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Translocation of Cu, Pb, Zn, Cd in Some Vegetables Grown in Polluted Area of Baia Mare, Romania

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Abstract. In this work, behaviors of lettuce and spinach, growing in a metal-polluted garden soils located near a nonferrous metallurgical center, were investigated through Cu, Pb, Zn and Cd soils and plant metal concentrations, their capacity to uptake these metals through concentration factor (CF) and translocated these metals from inedible to edible part of vegetables through translocation factor (TF). The present study highlights that both adults and children consuming vegetables grown in these garden soils ingest significant amount of these metals. The soil and plant samples had been collected from 2 places located at small distances from existing pollution sources Ferneziu and Tăuții de Sus. For the comparative analysis of the data, also a reference zone had been selected from Sighetu-Marmației, area not under the direct impact of mentioned pollution sources. Heavy metal (Pb, Cu, Zn, Cd) content from soil and plant samples were analysed on a Perkin Elmer Flame AAS. The values for Cu in total forms in soil varied between 31.278 mg·kg^{-1} (Sighet) and 216.042 mg·kg^{-1} (Ferneziu). The concentrations of Pb in total forms ranged from 38.671 mg·kg^{-1} (Sighet) at 2036.362 mg·kg^{-1} (Ferneziu). The concentration of Zn varied between 137.681 mg·kg^{-1} (Sighet) and 1992.322 mg·kg^{-1} (Ferneziu). The minimum value obtained for Cd was 0.789 mg·kg^{-1} (Sighet) and the maximum value was 7.854 mg·kg^{-1} (Ferneziu). The order of average CF values are similar in lettuce root and spinach leaves being followed: Cd >Cu> Zn >Pb. The order of average CF values at spinach from soil to root was: Cd> Pb> Cu >Zn. The CF average values from soil to lettuce leaves was in the following order: Cd> Pb >Cu >Zn. In lettuce the TF (root to leaves) values in varied in the following order Zn> Cu> Cd >Pb in all studied areas. In spinach the order of variation for TF values was Cu>Cd>Zn>Pb. This study reveals an intense soil and plant pollution in the areas adjacent to sources of pollution (Tăuții de Sus and Ferneziu) and indicate a translocation of these heavy metals in plants. Their consumption make possible an inevitably transfer of heavy metals in the body, which can have serious consequences on human health.

Keywords: soil contamination, heavy metals in foods, Baia Mare, metal toxicity, garden soils, translocation factor.

INTRODUCTION

The accumulation of heavy metals in agricultural soils is of increasing concern because of food safety issues, potential health risks, and its detrimental effects on soil ecosystems (Cui et al., 2004). Pb is of great concern because of their toxicity to human health and other organisms, whereas Zn and Cu are essential elements for plants and human body. Vegetables take up Cu, Pb, Zn, and Cd and accumulate them in their edible and inedible parts with various concentrations. The intake of the edible parts of vegetables is an important path for heavy metals in the soil to harm human health. It is therefore important to control and limit the accumulation of heavy metals in vegetables.

The researches conducted in Baia Mare area have highlighted multiple pollution by heavy metals (Pb, Zn, Cu, Cd) in the residential, agricultural and forestry soils felt at a distance over 25-30 km around the major pollution sources, as a consequence of the high emission levels and high frequency of exceeding the maximum admissible concentrations of heavy metals in the ambient air, (Cordoş et al., 2007; Damian et al., 2008; Frențiu et al., 2008, 2009; Ivasuc, 2008; Oprea et al., 2010).

Location of the most important sources of pollution in urban area in the east and northeast respectively, on the dominant wind directions and with reduced dispersion of pollutants (slow air circulation, calm atmosphere and frequent thermal inversions) generated by the relief area (depression) causes a high level of pollution emissions across the specific area of BaiaMare.

