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# Current status of heavy metals soil and plant pollution in Baia Mare area

## **Summary of PhD thesis**

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## CONTENTS

<b>INTRODUCTION .....</b>	<b>3</b>
OVERVIEW OF HEAVY METAL SOILS AND PLANT POLLUTION IN ROMANIA AND IN OTHER COUNTRIES.....	3
1. RESEARCHES CONDUCTED IN ROMANIA.....	3
2. RESEARCHES CONDUCTED INTERNATIONALLY .....	4
<b>THESIS OBJECTIVES.....</b>	<b>4</b>
<b>MATERIALS AND METHODS .....</b>	<b>4</b>
1. RESEARCH FIELD.....	4
2. RESEARCH LABORATORY .....	4
<b>RESULTS AND DISCUSSION .....</b>	<b>5</b>
SOIL TYPES FROM THE ANALYZED AREA.....	5
SOURCES OF HEAVY METAL POLLUTION .....	5
<b>CURRENT STATUS OF HEAVY METALS IN SOILS FROM BAI A MARE AREA .....</b>	<b>6</b>
1. CONTENT IN TOTAL FORMS (SOLUBLE AND INSOLUBLE FORMS).....	6
2. PERCENTAGE DISTRIBUTION OF TOTAL FORMS OF HEAVY METALS ON THE TWO DEPTHS .....	7
3. CONTENT IN MOBILE FORMS .....	7
4. HEAVY METALS SOIL POLLUTION IN BAI A MARE AREA COMPARED WITH ZLATNA, COPȘA MICĂ AREAS .....	9
5. CORRELATIONS OF HEAVY METALS IN SOIL.....	9
6. REACTION AND THE MACROELEMENTS CONTENT IN SOILS FROM THE BAI A MARE AREA.....	9
7. DISTRIBUTION MAPS OF HEAVY METALS.....	10
<b>HEAVY METALS TRANSLOCATION FROM SOIL IN CROPS PLANTS .....</b>	<b>10</b>
HEAVY METALS IN EDIBLE PARTS OF VEGETABLES AND PASTURE PLANTS .....	10
<b>HEAVY METALS CORRELATIONS IN SOIL-PLANT SYSTEM.....</b>	<b>11</b>
<b>THE BIOCONCENTRATION (BCF) AND THE TRANSLOCATION FACTOR (TF) .....</b>	<b>11</b>
PLANT NUTRIENT CONTENT OF THE BAI A MARE AREA .....	12
<b>METHODS FOR LIMITING POLLUTION EFFECTS OF HEAVY METALS .....</b>	<b>12</b>
<b>CONCLUSIONS ON HEAVY METALS SOIL AND PLANT POLLUTION IN BAI A MARE AREA .....</b>	<b>12</b>
<b>SELECTIVE BIBLIOGRAPHY.....</b>	<b>14</b>

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**Keywords:** heavy metals, soil, plants, SC. Romplumb SA, SC Cuprom SA, Baia Mare, transfer, translocation

## INTRODUCTION

The complexity of soil contamination with heavy metals in Baia Mare area makes this work to address an issue of our times: pollution, in order to decipher the ways that the heavy metals are taken from soil by plants. This work was performed using conventional methods of polluted soils research in the field, combined with modern laboratory investigation of the contents of heavy metals in soil and plants.

In this way, the research paper presents new data on the surface and deep soil pollution extension with heavy metals. The transfer of heavy metals from soil into plants is influenced by various factors. Their presence in plants may have negative effects on various processes: photosynthesis, respiration, perspiration, membrane permeability, affecting the whole process of plant growth. From the study purchased in this paper, has been demonstrated that the transfer of heavy metals from soil to plants is determined on the one hand, by soil characteristics, and on the other hand by the ability of different plant species to take some heavy metals and to transfer them differently from the root into the stem and leaf. Their highlight was possible by calculating the bioconcentration and translocation factors based on the analyzes conducted for two years, 2010 and 2011.

So, the heavy metal pollution severely reduced functional diversity of soil microbial community and prevents the specific ways of nutrients movement.

## OVERVIEW OF HEAVY METAL SOILS AND PLANT POLLUTION IN ROMANIA AND IN OTHER COUNTRIES

### 1. Researches conducted in Romania

Investigations regarding the role of metallic chemical elements, called micronutrients, involved in plant nutrition such as Fe, Mn, Cu, Zn, Co were initiated. In parallel, began researches regarding the distribution of heavy metals in agricultural soils of Romania (Mihailescu et al., 1986, 1987, 1980): Oltenia, central part of Moldavia, Transylvania Basin, Banat, Banato-Crisan Plain (Lăcătușu et al., 1997b) and soils of north-central part of the Romanian Plain (Lăcătușu et al., 1994).

