

THE ACCUMULATION AND REMANENCE OF HEAVY METALS IN SOIL, A RISK FACTOR FOR THE POPULATION HEALTH IN COPȘA MICĂ AREA

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Abstract

This paper presents the dynamics of sulphur compounds noxae in the synergic action with heavy metals (Cu, Pb, Zn, Cd) in the soils and forest vegetation of Copșa Mică area, the two moments with different noxae regime, namely: 1986-1988 period with highly increased noxae concentrations exhausted through low chimneys into atmosphere, the areal generators of more reduced influence and 2006-2008 period with more reduced noxae concentrations because of the decrease of industrial capacities, exhausted through an over 350 m high chimney, generator of extended areas of pollution influence.

Consequently, the determinations carried out with an over 350 m high chimney in 1986-1988 emphasized much higher values over the admissible limit (LMA) of noxae concentrations in the vegetation and litter in accordance with their level in the air and soils and in the first 10 cm, their values were in normal limits, because of the relatively recent pollution source (10-15 years).

After about 25 years, investigating again the same sample surface plots, a regularity fact was emphasized, namely: this time the noxae concentrations in the vegetation and litter record normal values at the most admissible limits in accordance with the values recorded in the atmosphere but on the contrary at the soil depth of the first 10 cm, because of their accumulation, the concentrations values record very high levels much higher than admissible highest limit.

Considering that in the first 10-15 cm of soils, the plants necessary for peoples' nutrition (cereals, vegetables, pastures and grasslands) grow, we also extended our investigations in the people's vegetables gardens from different localities in the area of Copșa Mică polluted until the beginning of 2009.

The paper presents extremely high alarming values of heavy metals, especially Cd and Pb in the vegetables cultivated in the people's gardens. Such heavy metals can induce neoplasm in the population of the affected areas.

These high values are recorded in the conditions where the pollution source from Copșa Mică was ceased beginning with 2009.

Key words: food security, concentrations, noxae, heavy metals, soils, vegetation, litter, cultivated plants.

INTRODUCTION

The Food and Agriculture Organization (FAO) has chosen this year the slogan “United against famine” and the 16th of October was declared “The Food World Day”. This is the reason why I consider that the symposium dedicated to food security followed the same coordinates. This manifestation was organized by the Faculty of Environment Protection from the University of Oradea (Romania) in collaboration with the University of Debrecen (Hungary), University of Szeged (Hungary), Technological Education Institute of Thessaloniki (Greece), University of Valladolid (Spain), University of Leon (Spain), University of Agriculture Nitra (Slovakia), University “Politehnica” Timișoara (Romania), Academic Society for Environmental Protection (Romania), under the patronage of Romanian Ministry of Education, Researches and Innovation.

Food security also includes besides the prevention against famine, the quality of food which people consume.

Being concerned with the high concentrations of heavy metals in A₀ horizon and soils from Copșa Mică area, the investigations were extended in the area of the forest fund stock, the population’s gardens in the localities of the respective area.

Beginning with 2009 the pollution sources from Copșa Mică area were ceased. Considering the conditions where plants are no longer in direct contact with the noxae in the atmosphere, still the heavy metals concentrations are much higher than the admissible limit being recorded in the vegetables leaves and fruits frequently cultivated in the population’s gardens as well as in the grass from pastures and meadows where animals graze, finally reaching by trophic chains to human beings.

This is the explanation of the high incidence of some incurable diseases, malformations among the population of Copșa Mică area, the life average being more reduced in comparison with the population living in other areas which are not polluted.

Generally metals and especially heavy metals penetrate deeply the soil through precipitations, reach by biologic accumulation the plants which in their turn are consumed by people and animals. The medium contents of some heavy metals in the non-contaminated soils were established by **Lindsay** (1979), as follows: cadmium (Cd) – 0,06 mg/kg or ppm; copper (Cu) – 30 mg/kg; lead (Pb) – 10 mg/kg; manganese (Mn) – 600 mg/kg; zinc (Zn) – 50 mg/kg. Directive 86/278/EEC established the following limits for heavy metals contents: cadmium (Cd), 1-3 mg/kg dry matter; copper (Cu), 50-140 mg/kg dry matter, lead (Pb), 50-300 mg/kg dry matter and zinc (Zn), 150-300 mg/kg dry matter.

Table 1 presents the limit in heavy metals contents in soil (mg/kg dry matter), in some countries in Europe.

Table 1

**The limit of heavy metals contents in soil
(mg/kg dry matter)**

Metal	EEC Directive 86/278	Country								
		Austria	Germany	Spain	France	Greece	Sweden	Italy	Holland	England
Cadmium (Cd)	1 – 3	1 – 2	1.5	1 – 3	2	1 – 3	0.4	1.5	0.8	3
Copper (Cu)	50–140	60-100	60	50-210	100	50-140	40	100	36	80-135
Lead (Pb)	50-300	100	100	50-300	100	50-300	40	100	85	300
Zinc (Zn)	150-300	200-300	200	150-450	300	150-300	100-150	300	140	200-300

Source: Leonard, I., Dumitru, M., Vrânceanu, N., Moteliică, M.D., Tănase, V., 2007

In **Dumitru's** opinion (1990) the maximum admissible limits of some heavy metals in soil are: cadmium (Cd), 3 ppm (mg/kg); lead (Pb), 100 ppm, copper (Cu), 300 ppm, zinc (Zn), 300 ppm, manganese (Mn), 1000 ppm.

Heavy metals contents are effective on the biologic activity of soils consisting in a significant increase of the number of fungi and bacteria, the decrease of the dehydrogenasic activity; nitrogen fixation due to the clover which is completely absent because of the toxicity on *Rhizobium*, culminating in the decrease of genetic diversity (Mc.Grath, 1992); effects on some living creatures in the soil (especially earthworms) become toxic for them at concentrations of 110 ppm Cu and 1100 ppm Zn; there is a high risk for the trophic chain in the situation of overloading soils with these elements even if they are not transferred into plants (Bayer et al, 1982); the concentration of cadmium in the earthworms tissue.

Consequently, for a 0.2-1.2 ppm Cd soil concentration, a concentration of 7.6-17.5 ppm Cd was dosed in the earthworms tissue (Czarnowska and Jopkiewicz, 1978, mentioned by Leonard I. et al., 2007), the blocking of the nitrogen symbiotic fixation by leguminous plants at 330 ppm Zn, 99 ppm Cu and 10 ppm Cd concentrations (Rothamsted, 1987, mentioned by Leonard I. et al., 2007).

Heavy metals presence in the cultivated plants affect them, namely: beet leaves are an important accumulator of heavy metals therefore the zootoxic level for cadmium is 1 ppm in dry matter and the phytotoxic level is 8 ppm d.m. (Hani and collaborators, 1983, mentioned by Leonard I. et al 2007); the plants exposed to extreme levels of copper in soil can be toxic for most animals. Consequently, sheep are very sensitive at copper, toxicity occurs when fodder contains 12-15 ppm Cu (White, 1976); the critical values in the plant, dangerous for animals are 100 ppm Zn, 50 ppm Pb, 30 ppm Cu (Pfeiffer and collab., 1990), mentioned by Leonard I. et al 2007);

the maximum admissible levels of metals in the leaves of salad are: 20 ppm Cu, 1,2 ppm Pb, 120 ppm Zn and below 0,1 ppm Cd d.m.; English regulations stipulate for vegetables on sale a concentration of Pb below 1 ppm in fresh matter which coincides about 10 ppm in dry matter (Davies, 1990, mentioned by Leonard I. et al, 2007); some heavy metals, for example cadmium can accumulate at dangerous levels for consumers without affecting plants productivity, leading to a high risk for the trophic chain; it was noticed a direct correlation between the maximum concentrations of cadmium in the maize grain and zinc concentrations. Thus, for a 1 ppm Cd concentration and a 40 ppm Zn volume cause people food poisoning (John, M.K., 1973); heavy metals availability for plants is not constant, it varies considering the species, climate and soil; green salad has a high capacity to accumulate heavy metals like zinc, copper and cadmium in comparison with potatoes and carrots which have a low affinity for these heavy metals in soil, and clover absorbs copper faster than the graminaceae species. The tolerance levels of various crops for heavy metals are in a decreasing order: herbs, graminaceae, cereals, potatoes and sugar beet.