The first objective of this research was to evaluate the level of garden soils contamination with lead, copper, zinc and cadmium in Tăuții de Sus and Ferneziu located in the vicinity of the nonferrous metallurgical center from Baia Mare. The reference zone was Sighetu-Marmației less polluted area. The soil in Baia Mare area is affected by the emissions of powders containing metals from metallurgical factories. The second objective was to evaluate the contamination level at lettuce (*Lactuca sativa* L. *var.capitata*) and spinach (*Spinacia* L. *oleracea var. matador*) grown on mentioned garden soils. The third objective was to evaluate the two vegetables uptake capacity of Cu, Pb, Zn and Cd from soil through the concentration factor (CF). The fourth objective was to know the behavior of elements in both species of plants throught the translocation factor (TF) that highlights the transition of metals from inedible to edible parts of vegetables.

Therefore, the relationships between metal contents in soils, plant roots and leaves studied in the present work would offer interesting and essential results that contribute to the knowledge of the bio-monitoring process.

MATERIALS AND METHODS

Location of the studied area: the soil and plant samples had been collected from 2 places located at small distances from existing pollution sources from Baia Mare depression, (fig.1). For the comparative analysis of the data, also a reference zone had been selected from Sighetu-Marmației, area not under the direct impact of mentioned pollution sources.

Soil and plant sampling: soil and vegetables samples from vegetables gardens within the areas influenced by emissions loaded with heavy metals, were collected in may 2010. The soil samples collected from the 0-20 cm depth of the A horizon, were been analyzed from point of view of general physical and chemical properties as pH, quantity of humus, quantity of clay and total forms of heavy metals (Cu, Pb, Zn, Cd).

The plant samples from the garden soils come from a very common vegetables used in human alimentation, lettuce (*Lactuca sativa* L. *var. capitata*) and spinach (*Spinacia oleracea* L. *var. matador*) These plant species was chosen because between vegetables, have the greatest capacity to accumulate heavy metals without manifest visible phytotoxicity symptoms, which enhances the risk to human health.

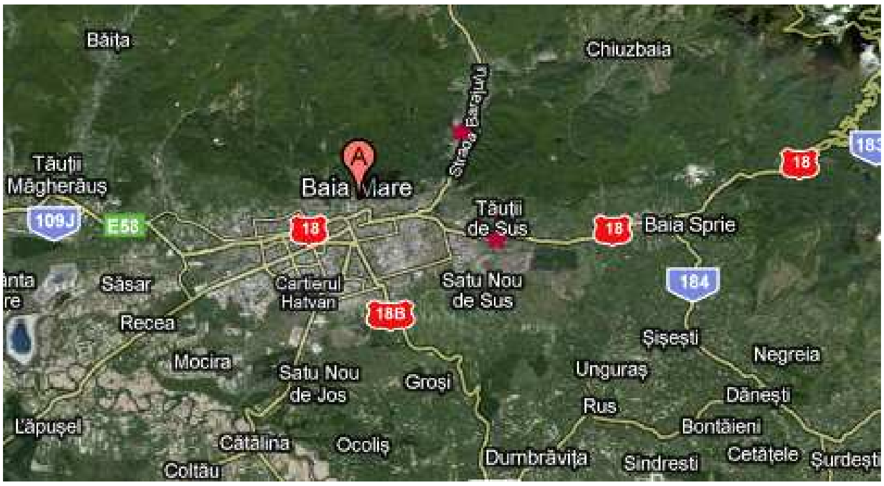

Fig. 1. Map of sampling points from the localities under the direct impact of pollution sources

Chemical analysis: the soil reaction (pH values) carried out by potentiometric method in aqueous suspension, using a double glass-calomel electrode. Determinations related to clay and humus content are part of the Romanian agrochemical system that establishes the heavy metals chemistry, (ICPA, 1981). The soil samples were digested with HCl:HNO_3 (3:1) acid mixture, (SR ISO 11466 :1999). The total content of heavy metals (Cu, Pb, Zn and Cd) was measured with flame atomic absorption spectrometer according to the SR ISO 11047:1999 method.

All the collected plant samples were washed with double distilled water to remove airborne pollutants. Each individual vegetable was separated into root and leaf sub-samples. All sub-samples were weighed and air-dried for a day, to reduce water content. All the samples were then oven-dried in a hot air oven at 70–80 °C for 24 h, to remove all moisture. Dried samples were powdered using a pestle and mortar and passed through a 0.15-mm sieve. The powdered samples were accurately weighed (1g each) and placed in beakers. Samples was treated with 5 ml ultra-pure 65%HNO_3. The beaker was covered with a watch-glass and the suspension was heated up to 130°C for 1 h. A total amount of 4 ml 20% H_2O_2 was added in aliquots of 0.5 ml. After cooling the solution was quantitavely transferred to a 50 ml volumetric flask and diluted to the mark with distilled water, (Marinussen and van der Zee, 1997).