Research has been concentrated in industrial areas related to the extraction and processing of complex sulphide ores such as those from Baia Mare, Copsa Mica, Zlatna or areas of influence of chemical plants producing fertilizers, especially phosphorus as were those of Călugărească Valley, Năvodari or Turnu Magurele, eastern part of Bucharest, in Pantelimon-

Brănești both in the period that the two industrial units have worked on the platform (Acumulatorul and Neferal) (Răuță et al., 1980) and during the period that the activity were reduced or stopped (Lăcătușu et al., 2000, 2011).

## **2. Researches conducted internationally**

On international plant there have been conducted a series of studies to highline the degree of soil pollution with heavy metals due to various anthropogenic sources, to assess the effects of pollution on plants by determining the different transfer factors from soil micronutrients in plants. An important source of information about the abundance of heavy metals in soils in Romania and Europe is the European Geochemistry Atlases (Russia, Serbia, Austria, Denmark, Germany, Poland, Lithuania, Latvia, Sweden, France, and so on). To these are added researches by different authors for countries like USA, Ethiopia and Canada.

## **THESIS OBJECTIVES**

The main goal of the research in this paper is the evaluation of soil contamination with heavy metals in Baia Mare, in 2010-2011.

The second objective is the determination of heavy metals in samples of vegetables from cultures of carrot, lettuce and parsley and interpretation of the level of contamination and food safety hazard by comparing the values obtained with the maximum limits and normal content.

The third objective is the general characterization of soil and its physical and chemical properties.

The fourth objective is to conduct a comparative study between the current state and the previous state of soil pollution with heavy metals in Baia Mare area and present situation of soil pollution with heavy metals in the country, highlighting the heavily polluted areas like Copsa Mica, Zlatna , Năvodari, Călugărească Valley, Turnu Magurele.

## **MATERIALS AND METHODS**

### **1. Research Field**

Soil sampling was done in 25 locations of the Baia Mare area, in environments, soil and vegetation influenced by two polluting units SC. Romplumb SA, SC Cuprom SA, at two depths: 0-10 cm, 20-40 cm. Investigated area was about 20 km<sup>2</sup>.

### **2. Research Laboratory**

It have been done: the analysis of total and mobile forms of heavy metals analysis, pH analysis, the amount of basic cations exchange (SB), total cation exchange capacity, base saturation degree, the amount of exchangeable hydrogen (SH), total nitrogen, amount of humus.

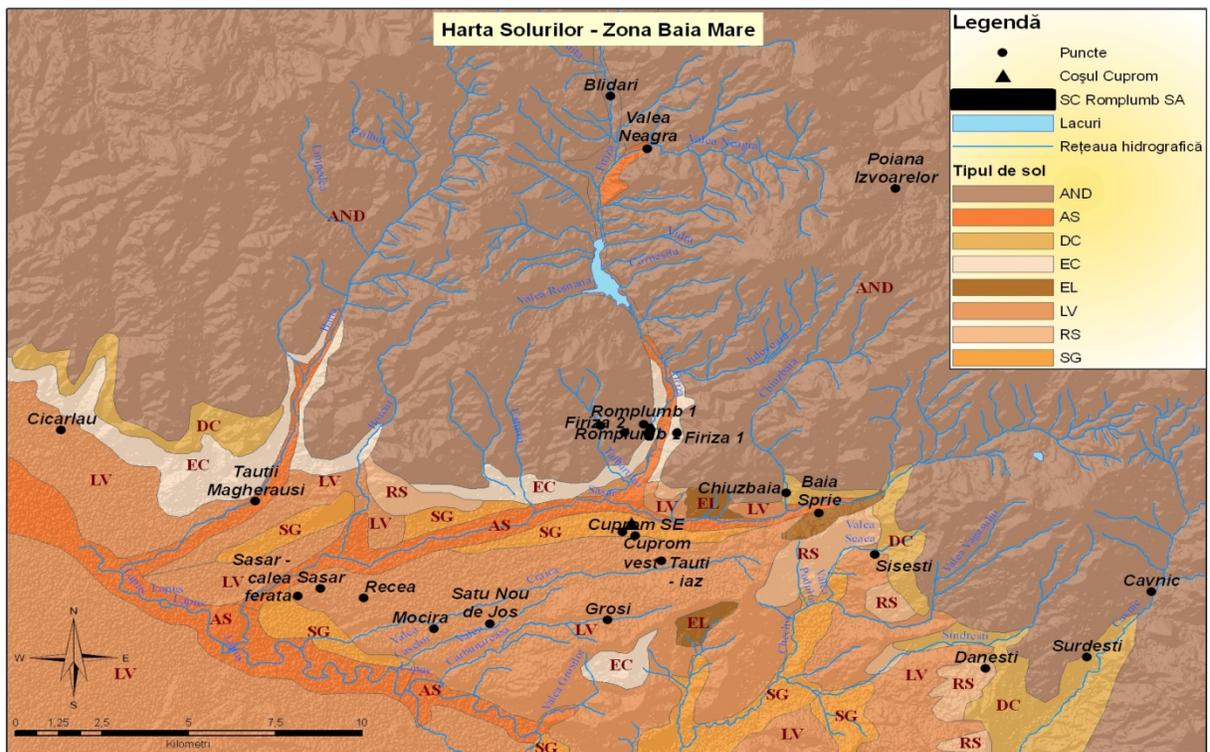
Laboratory analyzes were completed within the institution Ecole Nationale Superieure des Mines de Saint-Etienne, central SPIN General Department through international mobility obtained from the European Social Fund through POSDRU/88/1.5/S/47646 contract.