Table 2 presents the maximum levels of Cd and Pb, the most toxic heavy metals in some foodstuffs, according to EC Regulation No. 1881/2006

Table 2

Maximum levels for certain contaminants in foodstuffs
Heavy metals

No.	Foodstuffs	Maximum levels (mg/kg wet weight)
1	Lead	
1.1.	Raw milk, heat-treated milk and milk for the manufacture of milk-based products	0.020
1.2.	Cereals, legumes and pulses	0.20
1.3.	Vegetables, excluding brassica vegetables, leaf vegetables, fresh herbs and fungi. For potatoes the maximum level applies to peeled potatoes	0.10
1.4.	Brassica vegetables, leaf vegetables and cultivated fungi	0.30
1.5.	Fruit, excluding berries and small fruit	0.10
1.6.	Fruit juices, concentrated fruit juices as reconstituted and fruit nectars	0.050
1.7.	Wine (including sparkling wine, excluding liqueur wine), cider, perry and fruit wine	0.20
2.	Cadmium	
2.1.	Meat (excluding offal) of bovine animals, sheep, pig and poultry	0.050
2.2.	Cereals excluding bran, germ, wheat and rice	0.10
2.3.	Bran, germ, wheat and rice	0.20
2.4.	Soybeans	0.20
2.5.	Vegetables and fruit, excluding leaf vegetables, fresh herbs, fungi, stem vegetables, pine nuts, root vegetables and potatoes	0.050
2.6.	Leaf vegetables, fresh herbs, cultivated fungi and celeriac	0.20

2.7.	Stem vegetables, root vegetables and potatoes, excluding celeriac. For potatoes the maximum level applies to peeled potatoes	0.10
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Some of the heavy metals effects on the fauna and microfauna in soils and on cultivated and spontaneous plants were presented here.

MATERIAL AND METHOD

The research material consisted in soil, vegetation and litter samples collected from the same stand from those approximately 30 permanent sample surface plots set 25-35 years ago as well as soil and vegetation samples which have been collected from people's vegetables gardens in 12 localities – Copșa Mică area, this year (fig.1, table 3 and 4).

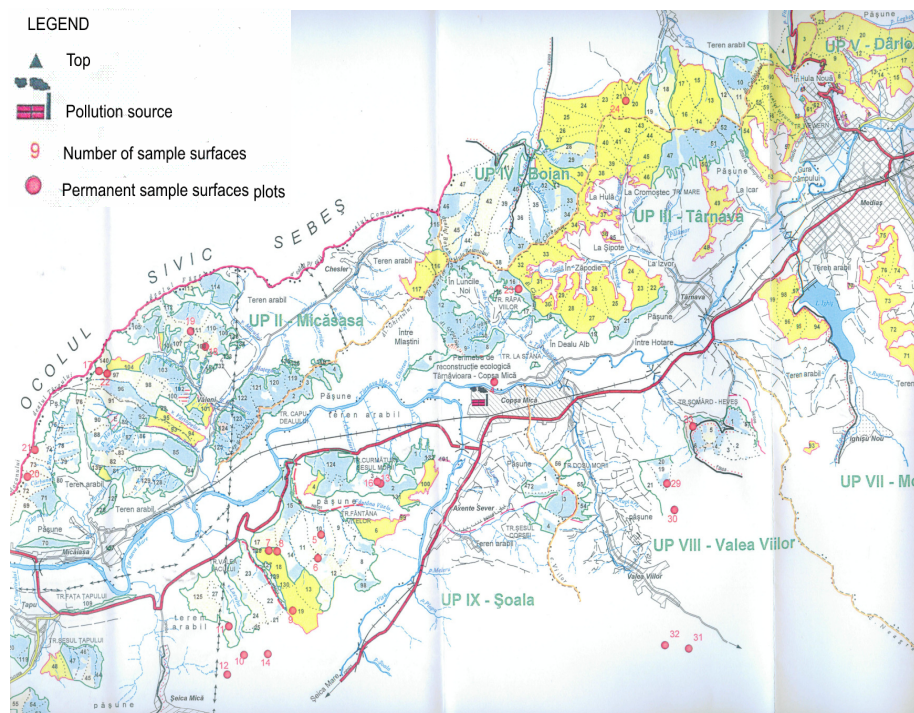


Fig. 1 The map of Copșa Mică area with permanent sample surface plots

Heavy metals (Cu, Pb, Cd, Zn, Mn), sulphur, calcium, magnesium, phosphorus, potash, nitrogen, total humus and other compounds samples have been determined from soil and vegetables samples.

The following determinations have been done by specific laboratory methods, namely:

- *in soil samples*: pH in aqueous suspension; organic matter contents, by Walkley-Black method, modified by Gogoășă; sulphur as SO_4 in aqueous extract; extractive aluminum, Coloman method in KCl 1N; granulometric practical size distribution by the dropping method; soluble P and K by Egner Riem Domingo method (double ammonium lactate); mobile Ca and Mg in 0.5 N NaCl , flamephotometric; total N by Kjeldahl method; heavy metals (Cu, Pb, Zn, Cd, Mn) were dosed as total and soluble forms by atomic absorption spectrophotometric method. The total forms in HCl solution were obtained by disintegration with a mixture of strong acids (HNO_3 , H_2SO_4 , HClO_4) and the soluble ones by extraction (Cu, Zn and Mn in DTPA, pH 7,3, and Pb and Cd in CH_3COOH);

- *in the dry matter*: of vegetation samples: heavy metals (Cu, Pb, Zn, Cd, Mn) by atomic absorption spectrophotometric method, sulphur as SO_4 by granulometric method, N by Kjeldahl method; P and K by Egner Riem Domingo method, Ca and Mg, flame-photometric method.

Table 3

The dynamics of pH and concentrations of heavy metals (Pb, Cu, Zn) in Ao horizon of soils in the stands under the influence of pollution in Copșa Mică area
(Ianculescu et al., 2009)

Production unit P.U.	Forest management unit FMu	12 km distance from the source	pH		Heavy metals concentrations, in ppm					
			1985	2006	Pb, total		Cu, total		Zn, total	
					1987	2006	1987	2006	1987	2006
I	2	3	4	5	6	7	8	9	10	11
I	1A	3.0	4.32	3.9	211.0	6663.0	27.9	458.3	375.0	1560
	13A	7.0	4.2	5.2	20.0	453.4	12.5	52.3	40.0	271.4
	30A	7.5	4.6	5.5	59.1	362.5	11.3	49.0	118.6	235.0
	37B	17.0	5.3	5.2	31.5	252.1	6.0	36.6	58.5	126.2
	84A	14.0	4.0	4.3	106.0	978.1	17.5	92.7	197.0	485.1
II	104F	12.0	4.4	4.9	21.3	493.6	16.9	51.5	51.6	273.8
III	122C	6.5	3.8	3.7	490.0	2086.0	61.4	186.6	770.0	780.5
	17A	2.0	3.95	5.8	116.0	4262.0	33.0	467.4	240.0	1672.0
IV	18B	1.8	4.0	4.2	249.7	924.5	47.7	415.9	692.0	1097.0
	17E	16.0	4.4	5.5	114.0	295.0	21.3	466.7	221.0	265.3
V	79E	22.0	7.0	5.5	33.0	269.3	21.0	64.2	56.0	173.6
VII	91E	17.0	5.3	5.9	56.0	258.7	18.5	55.6	142.0	174.7
	3A	17.0	4.1	3.1	73.9	1103.0	15.8	127.1	112.3	446.3
	46A	6.0	4.2	5.3	93.3	543.9	10.5	64.2	156	309.3
	99B	5.0	3.7	4.3	1116.0	4069.0	463.0	271.3	1148	983.1
VIII	46A	6.0	4.2	5.3	93.3	543.9	10.5	64.2	156.0	309.3
IX	4E	3.0	4.3	4.2	94.0	1659.0	17.4	16.7	199.0	1339.0
VIII	6A	3.6	5.8	4.6	84.0	1000.0	20.7	81.9	410.0	475.6

Table 4

The dynamics of heavy metals concentrations (Pb, Cu, Zn, Cd) in litter and vegetation for some stands under pollution influence, Copsa Mică area (Ianculescu et al., 2009)