Standards: the reagents used during the process were of analytical grade. Standard solutions of heavy metals (1000 mg/l), namely copper (Cu), zinc (Zn) and lead (Pb) were procured from Merck. Solutions of varying concentrations were prepared for all the metals by diluting the standards.

Data analysis: All data were analyzed using the SPSS 17.0 statistical package. Total metal concentrations were expressed as average, minimum and maximum values for soils, roots and leaves at both vegetables. Data from exposed plants were compared with those from controls and with standards.

Concentration factor (CF): expresses the ratio of metal concentration (M) between plants and soils. The ratio was calculated for both species of plants revealing the behavior about the pollutants studied, CF=(Mplant/Msoil).

Translocation factor (TF): is the ratio of metal concentration in plant's aerial parts (C_{aerial}) and metal concentration in plant's root (C_{root}), (Marchiol et al., 2004). TF= (C_{aerial} / C_{root}

RESULTS AND DISCUSSION

Tab.1

Heavy metals contents of vegetables and soils in Tautii de Sus area

Experimental values	Cu	Pb	Zn	Cd
Soil total content (mg·kg^{-1})	137.130 [a] (67.329–206.986)	1010.483 (754.735-1273.472)	551.325 (385.442-723.536)	5.244 (4.928-5.514)
Lettuce root (mg·kg^{-1})	12.156 (10.321-13.045)	88.047 (79.642-100.031)	89.678 (86.409-92.708)	0.837 (0.755-0.913)
Lettuce leaves (mg·kg^{-1})	10.369 (7.549-12.013)	65.081 (49.512-82.668)	80.519 (78.526-82.531)	0.794 (0.65-0.98)
Spinach root (mg·kg^{-1})	13.784 (12.152-15.072)	112.878 (100.761-126.354)	89.859 (84.311-98.354)	0.766 (0.602-0.962)
Spinach leaves (mg·kg^{-1})	15.592 (14.625-16.355)	56.525 (51.477-64.376)	79.509 (77.499-81.024)	0.839 (0.703-0.944)
The maximum levels in soil [b] (mg·kg^{-1})	100 [d] – 200 [e]	50 [d] – 100 [e]	300 [d] – 600 [e]	3 [d] – 5 [e]
The maximum levels in vegetables[c] (mg·kg^{-1})	73.3	0.3	99.4	0.2

[a] Average (min. – max.)
[b] Maximum levels for heavy metals from soil according with the Romanian standard 756/1997
[d] Warning threshold; [e] Intervention threshold for sensitive type of land
[c] Maximum levels in vegetables according with FAO/WHO- Codex alimentarius commission, 2001

Tab.2

Heavy metals contents of vegetables and soils in Ferneziu area

Experimental values	Cu	Pb	Zn	Cd
Soil total content (mg·kg^{-1})	185.319 [a] (138.225–216.042)	1548.858 (1288.369-2036.362)	1756.332 (1359.510-1992.322)	5.460 (3.373-7.854)
Lettuce root (mg·kg^{-1})	13.571 (12.843-14.651)	121.893 (121.893-138.721)	95.233 (90.824-99.562)	0.988 (0.942-1.017)
Lettuce leaves (mg·kg^{-1})	11.291 (10.212-12.077)	74.804 (93.680-100.432)	93.027 (87.652-97.621)	0.953 (0.673-1.132)
Spinach root (mg·kg^{-1})	14.109 (12.844-15.672)	165.401 (139.673-181.672)	95.809 (91.673-100.653)	0.977 (0.772-1.121)
Spinach leaves (mg·kg^{-1})	15.254 (14.643-16.162)	92.555 (82.859-102.412)	87.526 (84.442-89.643)	0.997 (0.778-1.113)
The maximum levels in soil [b] (mg·kg^{-1})	100 [d] – 200 [e]	50 [d] – 100 [e]	300 [d] – 600 [e]	3 [d] – 5 [e]
The maximum levels in vegetables[c] (mg·kg^{-1})	73.3	0.3	99.4	0.2