## RESULTS AND DISCUSSION

### Soil types from the analyzed area

The soil types identified in the conducted research are as follows: Luvisols, Eutricambosols, Districambosols, Aluvisols, Lithosols, Regosols, Entiantrosols, Stagnosols (fig. 1).

**Figure 1 - Map of soil sampling points in the Baia Mare**  
(with changes after the sheet of paper of Baia Mare soil map, scale 1:200.000)



### Sources of heavy metal pollution

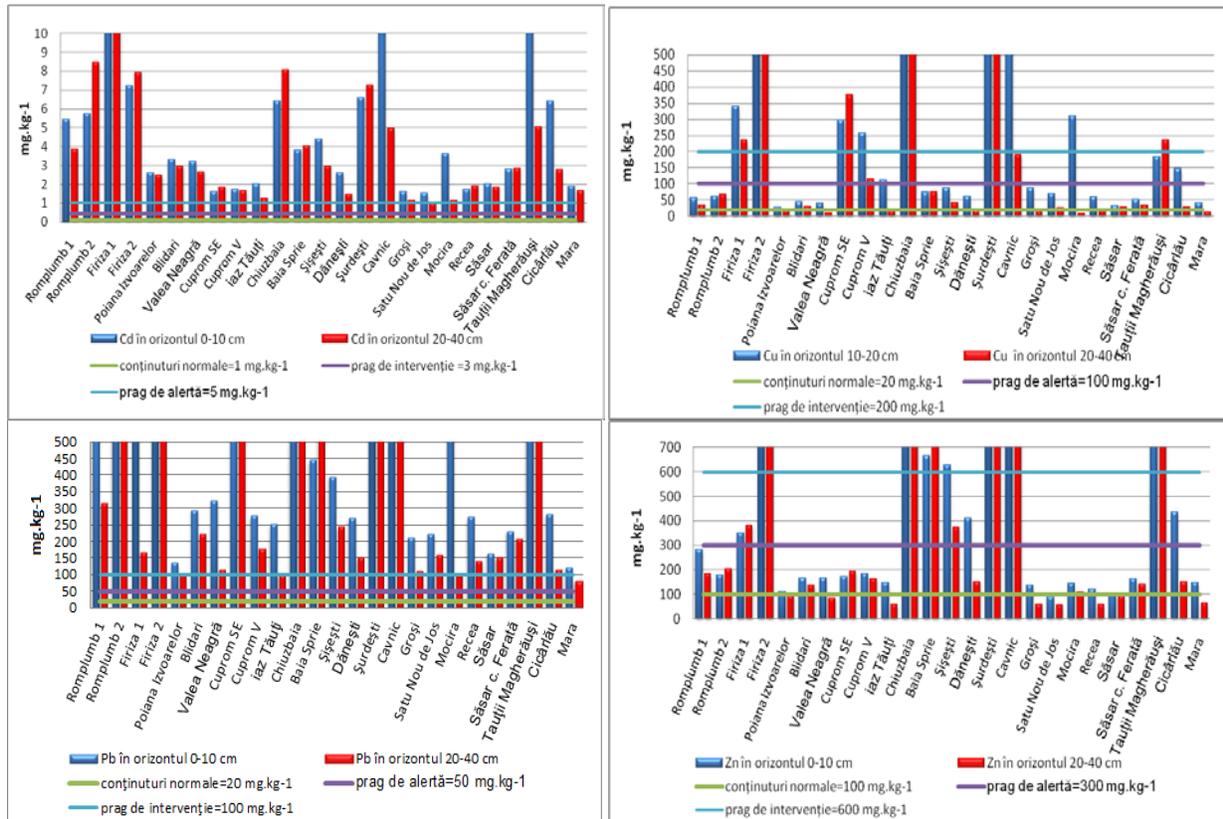
The major industrial emitters' analyzed area was represented by two non-ferrous metallurgy plants:

- SC Cuprom S.A. Bucharest - Baia Mare Branch (factory work was stopped in 2008)
- SC Romplumb S.A. – Firiza
- In 1999 joined their factory "Transgold"
- Non-ferrous ore mining units,
- Ferrous ore preparation plants.

## CURRENT STATUS OF HEAVY METALS IN SOILS FROM BAIJA MARE AREA

### 1. Content in total forms (soluble and insoluble forms)

To assess the soil pollution/contamination degree, the obtained values were compared with normal reference values and with alert and intervention thresholds for sensitive or less sensitive use of soils under Order no. 756/1997 of the Ministry of Waters, Forests and Environmental Protection (fig. 2).