Production unit P. U.	Forest management unit F/Mu	12 km distance from the source	Litter						Vegetation					
			Concentrations in ppm						Concentrations in ppm					
			Pb		Cu		Zn		Pb		Cu		Zn	
I	30A	7.5*	1987	2006	1987	2006	1987	2006	1987	2006	1987	2006	1987	2006
			371.0*	62.2	34.9	10.8	490.0	472.0	52.8	3.3	18.4	5.7	97.6	41.1
			96.0	49.7	27.4	21.0	330.0	562.0						
II	84A	14.0*	1055.0	195.8	73.0	5.7	501	2011.0	469.0	9.1	28.6	4.0	430.0	79.0
			3001.0	367.7	252.0	127.3	537	2409.0						
	104F	12.0	1472.0	179.7	113.0	37.1	1652.0	1280.0	111.0	6.0	26.3	7.5	215.0	67.1
			1262.0	210.0	98.0	59.4	1410.0	1726.0						
	122C	6.5	3550.0	389.4	805.0	121.9	2010.0	2600	356.0	15.6	36.9	5.8	580.0	72.8
			3510.0	656.0	438.0	270.9	1790.0	2710						
III	18B	1.8	2720.0	577.0	575.0	196.5	1850.0	2994.0	555.0	28.2	70.1	8.3	800.0	149.4
			2020.0	1080.0	415.0	402.3	1810.0	3488.0						
IV	17E	16.0	499.0	61.9	11.0	10.5	481.0	358.0	16.0	3.0	9.4	6.4	40.0	28.6
			-	157.8	-	40.5	-	963.0						
V	79E	22.0	8.0	2.1	19.3	1.3	32.0	159.0	6.3	2.1	10.4	6.2	28.0	25.3
			-	9.6	-	1.4	-	196.0						
	91E	17.0	146.0	36.9	11.0	7.5	547.0	268.0	35.0	2.4	11.0	5.1	61.0	35.4
			-	58.9	-	20.6	-	314.0						
VII	3A	17.0	1620.0	294.7	116.0	130.0	1450.0	2194.0	143.0	5.3	34.2	4.7	145.0	46.1
			663.0	-	74.8	-	698.0	-						
VIII	6A	3.8	1380.0	408.7	56.5	109.0	1480.0	2386.0	22.0	4.2	5.1	31.4	106.0	28.5
			350.0	202.6	96.0	64.9	920.0	1666.0						
	46A	6.0	146.0	129.9	11.0	15.6	547.0	951.0	35.0	2.7	11.0	2.2	61.0	25.5
			-	323.9	-	54.2	-	2029.0						
IX	4E	3.0	1850.0	332.3	145.0	85.2	1600.0	2292.0	72.0	7.4	25.2	107.2	206.0	46.7
			835.0	673.3	111.0	208.3	1400.0	2939.0						

$$* \frac{371.0}{96.0} - \frac{0I_1}{0I_2} = \frac{\text{undecomposed leaves}}{\text{decomposed leaves}}$$

RESULTS

Tables 3 and 4 present the dynamics of heavy metals contents (Cu, Pb, Zn, Cd) in A₀ horizon of soils, and stand litter and vegetation samples from the permanent sample surface plots in Copșa Mică area (fig. 1).

The graphs of figures 2, 3 and 4 present the dynamics of Pb concentrations in A₀ horizon in the stand litter and vegetation from the permanent sample surface plots.

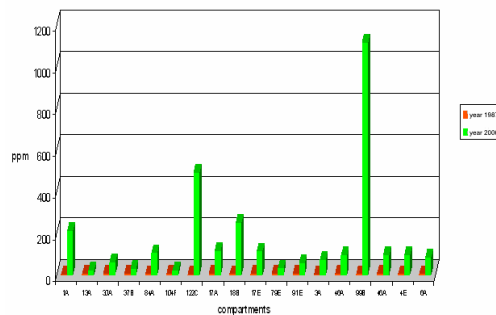


Fig. 2. The dynamics of Pb concentration in A₀ horizon of soils of some forest stands in Copșa Mică area

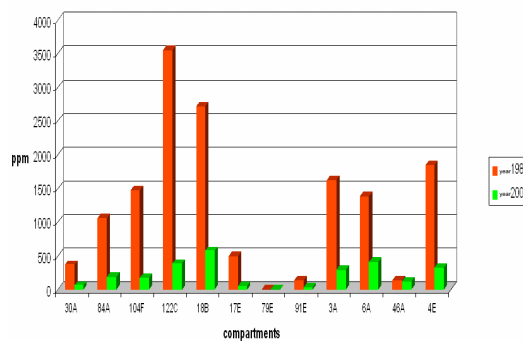


Fig.3. The dynamics of Pb concentrations in the litter of the forest stands in the permanent sample surface plots in Copșa Mică area

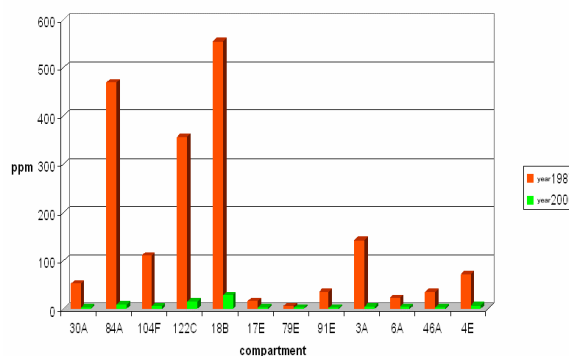


Fig.4. The dynamics of Pb concentrations in the vegetation samples from the forest stands in the permanent sample surface plots in Copșa Mică area

On one hand, the results point out the capacity of soils to accumulate large quantities of noxae which exceed by far the admissible limits and on the other hand the remanence capacity in soil especially for heavy metals (Pb, Cu, Zn, Cd) extremely dangerous for living creatures. The same regularity is obeyed for all stands from permanent sample surface plots in Copșa Mică area (Ianculescu et al, 2009) namely: considering that about 25-30 years ago, the contents in heavy metals and sulphur compounds from litter and vegetation recorded very high levels by far over the admitted maximum limits being in accordance with their concentrations in the atmosphere, on the contrary in soils A_0 horizon the respective noxae concentrations were in the maximum admitted limits because the pollution sources had been working for a recent period of time (about 10-15 years); investigating again after 25-30 years on the same permanent sample surface plots and with the same harvesting methods, the same period of time to harvest, namely August, and the same methods of determination, the results obtained were reverse: the contents in heavy metals and sulphur compounds in litter and vegetation are in the admitted limits being in accordance with their concentrations in the atmosphere (owing to the industrial capacities which reduced their activity and their release by about 150 m. high chimneys). On the contrary, in A_0 horizons of soils because of their accumulation and remanence, the contents record high values by far their admitted limit (Ianculescu et al, 2008, 2009).

Feeling alarmed about the high levels of contents, especially of Cd and Pb, in the forestry ecosystems, extremely toxic for animals and human beings health, we extended the investigations out of the forestry fund, namely in the population's gardens in Copșa Mică area in the conditions where the industrial capacities of S.C. SOMETRA S.A were ceased, the enterprise which was guilty of the pollution in that area. We have to mention that S.C. CARBOXIM S.A., the producer of carbon black from methane gas, a less toxic noxa, was closed in 1992.

The results of soil and vegetation samples analyses from the population's vegetables gardens, Copșa Mică area, are presented in tables 5 and 6 and figures 5-27 show Cd and Pb contents considered to be the most toxic in soil and vegetables.