[a] Average (min. – max.)
[b] Maximum levels for heavy metals from soil according with the Romanian standard 756/1997

[d] Warning threshold; [e] Intervention threshold for sensitive type of land
[c] Maximum levels in vegetables according with FAO/WHO- Codex alimentarius commission, 2001

Tab.3

Heavy metals contents of vegetables and soils in Sighetu-Marmatiei area

Experimental values	Cu	Pb	Zn	Cd
Soil total content ($mg \cdot kg^{-1}$)	44.981 [a] (31.278–51.651)	56.652 (38.671-74.933)	157.808 (137.681-172.875)	1.005 (0.789-1.161)
Lettuce root ($mg \cdot kg^{-1}$)	5.387 (4.611-6.011)	2.985 (2.653-3.421)	11.999 (10.700-14.056)	0.214 (0.125-0.318)
Lettuce leaves ($mg \cdot kg^{-1}$)	4.300 (3.662-4.781)	2.027 (1.662-2.602)	10.601 (9.963-11.378)	0.063 (0.051-0.079)
Spinach root ($mg \cdot kg^{-1}$)	5.457 (4.972-6.089)	8.114 (6.938-9.645)	16.614 (15.672-18.221)	0.075 (0.065-0.083)
Spinach leaves ($mg \cdot kg^{-1}$)	6.013 (5.792-6.115)	3.722 (2.881-4.880)	13.490 (12.726-14.272)	0.059 (0.052-0.068)
The maximum levels in soil [b] ($mg \cdot kg^{-1}$)	100 [d] – 200 [e]	50 [d] – 100 [e]	300 [d] – 600 [e]	3 [d] – 5 [e]
The maximum levels in vegetables[c] ($mg \cdot kg^{-1}$)	73.3	0.3	99.4	0.2

[a] Average (min. – max.)
[b] Maximum levels for heavy metals from soil according with the Romanian standard 756/1997
[d] Warning threshold; [e] Intervention threshold for sensitive type of land
[c] Maximum levels in vegetables according with FAO/WHO- Codex alimentarius commission, 2001

Heavy metals in soil

The main physico-chemical parameters determined for soils from the studied areas are as follows: humus contents are within the range of 2.18-5.88%, with average value being approximately 3.69%; values of pH was between 4.05-7.70, (Tab.4,5); clay content varied between 10.25–28.20%, with average value of 19.31%.

The values for Cu in total forms in soil varied between 31.278 $mg \cdot kg^{-1}$ (Sighet) and 216.042 $mg \cdot kg^{-1}$ (Ferneziu) with an average content of 122.477 $mg \cdot kg^{-1}$, (Tab.1,2,3). The maximum value obtained exceeded 2.16 times the warning threshold (100 $mg \cdot kg^{-1}$) and 1.08 times the intervention treshold for Cu at sensitive type of land.

The concentrations of Pb in total forms ranged from 38.671 $mg \cdot kg^{-1}$ (Sighet) at 2036.362 $mg \cdot kg^{-1}$ (Ferneziu) with an average content of 871.998 $mg \cdot kg^{-1}$, (Tab.1,2,3). A percentage of 86.66% of obteind values exceeded the warning threshold (50 $mg \cdot kg^{-1}$) and 66.66% exceeded the intervention threshold (100 $mg \cdot kg^{-1}$) for Pb at sensitive type of land.

The values for Zn in Tăuți area exceeded 38-2.41 times the warning threshold (300 $mg \cdot kg^{-1}$), some values even exceeded the intervention threshold (600 $mg \cdot kg^{-1}$) for Zn at sensitive type of land. At Ferneziu area the obtained values exceeded 2.26-3.32 times the intervention threshold. The concentration of Zn varied between 137.681 $mg \cdot kg^{-1}$ (Sighet) and 1992.322 $mg \cdot kg^{-1}$ (Ferneziu) with an average values of 821.822 $mg \cdot kg^{-1}$, (Tab.1,2,3).

