	81	I	81	I	Hayfield	36	IV
Arable	13	V	34	IV	27	IV	Orchard	17	V

Calculated about: Develop methodology of pedological studies, Interpreting soil surveys for different purposes, second part 1987, Research Institute for Soil Science and Agrochemistry, Bucharest, Romania.

For the area situated south of metallurgical platform where soils have different uses, heavy metal concentrations exceed normal concentrations and the allowable maximum limit, especially for Pb, Zn and Cu.

In some cases, the reference limits of alert threshold and intervention are exceeded by norms, (Order no. 756/1997). Gas emissions have caused a low pollution of soils in the area. Although the dominant wind direction is E-W and NW-SE, corresponding to the geographical position of the area situated south from industrial platform, the average annual wind movements rate from west and north-east wind is 30%. These data could explain the relatively low contribution of gaseous emissions within the southern area of industrial platform that correspond to Groși locality.

All examined samples present higher content than normal values and exceed the allowable maximum levels for lead. The allowable maximum values (100 ppm) for lead is exceeded by 1.14 to 1.65 times. Copper is the heavy metal with implications for pollution of luvisol type and it is in normal limits in eutricambosols. Compared with lead and copper, cadmium and zinc have concentrations below the allowable maximum limit and can be explained by high mobility of both metals that could percolate in depth.

For farm land, the objective of the bonitation is to establish notes and classes of favourability for different agricultural crops and quality classes for their agricultural land used as: arable land, vineyards, orchards, pastures and meadows.

In establishing the class of bonitate, landslides have had a negative role in the investigated area. The small notes of bonitate are due to gleyization and pseudogleyization and pollution from human activities and in particular the steel industry.

These landslides are the type of subsidence which has occurred due to slope base erosion when the vertical layers are breaking and falling. Their weight creates a thrust of stagnic and gleyic luvisols encouraging the formation of downstream sliding in steps. In these soils, slippage occurs because of a soil layer that is strongly wetted. These slides are extended on large areas forming a micro relief in steps and transverse cracks.

Each of these soils was assessed to rank their quality. These soils are used mostly as arable land or as grassland. Only few plots are used for orchards. In the case of their use as grassland the notes of bonitate range from 58-81 as they fall within the class III of bonitate and first class. From this point of view soils should be used only for grassland.

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