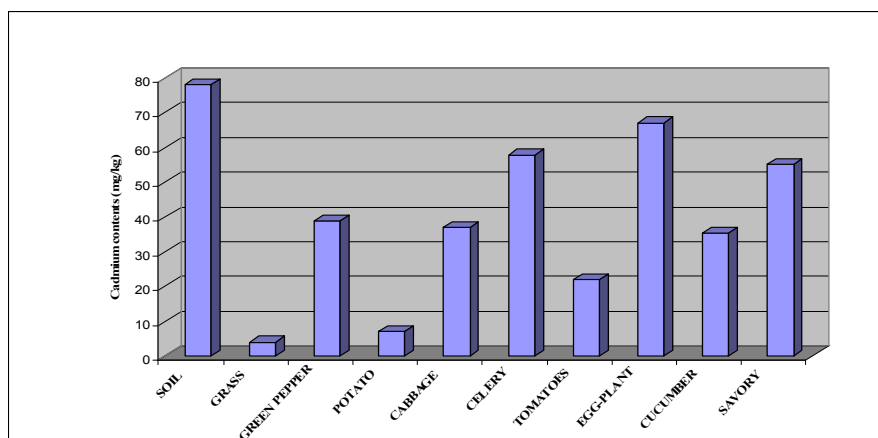


Fig. 5. The contents of cadmium from some samples taken from soil and some cultivated plants from population's gardens, in Copșa Mică, in August 2010

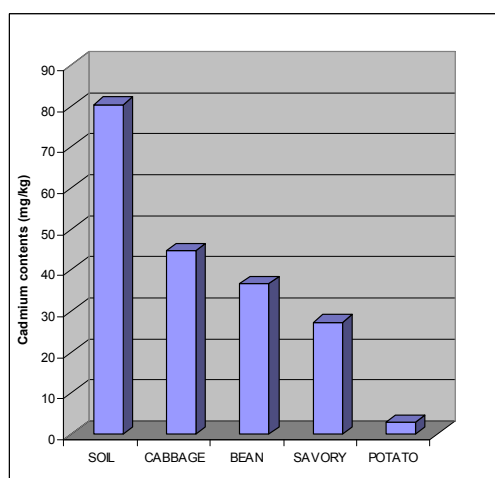


Fig. 6. The contents of cadmium from some samples taken from soil and some cultivated plants from population's gardens, in Târnăvioara, in August 2010

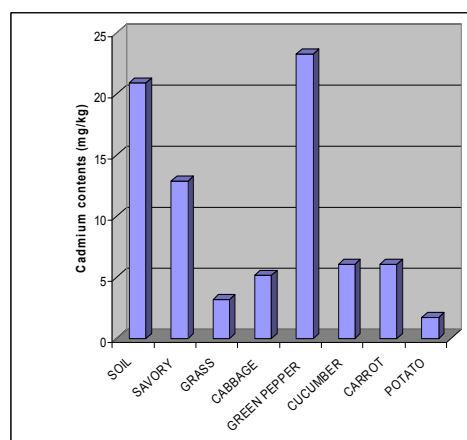


Fig. 7. The contents of cadmium from some samples taken from soil and some cultivated plants from population's gardens, in Micăsasa, in August 2010

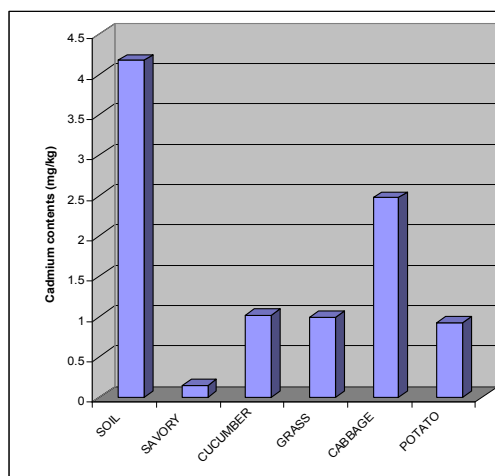


Fig. 8. The contents of cadmium from some samples taken from soil and some cultivated plants from population's gardens in Valea Lungă in August 2010

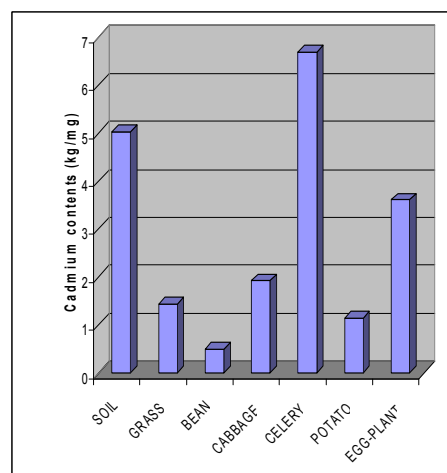


Fig. 9. The contents of cadmium from some samples taken from soil and some cultivated plants from population's gardens, in Seica Mică, in August 2010

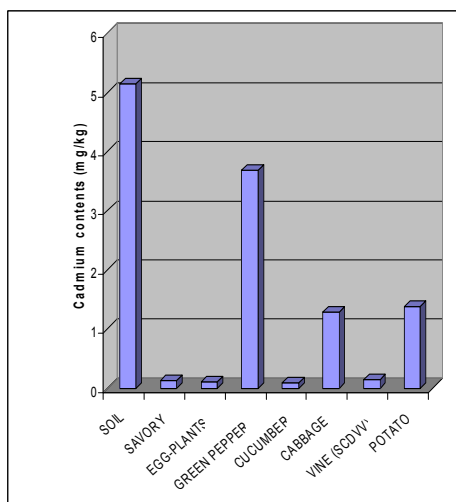


Fig. 10. The contents of cadmium from some samples taken from soil and some cultivated plants from population's gardens, in Blaj, in

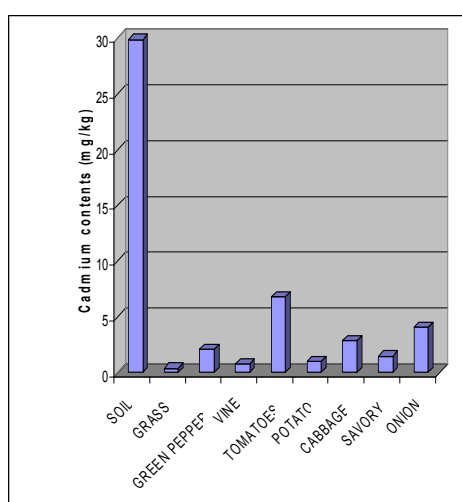


Fig. 11. The contents of cadmium from some samples taken from soil and some cultivated plants from population's gardens, in Târnava, in

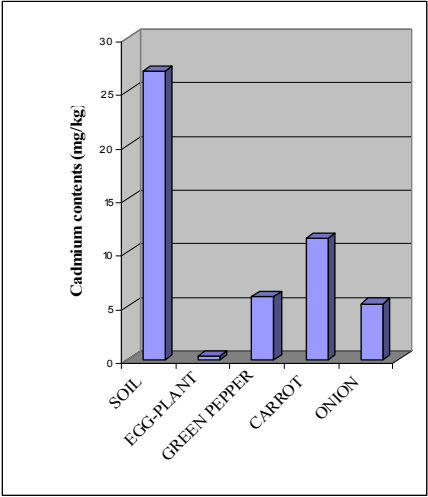


Fig. 12. The contents of cadmium from some samples taken from soil and some cultivated plants from population's gardens, in Axente Sever, in August

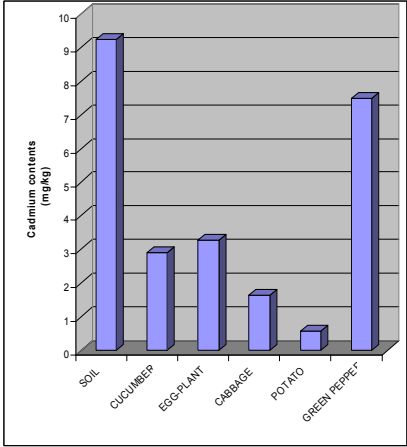


Fig. 13. The contents of cadmium from some samples taken from soil and some cultivated plants from population's gardens, in Valea Viilor, in August 2010

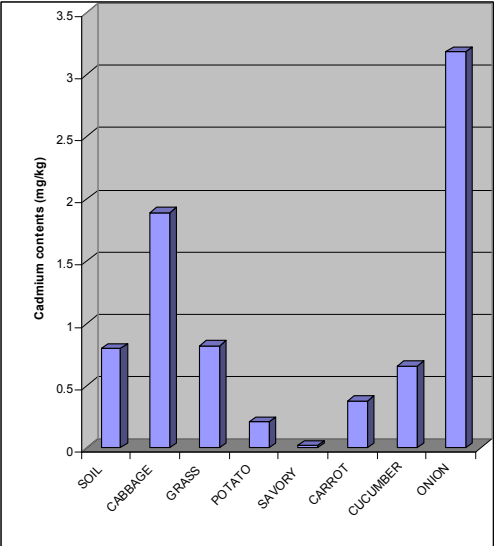


Fig. 14. The contents of cadmium from some samples taken from soil and some cultivated plants from population's gardens, in Bazna, in August 2010