**Figure 2 - Total contents of Cd, Cu, Pb, Zn compared to the normal values, the alert and the intervention threshold**

Based on the heavy metal content of the samples obtained from the two sampling depths were calculated the statistical parameters. The results are presented in Table 1:

**Table 1 - Statistical parameters of heavy metal content (mg.kg-1) determined in soil (total form)**

Total forms	Depth	Average	Mediana	X <sub>min</sub>	X <sub>max</sub>	Standard deviation	MAL
<b>Cd</b>	0-10 cm	4,80	3,30	1,52	18,08	4,02	3,00
<b>Co</b>	0-10 cm	20,47	20,14	12,62	32,90	5,24	30,00
<b>Cr</b>	0-10 cm	57,44	61,56	14,95	207,61	41,20	100,00
<b>Cu</b>	0-10 cm	350,07	85,99	26,78	2592,60	642,57	100,00
<b>Fe</b>	0-10 cm	35222,14	36589,82	15303,20	73169,60	16107,60	-
<b>Mn</b>	0-10 cm	1174,87	825,55	141,79	5884,47	1361,90	1500,00
<b>Ni</b>	0-10 cm	23,40	23,17	9,36	51,03	9,45	50,00
<b>Pb</b>	0-10 cm	998,61	323,42	122,29	5607,10	1451,78	100,00
<b>Zn</b>	0-10 cm	1187,15	177,48	89,04	8886,60	2335,91	300,00
<b>Cd</b>	20-40 cm	4,39	2,80	1,06	27,48	5,34	3,00
<b>Co</b>	20-40 cm	21,49	20,13	12,83	40,17	6,20	30,00
<b>Cr</b>	20-40 cm	54,30	60,69	11,75	113,17	29,39	100,00
<b>Cu</b>	20-40 cm	244,22	36,70	9,13	1510,10	473,04	100,00
<b>Fe</b>	20-40 cm	36352,02	32151,60	15928,83	89667,50	18732,57	-
<b>Mn</b>	20-40 cm	1045,29	890,72	113,77	5204,40	1055,87	1500,00
<b>Ni</b>	20-40 cm	23,50	24,25	8,53	55,94	10,46	50,00
<b>Pb</b>	20-40 cm	773,65	180,06	83,05	8039,90	1654,27	100,00
<b>Zn</b>	20-40 cm	1040,54	155,09	59,88	11445,00	2379,25	300,00

\*MAL= maximum allowed limit, after Kloke, 1980

## 2. Percentage distribution of total forms of heavy metals on the two depths

The interpretation of soil tests conducted in 2010 reveals that for the main indicators: cadmium, copper, lead, zinc, the distribution at the two depths shows a higher percentage belonging to the upper horizon.

## 3. Content in mobile forms

After analyzing these forms of heavy metals, there is a high mobility in soils from Baia Mare area, this being due to the acid reaction (pH often below 5) and to the higher exchangeable aluminum content, leading to microorganisms inhibition.