The minimum value obtained for Cd was 0.789 mg·kg^{-1} (Sighet) and the maximum value was 7.854 mg·kg^{-1} (Ferneziu) with an average value of 3.904 mg·kg^{-1} , (Tab. 1,2,3). At Ferneziu and Tăuți areas the obtained values exceeded 1.12-2.618 times the warning threshold (3 mg·kg^{-1}) and 40% of the obtained values exceeded 1.01-1.57 times the intervention threshold 5 (mg·kg^{-1}).

Heavy metals in plants

The concentrations of Cu in lettuce leaves ranged from 3.662 mg·kg^{-1} (Sighet) at 12.077 mg·kg^{-1} (Ferneziu) with an average content of 8.656 mg·kg^{-1}, and in spinach leaves ranged between 5.792 mg·kg^{-1} (Sighet) and 16.355 mg·kg^{-1} (Ferneziu) with an average value of 12.286(Tab.1,2,3). All values are located below the maximum permited level provided by FAO/WHO- Codex alimentarius commission, 2001

The values for Pb in lettuce leaves varied between 1.662 mg·kg^{-1} (Sighet) and 100.432 mg·kg^{-1} (Ferneziu) with an average content of 53.596 mg·kg^{-1} and in spinach leaves ranged from 2.881 mg·kg^{-1} (Sighet) at 102.412 mg·kg^{-1} (Ferneziu) with an average value of 50.934. (Tab.1,2,3). The values obtained for the two vegetables exceed the 5.54 to 341.37 times the maximum permitted levels.

The concentrations of Zn in lettuce leaves ranged from 9.963 mg·kg^{-1} (Sighet) at 97.621 mg·kg^{-1} (Ferneziu) with an average content of 61.383 mg·kg^{-1}, and in spinach leaves ranged between 12.726 mg·kg^{-1} (Sighet) and 89.643 mg·kg^{-1} (Ferneziu) with an average value of 60.175 (Tab.1,2,3). All values are located below the maximum permited level provided by FAO/WHO- Codex alimentarius commission, 2001.

The concentrations of Cd in lettuce leaves ranged from 0.051 mg·kg^{-1} (Sighet) at 1,132 mg·kg^{-1} (Ferneziu) with an average content of 0.604 mg·kg^{-1}, and in spinach leaves ranged between 0.052 mg·kg^{-1} (Sighet) and 1,113 mg·kg^{-1} (Ferneziu) with an average value of 0.632 (Tab.1,2,3). The values obtained for the two vegetables in the polluted areas exceed the 3.26 to 5.66 times the maximum permitted levels.

Concentration factor (CF)

The concentration factor CF (also called uptake factor, accumulation factor) is ratio between plant and soil concentrations of elements. This represents an index of soil–plant transfer that favors the understanding of plant uptake characteristics and it is widely used in biomonitoring studies, (Chamberlain et al., 1983; Mingorance et al., 2005). Ratios >1 indicate that plants are enriched in elements (accumulator), ratios around 1 indicates that plants are not influenced by elements (indicator), and ratios <1 shows that plants exclude the elements from uptake (excluder), (Baker, 1981).

The order of average CF values are similar in lettuce root and spinach leaves being followed: Cd >Cu> Zn >Pb. The order of average CF values at spinach from soil to root was: Cd> Pb> Cu >Zn. The CF average values from soil to lettuce leaves was in the following order: Cd> Pb >Cu >Zn.

Analyzing these data we can see that the highest average value 0.12 was for CF from soil to root in spinach, followed by average CF value from soil to root in lettuce 0.11 and average CF value from soil to leaves in spinach 0.09. The lowest CF average value was obtained at lettuce for heavy metal translocation from soil to leaves 0.08, (Tab. 4).

The obtained values were <1 for all elements indicating a low translocation from soil to plant leaves in all sampling area. Therefore, both species act as excluders for all studied elements.

Translocation factor (TF)

Translocation factor (TF) of heavy metals from roots to leaves is an indicator that helps to understand the mobility of heavy metals in plants and the human health risk they represent when reached in the edible parts of the vegetables, (Tab.5).

The mean values of TF for Cu in lettuce roots to leaves was between 0.79-0.85 which indicated that there was a strong capability of root tissues to hold Cu against the transport to leaves under both condition Cu excess and Cu normal content. The mean of TF for Cu in spinach was between 1.08-1.13 a higher values than in lettuce, indicating an easier copper translocation to leaf in spinach.