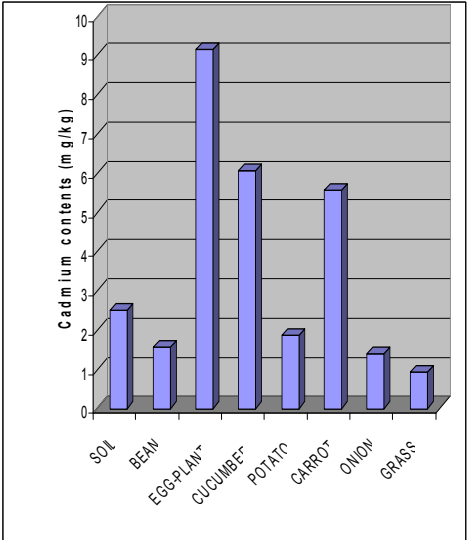


Fig. 15. The contents of cadmium from some samples taken from soil and some cultivated plants from population's gardens, in Mediaș

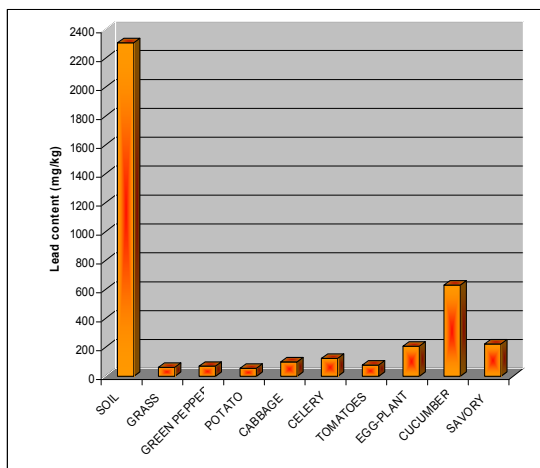


Fig. 16. The contents of lead (Pb) from some samples taken from soil and some cultivated plants from population's gardens, in Copșa Mică,

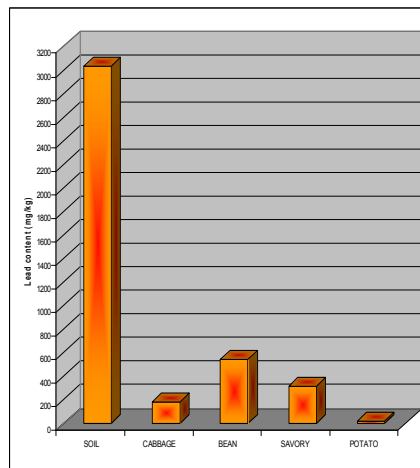


Fig. 17. The contents of lead (Pb) from some samples taken from soil and some cultivated plants from population's gardens, in

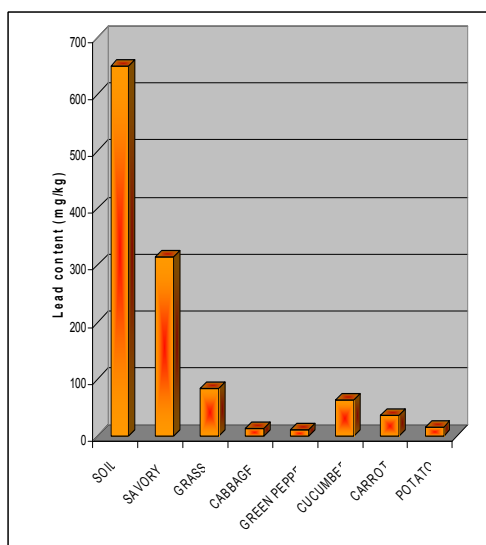


Fig. 18. The contents of lead from some samples taken from soil and some cultivated plants from population's gardens, in Micăsasa, in

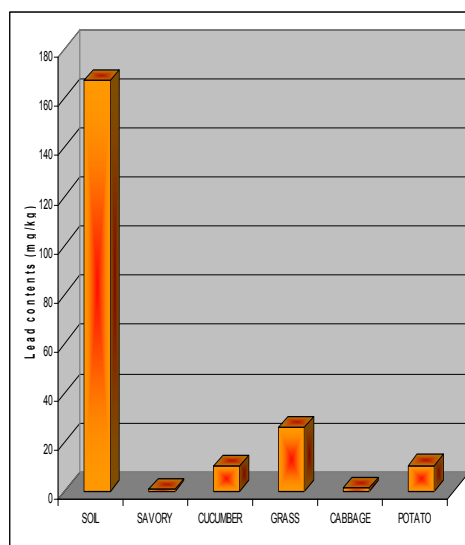


Fig. 19. The contents of lead from some samples taken from soil and some cultivated plants from population's gardens, in Valea Lungă,

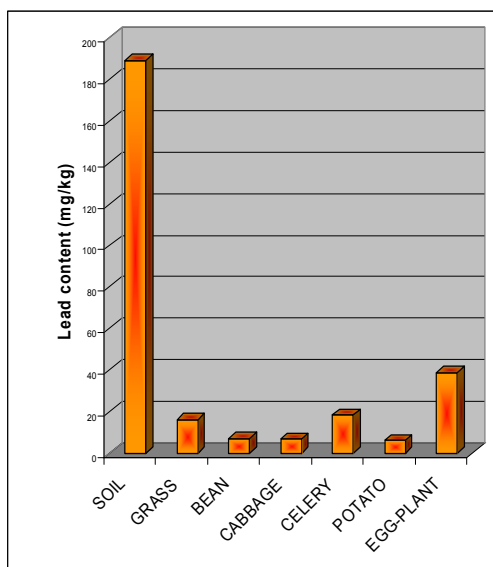


Fig. 20. The contents of lead from some samples taken from soil and some cultivated plants from population's gardens, in Seica Mică, in

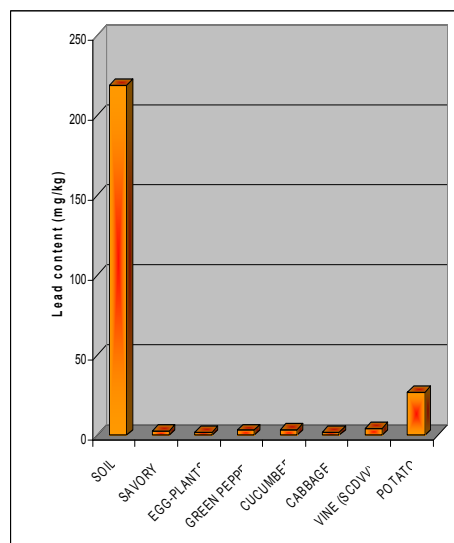


Fig. 21. The contents of lead from some samples taken from soil and some cultivated plants from population's gardens, in Blaj, in August 2010

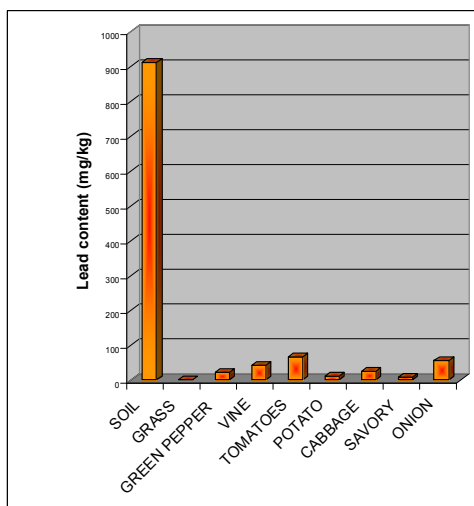


Fig. 22. The contents of lead from some samples taken from soil and some cultivated plants from population's gardens, in Târnava, in August 2010

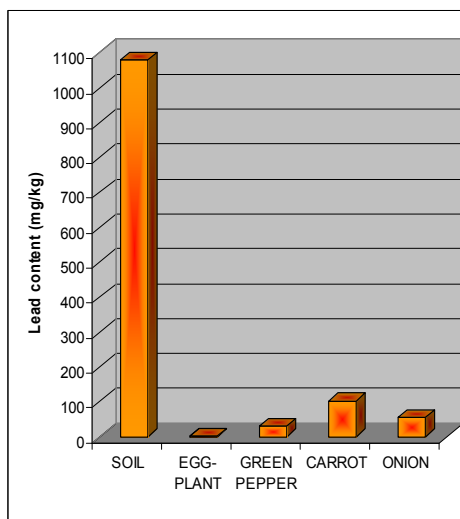


Fig. 23. The contents of lead from some samples taken from soil and some cultivated plants from