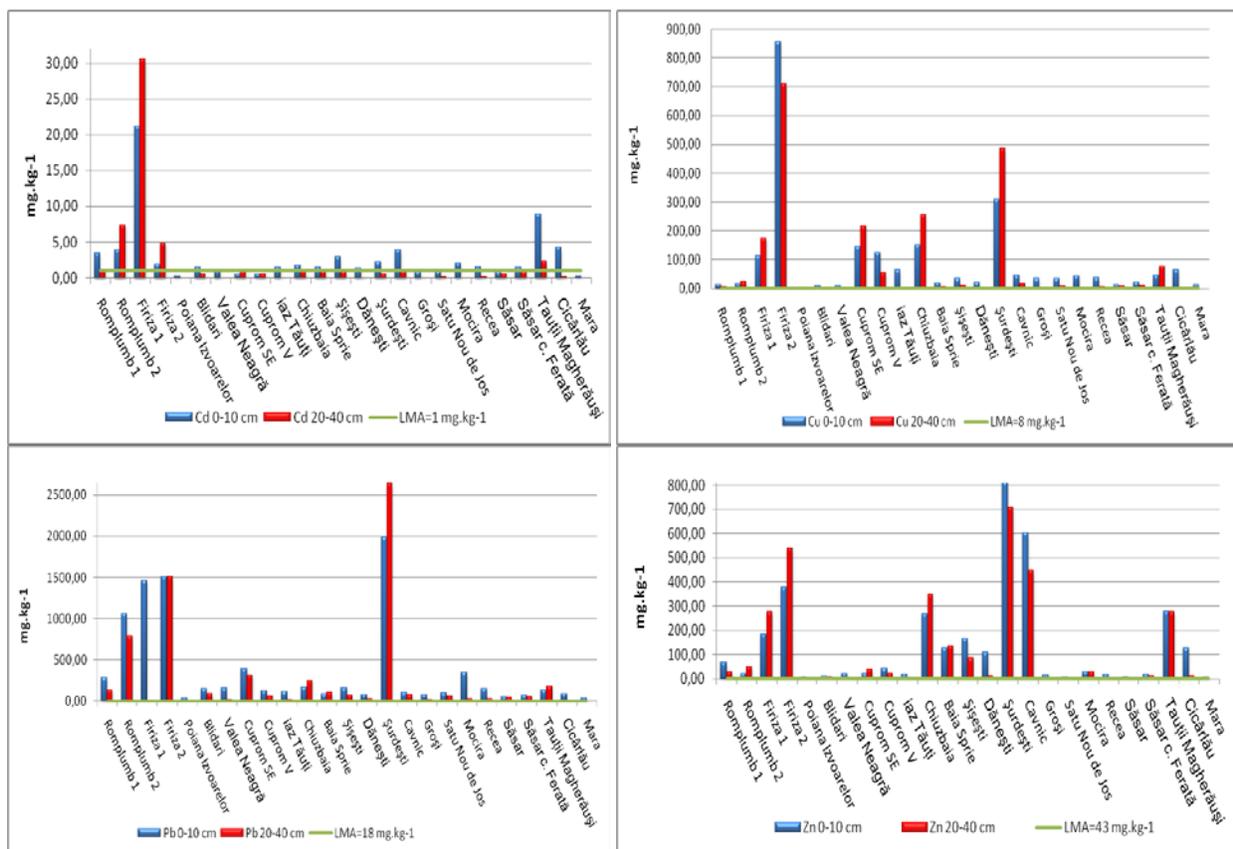


Figure 3 - Mobile contents of Cd, Cu, Pb, Zn in relation to MAL

The values of statistical parameters calculated for the mobile forms of heavy metals in both depths are shown in table 2:

Table 2 - Statistical parameters of heavy metal content ( $\text{mg}\cdot\text{kg}^{-1}$ ) determined in soil (mobile forms soluble in  $\text{EDTA}\cdot\text{CH}_3\text{COONH}_4$  solution)

Mobile forms	Depth	Average	Median	$X_{\min}$	$X_{\max}$	Standard deviation
Cd	0-10 cm	2,82	1,49	0,26	21,21	4,24
Co	0-10 cm	0,51	0,40	0,08	1,15	0,32
Cr	0-10 cm	0,09	0,04	0,00	0,37	0,10
Cu	0-10 cm	89,71	35,28	3,86	856,89	173,49
Fe	0-10 cm	286,38	199,31	8,00	1456,48	302,05
Mn	0-10 cm	110,02	85,67	12,20	283,92	80,32
Ni	0-10 cm	0,98	0,60	0,20	4,93	1,01
Pb	0-10 cm	357,40	134,58	32,98	1987,05	534,16
Zn	0-10 cm	134,85	28,82	6,77	813,49	201,75
Cd	20-40 cm	2,39	0,77	0,03	30,66	6,12
Co	20-40 cm	0,43	0,35	0,07	1,42	0,33

Mobile forms	Depth	Average	Median	X <sub>min</sub>	X <sub>max</sub>	Standard deviation
Cr	20-40 cm	0,07	0,02	0,00	0,42	0,11
Cu	20-40 cm	85,93	11,58	0,66	711,83	172,89
Fe	20-40 cm	204,48	128,94	2,94	1021,87	226,92
Mn	20-40 cm	75,95	51,87	5,56	302,90	69,11
Ni	20-40 cm	0,63	0,49	0,13	1,80	0,50
Pb	20-40 cm	266,88	64,93	5,75	2637,09	590,32
Zn	20-40 cm	124,48	26,85	1,11	708,40	195,98

#### 4. Heavy metals soil pollution in Baia Mare area compared with Zlatna, Copșa Mică areas

The data presented in the comparative study between Copșa Mică and Baia Mare on the typical luvisols and regosols, Baia Mare presents the highest values for the four heavy metals, Cu, Cd, Pb, Zn. Analyzes were made at a difference of 15 years (Lăcătușu et al., 1995, 2010), which demonstrates the residual character of heavy metals in relation to soil physico-chemical features.

In Zlatna the main soil types that were investigated (2006-2008) are aluvisols and districambosols (Damian et al., 2008a). Cadmium shows higher values in Zlatna instead copper, lead and zinc shows higer values in the Baia Mare area.

#### 5. Correlations of heavy metals in soil

Between the total content of heavy metals and the mobile heavy metal content from the soil solution were established directly proportional relationships statistically assured - significant and distinct significant. Correlation coefficient  $r$  has values close to 1 for the elements Pb (0.99 \*\* at 0-10 cm and 0.98 \* at 20-40 cm), Zn (0.94 \*\* at 0-10 cm and 0,90 \*\* at 20-40 cm), Cu (0.83 \* at 0-10 cm and 0.91 \*\* at 20-40 cm), Mn (0.73 \*\* at 0-10 cm and 0.55 \* at 20-40 cm). The order of correlation magnitudes for total and mobile forms of heavy metals in soil is: Pb > Zn > Cu > Mn.