The TF mean values for Pb is less than the average TF values for Cu at both vegetables. The mean values ranges between 0.67-0.76 at lettuce and between 0.45-0.55 at spinach. Antagonistic effects were observed in translocation of lead in these two vegetables compared to copper translocation. TF values were higher in lettuce than in spinach, which indicates an easier lead translocation from roots to leaves in lettuce.

The TF average values obtained for Zn are very similar for these two vegetables.The average TF values ranges between 0.88-0.97 at lettuce and between 0.81-0.91 at spinach. These values indicate a similar translocation of Zn in these two vegetables.

For Cd the average TF values in the polluted areas (Tăuții de Sus and Ferneziu) ranges between 0.94-0.96 at lettuce and between 1.02-1.09 at spinach. The values are lower in the reference area (Sighet) 0.29 at lettuce and 0.78 at spinach.

Tab.4

The concentration factor (CF) of heavy metals from soil to roots and leaves of the vegetables

Sampling area	pH in soil	Vegetables	Cu	Pb	Zn	Cd
			CF average values			
Tautii de Sus	6.15	Lettuce ^{r}	0.08	0.08	0.16	0.15
		Lettuce ^{l}	0.07	0.06	0.14	0.15
		Spinach ^{r}	0.1	0.1	0.16	0.14
		Spinach ^{l}	0.11	0.05	0.14	0.15
Ferneziu	4.05	Lettuce ^{r}	0.07	0.07	0.05	0.18
		Lettuce ^{l}	0.06	0.04	0.05	0.17
		Spinach ^{r}	0.07	0.1	0.05	0.17
		Spinach ^{l}	0.08	0.05	0.04	0.18
Sighetu-Marmatiei	7.7	Lettuce ^{r}	0.11	0.06	0.05	0.07
		Lettuce ^{l}	0.09	0.03	0.06	0.06
		Spinach ^{r}	0.12	0.14	0.1	0.07
		Spinach ^{l}	0.13	0.06	0.08	0.05

^{r} root; ^{l} leaves

Tab.5

The translocation factor (TF) of heavy metals from roots to leaves of the vegetables

Sampling area	pH in soil	Vegetables	Cu	Pb	Zn	Cd
Tautii de Sus	6.15	Lettuce	0.85	0.73	0.89	0.94
		Spinach	1.13	0.50	0.88	1.09
Ferneziu	4.05	Lettuce	0.83	0.76	0.97	0.96
		Spinach	1.08	0.55	0.91	1.02
Sighetu-Marmatiei	7.7	Lettuce	0.79	0.67	0.88	0.29
		Spinach	1.10	0.45	0.81	0.78

CONCLUSIONS

Regarding to the first objective of the present work, we conclude that the variation of heavy metals content depends very much on the position of sampling site from sources of pollution. In the areas adjacent to sources of pollution (Tăuții de Sus and Ferneziu) most values exceeded the warning threshold even the intervention threshold for Cu, Pb Zn and Cd at sensitive type of land. The soils from the polutted areas (Ferneziu and Tăuții de Sus), through the composition and physic-chemical properties influences the concentration level and the mobility of Cu, Pb, Cd and Zn.

Regarding the second objective, the obtained values for Pb and Cd far have exceeded the maximum permited levels for the polluted area and ingestion of these contaminated vegetables are a real danger for human health.

Regarding the third objective, results for CF show that the differences between average CF values were not significant, indicating a relatively similar behavior for the absorption and translocation of these metals in the two studied vegetables.

Regarding the fourth objective, the results show that TF (root to leaves) values in lettuce plants varied in the following order Zn> Cu> Cd >Pb in all studied areas. In spinach the order of variation for TF values was Cu>Cd>Zn>Pb.

In conclusion, reduction of pollution will have to start from the source of pollution, that is by industry retechnologization, expanding automatization, improving existing technology, workforce qualification, in other words, pollution prevention, followed by the introduction of technical and organizational systems against pollution.

Therefore, the government should pay more attention to environmental pollution around metalurgical industry centers to prevent negative effects on human health.

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