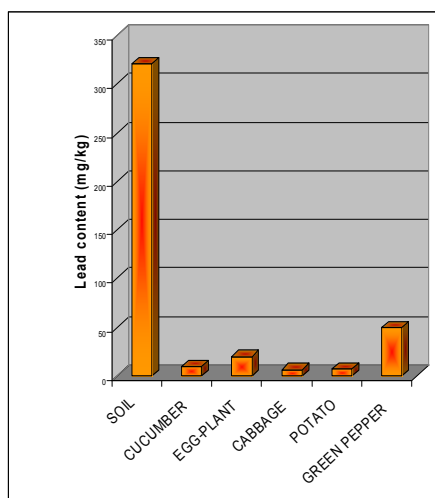


Fig. 24. The contents of lead from some samples taken from soil and some cultivated plants from population's gardens, in Valea Viilor, in August 2010

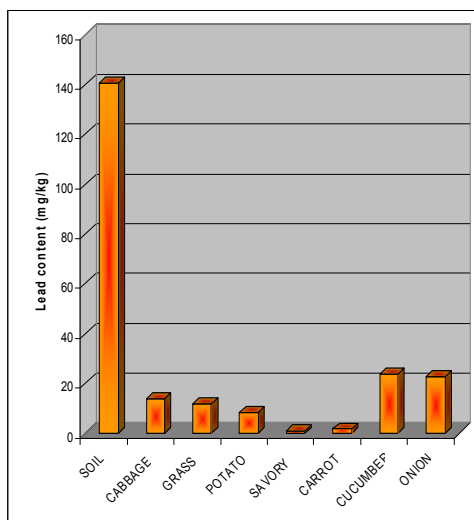


Fig. 25. The contents of lead from some samples taken from soil and some cultivated plants from population's gardens, in Bazna, in August 2010

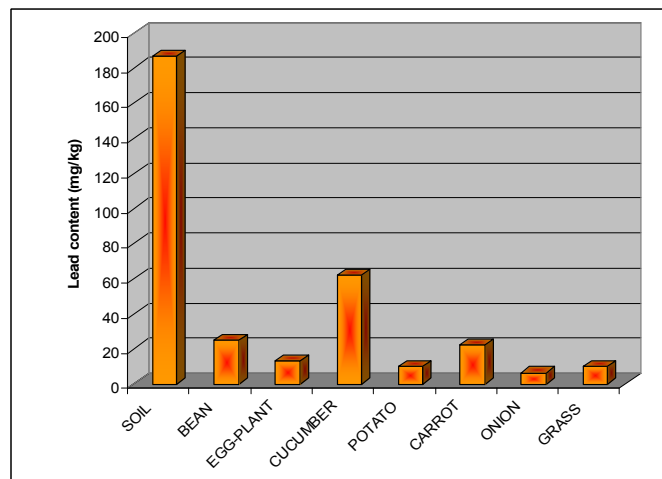


Fig. 26. The contents of cadmium from some samples taken from soil and some cultivated plants from population's gardens, in Mediaș (Headquarter Forest District) in August 2010

The concentrations of noxae in soil (table 5) have values by far higher than the admitted limit, as it was said above, because of the accumulation and remanence phenomena in all those 11 places where samples were collected, some of them being considered as control areas regarding the large distances from the pollution source, behaviour and

morphologic aspect of the forestry vegetation (to see the points in Blaj, Bazna, Valea Viilor, Șeica Mică, Mediaș - the headquarters of the forest district).

Considering the conditions where the pollution source was ceased beginning with 2009, the samples of vegetables and grass coming from these 11 points contain in all these points quantities over the admitted limit (table 6). If we refer to the admitted limits in various countries and the European Union Directives, we find out that the population of tens even hundreds of thousand inhabitants of Copșa Mică consume daily vegetables, fruits, cheese, dairy products with a high concentration of heavy metals especially Cd and Pb, which are considered to be cancerogenic. A few data about the European Union norms regarding the maximum admitted doses of Cd are extremely alarming. Thus, the weekly admitted dose of Cd per person is 0.007 mg/kg while in the population's gardens from Copșa Mică the concentrations were 67.06 mg/kg in egg plants, 57.83 mg/kg in celery, 38.86 mg/kg in green peppers, 37.04 mg/kg in cabbage, 21.95 mg/kg in tomatoes, 35.39 mg/kg in cucumbers, examples of the most utilized vegetable by people (table 6, fig. 6).

Table 5

Chemical analyses of some soil samples (0–10 cm) taken from population's gardens from Copșa Mică area (total forms)

Data to identify laboratory assays		Distance from the pollution source	Locality	Depth	N %	pH	Carbo nats	Ht %	K ppm	Ca ppm	Mg ppm	Mn ppm	Ni ppm	Cd ppm	Cu ppm	Fe ppm	Pb ppm	Zn ppm	P ppm
No.	No. Lab																		
Total forms																			
1	75	0.5 km	Copșa Mică	A ₀ (0-10)	0.135	7.03	0.5	2.59	3455	4022	64.30	9.10	102.8	78.16	397.1	54.49	2307	174.5	14.86
2	74	0.6 km	Tarnavioara	A ₀ (0-10)	0.137	7.28	2.9	2.60	1853	3216	58.89	7.40	91.3	80.36	397.0	49.43	3036	178.7	13.63
3	81	8.0 km	Micasasa	A ₀ (0-10)	0.146	7.96	4.7	2.57	2749	6233	92.82	9.80	112.1	20.90	257.8	20.17	650.3	58.7	8.73
4	78	13.0 km	Valea Lunga	A ₀ (0-10)	0.202	8.05	6.7	3.95	3054	687	100.7	12.13	148.6	4.17	589.9	34.98	167.0	27.8	11.47
5	77	9.0 km	Seica Mica	A ₀ (0-10)	0.107	7.87	0.4	2.06	1511	2261	58.89	7.02	86.0	5.03	211.2	49.51	188.9	23.3	12.47
6	80	18.0 km	Blaj	A ₀ (0-10)	0.241	7.20	0.8	4.64	2922	1954	84.17	13.42	132.9	5.14	241.6	31.95	219.1	32.5	26.43
7	76	4.0 km	Tarnava	A ₀ (0-10)	0.252	7.75	2.5	4.58	3190	5301	87.55	11.14	137.0	29.92	374.1	29.00	911.7	73.8	24.04
8	72	1.5 km	Axente Sever	A ₀ (0-10)	0.174	6.78	0.9	2.93	1885	4331	66.24	10.21	107.8	26.93	369.5	25.65	1080.0	89.4	24.35
9	73	4.0 km	Valea Viilor	A ₀ (0-10)	0.193	7.34	2.5	3.62	1753	3290	63.61	9.91	95.4	9.24	192.9	48.12	320.7	37.6	15.03
10	79	7.0 km	Bazna	A ₀ (0-10)	0.196	7.73	0.4	3.82	3123	4610	96.65	13.14	167.9	0.80	258.3	10.94	140.7	23.7	12.75
11	71	9.0 km	Mediaș (The headquarter of the forest district)	A ₀ (0-10)	0.129	6.39	-	2.48	1700	4727	60.74	13.61	120.2	2.49	129.5	27.43	186.9	19.8	9.16
12	82	18.0	SCDVV Blaj	A ₀ (0-10)	0.157	7.31	1.4	3.02	2359	5616	93.15	13.43	160.6	3.84	689.5	35.51	218.1	28.4	12.41

Table 6

Analyses of some samples taken from cultivated plants in the population's gardens in Copșa Mică area in August 2010 (total forms)