#### 6. Reaction and the macroelements content in soils from the Baia Mare area

##### 6.1. Soil pH values in the Baia Mare area

Regarding the actual acidity for the main soil types were found as follows:

- H<sub>2</sub>O pH values are lower in areas as: Cuprom, Mocira, Firiza which shows a clear acidification;
- for all types of soil is noted that the current acidity is approximately the same in the organic horizon (0-10 cm depth) than in the mineral horizon (20-40 cm depth);
- minimum values may be more important than averages in assessing the acidification process.

##### 6.2. Soil supply with sulfur, phosphor and potassium

Data on the total contents of the main macroelements (S, P, K) indicates a poor supply of soil, mainly in the Romplumb area.

## 7. Distribution maps of heavy metals

Spatial distribution maps of heavy metals in the soil, at the two depths, showed highly polluted areas around the two metallurgical plants, and on the direction of prevailing winds, western and eastern; where the contents of cadmium and lead are higher than the background of the investigated area. Thus, it highlights the area of the southern part of Cavnic locality, the southern part of the Tăuții Măgherauși locality (fig. 4). The higher content of heavy metals from the last area are due also to the plant Transgold accident from 2000 year.

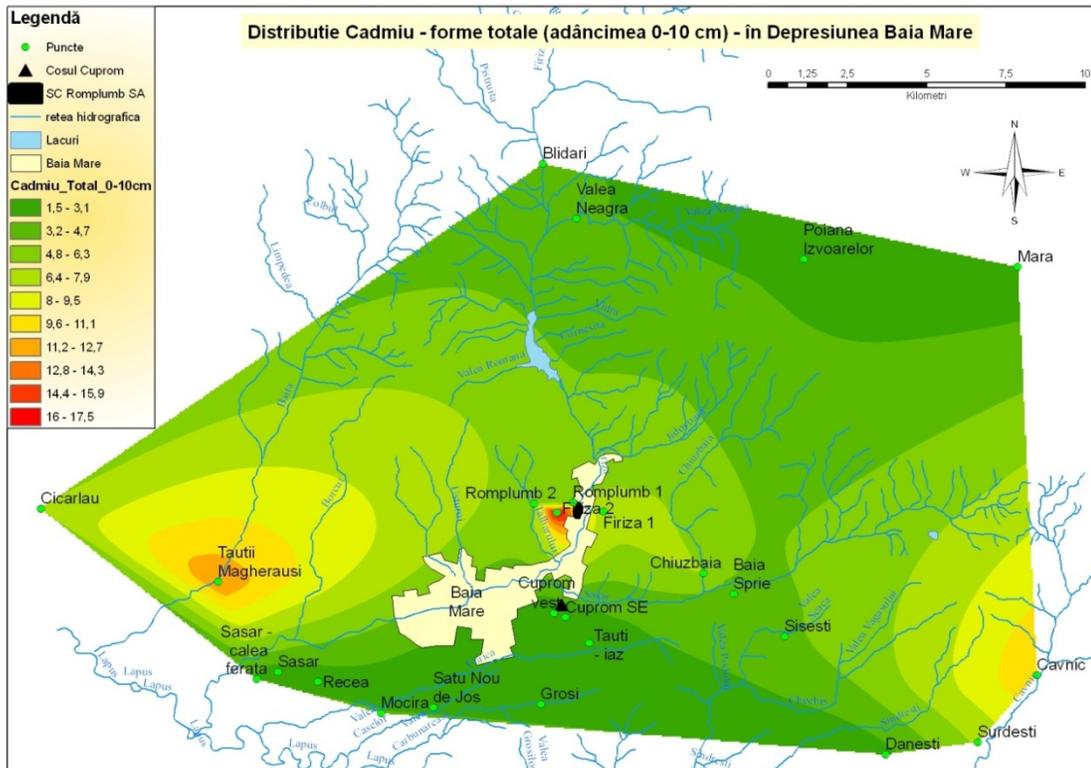


Figure 4 - Distribution of total forms (mg.kg-1) of cadmium at a depth of 0-10 cm