Data to identify laboratory assays	No.	No. lab	Locality	Distance from the pollution source	Species	S %	K mg/kg	Ca mg/kg	Mg mg/kg	Mn mg/kg	Cd mg/kg	Cu mg/kg	Fe mg/kg	Pb mg/kg	Ni mg/kg	Zn mg/kg	P mg/kg
1	83		BLAJ	18.0 km	SAVORY	0.14	151.60	3525.00	4468.00	36.00	0.12	73.79	1186.00	3.05	3.33	87.00	4211.00
2	84				EGG-PLANT	0.22	144.70	4154.00	4215.00	30.00	0.10	68.80	818.20	2.27	2.28	30.30	4610.00
3	85				GREEN PEPPER	0.40	235.00	3789.00	5353.00	154.00	3.68	42.93	3203.00	3.21	15.11	67.20	3839.00
4	86				CUCUMBER	0.56	173.00	1404.00	9010.00	90.00	0.09	58.32	1186.00	3.91	3.76	71.00	3256.00
5	87				CABBAGE	1.51	119.20	2603.00	6334.00	40.00	1.28	53.66	171.40	1.87	2.66	75.50	3941.00
6	88				VINE	0.22	40.11	2449.00	2825.00	121.80	0.15	552.40	160.80	4.57	4.61	30.00	2677.00
7	89				POTATO	0.12	130.40	5952.00	2551.00	117.10	1.38	43.12	3066.00	27.03	16.52	55.00	4322.00
8	90		TÂRNĂVOARA	0.6 km	CABBAGE	2.04	97.35	981.60	5808.00	99.70	44.79	123.20	1077.00	182.60	2.51	900.70	5054.00
9	91				BEAN	0.21	97.35	3347.00	4121.00	208.20	36.65	190.50	6352.00	548.00	27.73	900.90	3325.00
10	92				SAVORY	0.37	69.70	2039.00	3500.00	72.70	27.24	94.43	900.40	319.30	10.83	573.40	3278.00
10 bis	92 bis				POTATO	0.15	46.17	613.60	1134.00	14.10	2.83	36.27	245.90	25.33	2.40	46.10	3169.00
11	93		ȘEICA MICĂ	9.0 km	GRASS	0.31	97.70	5053.00	2679.00	131.10	1.44	49.55	1764.00	16.21	11.94	61.60	4217.00
12	94				BEAN	0.16	102.30	2230.00	3088.00	45.80	0.51	32.07	155.70	7.20	2.92	27.90	3286.00
13	95				CABBAGE	0.87	85.63	762.00	2942.00	63.00	1.93	49.81	535.80	6.78	2.62	35.50	4885.00
14	96				CELERY	1.19	86.98	6840.00	2252.00	116.20	6.70	340.80	1743.00	18.71	8.09	136.40	3105.00
15	97				POTATO	0.19	64.01	842.10	1718.00	23.10	1.14	33.86	437.70	6.24	3.32	23.20	3729.00
16	98				EGG-PLANT	0.20	71.29	3423.00	2724.00	162.50	3.61	547.20	5117.00	38.93	24.44	77.80	3411.00
17	99		MEDIAȘ (The headquarter of the forest district)	9.0 km	BEAN	0.21	53.65	3508.00	3800.00	229.20	1.55	988.00	2582.00	24.76	14.61	68.90	3816.00
18	100				EGG-PLANT	0.40	80.66	4007.00	3716.00	299.50	9.15	456.30	1422.00	13.26	16.02	67.10	5585.00
19	101				CUCUMBER	0.16	40.31	853.80	8472.00	392.10	6.07	205.20	2244.00	62.19	38.98	128.90	8233.00
20	102				POTATO	0.11	42.24	1200.00	1402.00	53.10	1.87	129.90	1063.00	10.15	10.38	27.60	2425.00
21	103				CARROT	0.13	56.75	5150.00	2278.00	119.50	5.55	54.81	3041.00	22.42	35.72	56.90	4584.00
22	104				ONION	0.34	38.07	3731.00	1179.00	26.50	1.38	28.77	232.90	6.42	6.32	36.50	4041.00
23	105		MICĂSASA	8.0 km	GRASS	0.26	2252.00	4590.00	1885.00	109.80	0.92	30.36	980.80	9.98	8.66	41.10	3388.00
24	106				SAVORY	0.19	1502.00	2394.00	4610.00	276.10	12.88	129.10	4514.00	315.60	28.84	330.10	2832.00
25	107				GRASS	0.23	1597.00	5841.00	2209.00	163.70	3.20	51.73	3270.00	83.94	10.58	96.30	2944.00
26	108				CABBAGE	0.77	3463.00	475.00	3214.00	90.00	5.16	19.14	346.80	13.09	0.56	123.00	4857.00
27	109				GREEN-PEPPER	0.54	768.30	2991.00	5552.00	53.10	23.28	72.41	322.00	11.89	1.15	195.20	4973.00
28	110				CUCUMBER	0.60	565.00	3348.00	6912.00	102.00	6.11	61.16	1581.00	63.75	2.45	111.90	5154.00
29	111		VALEA LUNGĂ	13.0 km	CARROT	0.15	2860.00	5100.00	1937.00	47.10	6.10	52.96	1285.00	37.51	12.08	60.00	3374.00
30	112				POTATO	0.12	1959.00	1141.00	1476.00	24.40	1.70	40.43	329.70	17.27	1.78	36.00	2184.00
31	113				SAVORY	0.27	87.37	2704.00	405.00	30.00	0.15	4.14	370.00	0.97	1.93	15.00	1950.00
32	114				CUCUMBER	0.33	1845.00	2993.00	4155.00	117.10	1.02	279.20	1924.00	10.37	7.50	56.30	4096.00
33	115				GRASS	0.28	597.30	3599.00	7658.00	232.70	0.99	190.60	4340.00	26.05	15.08	78.90	8770.00
34	116				CABBAGE	1.25	1883.00	2744.00	3606.00	173.50	2.47	46.91	216.40	1.47	0.25	41.50	4157.00
35	117				POTATO	0.10	430.90	4230.00	2559.00	129.20	0.93	49.75	1511.00	10.53	8.08	70.60	4422.00

Data to identify laboratory assays	No.	Locality	Distance from the pollution source	Species	S %	K mg/kg	Ca mg/kg	Mg mg/kg	Mn mg/kg	Cd mg/kg	Cu mg/kg	Fe mg/kg	Pb mg/kg	Ni mg/kg	Zn mg/kg	P mg/kg
36	118	AXENTE SEVER	1.5 km	EGG-PLANT	0.28	1494.00	2775.00	1212.00	34.50	0.32	43.28	928.50	3.00	3.93	20.70	1755.00
37	119			GREEN- PEPPER	0.36	598.80	5222.00	2243.00	73.00	5.90	156.90	593.40	30.51	2.62	47.20	4010.00
38	120			CARROT	0.11	441.60	3880.00	4625.00	187.10	11.30	467.00	4864.00	103.40	30.06	211.60	4526.00
39	121			ONION	0.36	422.90	5700.00	1994.00	54.20	5.26	48.68	1205.00	54.49	15.03	80.80	4588.00
40	122	BAZNA	7.0 km	CABBAGE	1.85	1841.00	3483.00	1392.00	32.40	1.89	26.78	236.50	13.83	0.58	71.60	4728.00
41	123			GRASS	0.21	3404.00	2677.00	3095.00	62.40	0.82	34.92	745.10	11.77	1.00	45.00	5492.00
42	124			POTATO	0.11	1693.00	6624.00	3207.00	125.40	0.21	82.80	3643.00	8.54	36.67	53.70	1794.00
43	125			SAVORY	0.36	1774.00	887.90	1527.00	30.30	0.02	46.39	733.30	0.97	2.77	16.20	2418.00
44	126			CARROT	0.10	2658.00	1718.00	5026.00	42.00	0.38	65.75	183.90	1.78	1.41	69.30	4267.00
45	127			CUCUMBER	0.19	2102.00	7161.00	2732.00	172.90	0.66	59.19	6438.00	23.60	55.96	61.80	3952.00
46	128			ONION	0.38	2591.00	867.30	6913.00	320.20	3.18	667.00	2437.00	22.74	37.50	118.90	8786.00
47	129	TÂRNAVA	4.0 km	GRASS	0.37	2752.00	4574.00	1273.00	31.00	0.39	43.57	634.10	1.92	5.71	34.30	3018.00
48	130			GREEN PEPPER	0.49	3080.00	5073.00	5767.00	141.00	2.10	53.40	449.00	21.27	3.85	85.40	6282.00
49	131			VINE	0.21	2755.00	2283.00	1313.00	86.80	0.77	29.90	184.90	41.86	0.71	40.60	5811.00
50	132			TOMATOES	0.88	496.90	4792.00	4234.00	607.00	6.76	526.00	2101.00	66.85	8.13	136.10	3998.00
51	133			POTATO	0.10	1875.00	1308.00	1203.00	18.80	0.98	41.08	291.20	10.44	0.08	23.30	2692.00
52	134			CABBAGE	1.64	684.00	3812.00	4640.00	89.90	2.89	41.11	355.90	26.37	1.17	104.30	4430.00
53	135			SAVORY	0.36	1496.00	5490.00	1251.00	22.70	1.50	22.52	206.30	8.36	0.18	33.50	3480.00
54	136	VALEA VIILOR	4.0 km	ONION	0.51	3251.00	2131.00	3487.00	398.30	4.06	368.00	502.20	55.00	2.60	117.60	3711.00
55	137			CUCUMBER	0.46	447.20	1057.00	9984.00	46.40	2.91	961.30	218.20	9.28	1.77	77.20	8910.00
56	138			EGG-PLANT	0.22	444.30	2396.00	3706.00	94.90	3.27	282.60	1200.00	19.95	8.61	61.50	4466.00
57	139			CABBAGE	1.49	3276.00	3377.00	3936.00	72.10	1.65	21.22	420.60	6.58	1.18	63.70	5138.00
58	140			POTATO	0.15	2659.00	1125.00	1319.00	21.70	0.59	35.19	277.30	7.76	1.47	37.80	4170.00
59	141			GREEN PEPPER	0.34	597.00	3390.00	4396.00	170.60	7.49	127.70	2714.00	49.96	17.09	167.50	3816.00
60	142	COPȘA MICĂ	0.5 km	GRASS	0.29	1432.00	6825.00	945.90	67.60	4.00	26.21	213.40	60.40	1.26	90.40	1782.00
61	143			GREEN PEPPER	0.57	692.50	2426.00	3495.00	68.70	38.86	2818.00	602.80	71.23	17.11	167.50	4986.00
62	144			POTATO	0.13	2486.00	1389.00	1640.00	27.40	6.89	56.72	492.80	54.44	3.21	94.60	4247.00
63	145			CABBAGE	1.89	2570.00	691.20	3977.00	173.50	37.04	426.40	847.60	95.70	3.51	303.10	4325.00
64	146			CELERY	1.37	2588.00	3115.00	4732.00	150.60	57.83	896.00	1628.00	119.70	16.60	433.30	9970.00
65	147			TOMATOES	0.60	3031.00	5000.00	3659.00	88.50	21.95	859.00	272.20	76.46	43.12	75.40	3385.00
66	148			EGG-PLANT	0.26	3049.00	2690	3570.00	114.80	67.06	187.8	1560.00	205.50	8.18	259.60	3390.00
67	149			CUCUMBER	0.42	1754.00	752.80	9631.00	303.10	35.39	753.00	6468.00	630.10	56.37	723.00	3504.00
68	150			SAVORY	0.31	2165.00	3840.00	4669.00	133.80	55.25	539.00	1035.00	223.20	6.81	604.30	4490.00