### HEAVY METALS TRANSLOCATION FROM SOIL IN CROPS PLANTS

#### Heavy metals in edible parts of vegetables and pasture plants

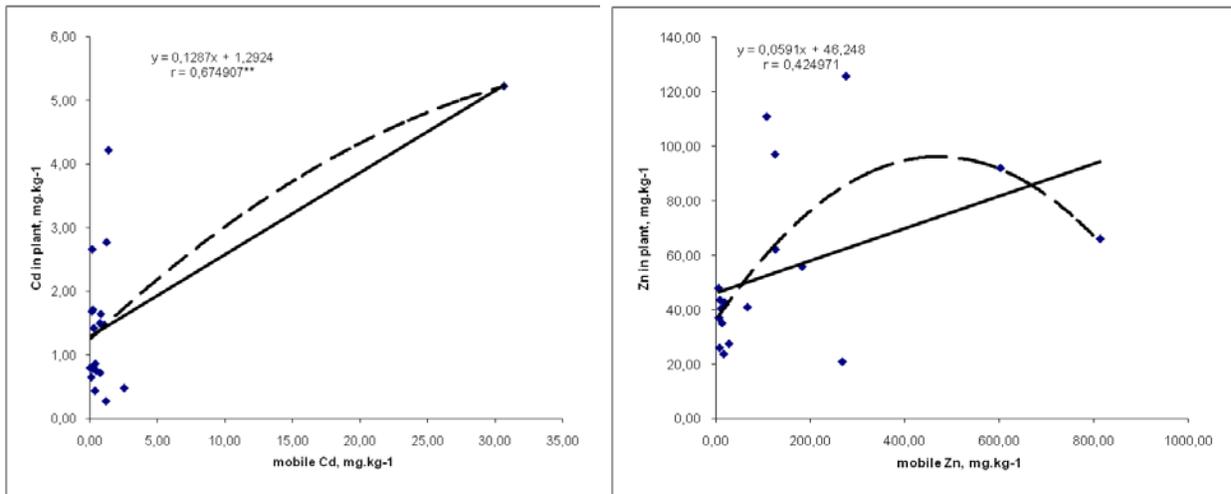
Were collected carrots, lettuce and parsley samples, on organs, roots and leaves. Their choice was made balanced, as are common in the human alimentation and it is consumed the root from carrots and parsley, and the leaves from lettuce. Also, salad is a good indicator of soil pollution with heavy metals and it is known as a good accumulator of these chemical elements. From analyzes conducted on vegetables and plants of meadows, clear differences are observed regarding heavy metal content. Analyzes also show multiple pollution with heavy metals (Cu, Cd, Pb, Zn).

Obvious differences in meaning of the accumulation of large quantities of heavy metals were recorded for vegetables whose edible part is the root (carrot, parsley) or leaves (lettuce). The highest values are found in salad. Also, the highest concentrations are found in leaves. Of all the plants the most vulnerable are the root plants.

### HEAVY METALS CORRELATIONS IN SOIL-PLANT SYSTEM

To determine the influence exerted by heavy metals in soils (total and mobile forms) on vegetables (carrots), simple correlations were performed using the statistical package SPSS 17, yielding Pearson correlation coefficient values.

The correlation coefficient  $r$  has significant values for the elements: total Cd (0.53 at 0-10 cm and 0.46 at 20-40 cm) and mobile (0.62 at 0-10 cm and 0.67 at 20-40 cm), mobile Zn (fig. 5), mobile Mn, Co total, total Ni. Values of correlation coefficients for mobile forms of heavy metals in soil join together as follows: Cd > Zn > Mn > Cr > Ni > Co.



**Figure 5 - Dependence of Cd and Zn content foam carrots towards the mobile content of Cd and Zn in soil (20-40 cm, 0-10 cm)**

### THE BIOCONCENTRATION (BCF) AND THE TRANSLOCATION FACTOR (TF)

1. The BCF order for *parsley* values was Zn > Cu > Cd > Pb, for *salad*: Cd > Zn > Cu > Pb respectively for *carrots*: Zn > Cu > Cd > Pb. The lowest value of BCF (0) for the three analyzed vegetables, lettuce, carrots, parsley, is the lead.

2. The TF order, from roots to leaves, for *parsley* was as follows: Pb > Cd > Cu > Zn, the highest value being for the lead, of 5.8, for the Romplumb area *parsley*, for *salad* the order was: Pb > Cu > Cd > Zn, the highest value (Pb) being in Mocira locality, of 4.5, and for the *carrots* was: Cd > Pb > Zn > Cu, Cd showing the highest value of 1.6 in the Recea locality. In contrast, the highest translocation is showed by the *parsley* and the lowest by the *carrots*.

## **Plant nutrient content of the Baia Mare area**

In Baia Mare area, the heavy metals contents in soil solution are much higher than the physiological optimum, which favors their uptake and accumulation in plants in high and very high quantities (up to 24 times higher than the maximum allowed, in case of the cadmium in lettuce leaves).

### **METHODS FOR LIMITING POLLUTION EFFECTS OF HEAVY METALS**

It is recommended measures both at:

- metallurgical units, responsible for the filtering of emissions sent into the atmosphere;
- tailing ponds, by greening the waste dumps and their vegetation cover;
- soil, by the amendment and the introduction of materials with absorptive role for heavy metals.

### **CONCLUSIONS ON HEAVY METALS SOIL AND PLANT POLLUTION IN BAIJA MARE AREA**

- The soil samples were analyzed both as a total and mobile content in solution of ammonium acetate-EDTA at a pH of 7 for the following: Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, and Zn. Also were analyzed the main physical and chemical properties of studied soils.

- Total cadmium in soils from Baia Mare area exceeds normal content. Mean values of total cadmium content at 0-10 cm depth exceeds 1.6 times the alert threshold. At a depth of 20-40 cm, the mean of total cadmium content exceeds the alert threshold of 1.46 times and its situated below the intervention threshold. Regarding mobile forms of cadmium content in the upper horizon of the soil, they recorded an average exceed by 2.8 times of the normal content, as well as at the depth of 20-40 cm (by 2.4 times).

- Total copper in soils analyzed area at a depth of 0-10 cm, is exceeding the normal content by 17.5 times, the alert threshold by 3.5 times and the intervention threshold by 1.8 times. At a depth of 20-40 cm, the total copper average content exceeds up to 12 times the normal content, up to 24 times the alert threshold and up to 1.2 times the intervention threshold. The average value of the mobile form of copper exceeds up to 11 times the normal content at the surface and up to 10.8 times at the depth.

- The average concentration of total lead in the upper horizon of Baia Mare soils, exceeding up to 50 times the normal concentration, up to 20 times the alert threshold and 10 times the intervention threshold. The mean of total lead content at a depth of 20-40 cm exceeds up to 39 times the normal contents, up to 15 times the alert threshold, and up to 7.7 times the intervention threshold. The mean mobile lead content at the surface exceeds up to 20 times the normal contents and up to 14.8 times at the depth.

- The average zinc content in full form at a depth of 0-10 cm exceeds up to 12 times the normal contents, up to 4 times the alert threshold of and up to 2 times the intervention

threshold. The average for mobile zinc concentrations at a depth of 0-10 cm exceeds the normal content up to 3 times and at the depth of 20-40 cm up to 2.9 times.

- The highest concentrations of heavy metals were determined in the surface soils horizon (0-10 cm depth).

- The concentration of heavy metals analyzed decreases with increasing the distance from the main sources of pollution. Soil types associated with the areas closest to the sources of pollution (eutricambosols, luvisols, and subordinate districambosols regosols) presents the highest degree of pollution. These soils have physical and chemical properties that promote retention and circulation of heavy metals. Among them an important role shows: the low pH, the low to medium content of organic matter and the cation exchange capacity.

- Positive correlations in majority statistically insured, were obtained between the total and mobile heavy metals in the soil at both depths.

- The comparative study conducted between Baia Mare and Zlatna, Copșa Mică areas pointed out that all three areas are heavily polluted with heavy metals, with slightly different content for a chemical element or another, depending on local conditions.

- Data on heavy metal concentrations obtained from analyzes of soil samples show that their values are higher than those obtained by various researches in the period 1996-2008.

- Copper in soil has an average value that excess up to 5 times MAC ( $5 \text{ mg.kg}^{-1}$ ). Exceeding the maximum allowable concentration of Cu ( $5 \text{ mg.kg}^{-1}$ ) was found in all analyzed plants, carrots and lettuce.

- Lead mean in soil exceeds up to 9.4 times the MAC.

- The average concentration of zinc exceeds up to 3.4 times the MAC in carrots.

- Results on analyzed plants in 2011, roots and leaves of lettuce, carrots, parsley, show that the highest values are found in the salad and by organs, in leaves.

- The highest average concentrations of cadmium are found in the lettuce leaves and roots, appearing a 24 times exceeding of MAC. Other vegetables, carrots and parsley (root and leaves), both the minimum and average values, exceed the maximum permissible concentration ( $0.1 \text{ mg.kg}^{-1}$ ).

- The mean lead for all analyzed vegetables and in all analyzed organs exceeds the maximum allowable concentration ( $0.5 \text{ mg.kg}^{-1}$ ).

- Highest average of zinc content occur in case of roots, leaves of salad and parsley roots, overruns the maximum allowable concentrations up to 4.8, 4.7, or 4.4 times. And in case of carrots the maximum allowable concentrations is exceeded, but lower compared with lettuce and parsley.

- Analyzing the results obtained in pasture plants results significant differences in the concentrations of heavy metals analyzed. Thus Cd is within normal range; Cu exceeds normal values falling into the toxicity threshold ( $20-100 \text{ mg.kg}^{-1}$ ). For Pb are also exceeded the normal values ( $5-10 \text{ mg.kg}^{-1}$ ), falling into the toxicity threshold ( $30-300 \text{ mg.kg}^{-1}$ ). Zn is included in the normal range.

- In the case of the correlations between total content of heavy metals in soil and the content from carrot roots is a wide variation range from 0.05 for Pb to 0.53\* for Cd in the upper horizon and from 0.06 for Zn to 0.46\* for Cd in the lower horizon. Correlation reports have maximum values for soluble soil solution forms being distinctly significant, compared to the total forms which are significant.

- The values of transfer factors for various vegetables analyzed vary depending on the species and location. The cadmium mean is 0.26, being also the highest BCF mean of vegetables. BCF of lead has a low average of 0.02, of copper 0.16 and of zinc 0.19.
- Contamination of soils and plants in Baia Mare area is significant, indicating a serious situation that requires the application of soil remediation methods.
- Due to the heavy metal content in the studied area, the accumulation of metals in vegetables grown near industrial site represents a potential risk to human health.
- It is not recommended the vegetable cultivation in the area polluted by heavy metals.

Considering the results obtained in field and laboratory work, as well as the interpretation of the results, we believe that the proposed objectives were fully accomplished.

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