The same alarming Cd contents considered to be the most toxic of heavy metals followed by Pb are recorded at village Târnăvioara (table 6, figure 7) – 44.79 mg/kg in cabbage, 36.65 mg/kg in beans and 27.24 mg/kg in savory.

Regarding Cd admitted doses, EU norms provide for: cereals / 0.1 mg/kg, vegetables (leaves) – 0.01 mg/kg, vegetables celery leaves and cultivated mushrooms (0.2 mg/kg); vegetables – fruits– 0.05 mg/kg; vegetables (stem + roots) and for potatoes – 0.1 mg/kg (data offered by prof. Gheorghe Mencinicopschi). It is easy to notice that in comparison with these admitted doses, the results obtained from all these 11 localities show high overtaking of cadmium concentration.

DISCUSSIONS

Analysing the results obtained we find out high overtaking in heavy metals contents (Cd, Pb, Cu, Zn) in soils in comparison with the doses admitted by Directive 86/278/EEC (tables 1 and 6) in all these 11 localities in Copșa Mică area. Similarly, heavy metals contents in the analyzed soils, pass over the maximum admitted limits presented by Lindsay (1979) and Dumitru (1990) and the things presented in the introduction of this paper.

Table 7 presents a hierarchy of vegetables species considering Cd and Pb contents absorbed from the soils samples taken from the 11 households. The highest capacity storing heavy metals species proved to be the green pepper, eggplants, cabbage, cucumbers, celery with some differences in hierarchy, according to the locality. The lower affinity for heavy metals contents in soils (Cd, Pb, Cu, Zn) was found out in potatoes and carrots (Leonard et al, 2007).

Analysing the way the vegetables species grade regarding the heavy metals absorbing capacity we find out that generally in each locality the hierarchy is the same considering Cd and Pb (table 7).

Table 7

Hierarchy of species considering Cd and Pb contents assimilated in soil

No.	Locality	Species	Cadmium (Cd) contents mg/kg	Species	Lead (Pb) contents mg/kg
1.	Copșa Mică	egg-plant	67.06	cucumber	630.10
		celery	57.83	savory	223.20
		savory	55.25	egg-plant	205.50
		green pepper	38.86	celery	119.70
		cabbage	37.04	cabbage	95.70
		cucumber	35.39	green pepper	71.23
		tomatoes	21.95	tomatoes	76.46
		potato	6.89	potato	54.44
		grass	4.00	grass	60.40
2.	Târnăvioara	cabbage	44.79	bean	548.00
		bean	36.65	savory	319.30

No.	Locality	Species	Cadmium (Cd) contents mg/kg	Species	Lead (Pb) contents mg/kg
		savory	27.24	cabbage	182.6
		potato	2.83	potato	25.33
3.	Micăsasa	green pepper	23.28	savory	315.60
		savory	12.88	grass	83.94
		cucumber	6.11	cucumber	63.75
		carrot	6.10	carrot	37.51
		cabbage	5.16	potato	17.27
		grass	3.20	cabbage	13.09
		potato	1.70	green pepper	11.89
4.	Valea Lungă	cabbage	2.47	grass	26.05
		cucumber	1.02	cabbage	10.53
		grass	0.99	potato	10.53
		potato	0.93	cucumber	10.37
		savory	0.15	savory	0.97
5.	Șeica Mică	celery	6.70	egg-plant	38.93
		egg-plant	3.61	celery	18.71
		cabbage	1.93	grass	16.21
		grass	1.44	bean	7.20
		potato	1.14	cabbage	6.78
		bean	0.51	potato	6.24
6.	Blaj	green pepper	3.68	potato	27.03
		potato	1.38	vine	4.57
		cabbage	1.28	cucumber	3.91
		vine	0.15	green pepper	3.21
		savory	0.12	savory	3.05
		egg-plant	0.10	egg-plant	2.27
		cucumber	0.09	cabbage	1.87
7.	Târnava	tomatoes	6.76	tomatoes	66.85
		onion	4.06	onion	55.00
		cabbage	2.89	vine	41.86
		green pepper	2.10	cabbage	26.37
		savory	1.50	green pepper	21.27
		potato	0.98	potato	10.44
		vine	0.77	savory	8.36
		grass	0.39	grass	1.92
8.	Axente Sever	carrot	11.30	carrot	103.40
		green pepper	5.90	onion	54.49
		onion	5.26	green pepper	30.51
		egg-plant	0.32	egg-plant	3.00
9.	Valea Viilor	green pepper	7.49	green pepper	49.96
		egg-plant	3.27	egg-plant	19.95
		cucumber	2.91	cucumber	9.28
		cabbage	1.65	potato	7.76
		potato	0.59	cabbage	6.58
10.	Bazna	onion	3.18	cucumber	23.60
		cabbage	1.89	onion	22.74
		grass	0.82	cabbage	13.83
		cucumber	0.66	grass	11.77
		carrot	0.38	potato	8.54
		potato	0.21	carrot	1.78
		savory	0.02	savory	0.97
11.	Mediaș	egg-plant	9.15	cucumber	62.19
		cucumber	6.07	bean	24.76
		carrot	5.55	carrot	22.42
		potato	1.87	egg-plant	13.26
		bean	1.55	potato	10.15

No.	Locality	Species	Cadmium (Cd) contents mg/kg	Species	Lead (Pb) contents mg/kg
		onion	1.38	onion	6.42
		grass	0.92	grass	9.98

Considering soils high capacity in storing heavy metals as well as their remanence in soil, the situation is getting extremely alarming for peoples and animals' health in Copșa Mică area even in the conditions of reducing, in admissible limits, noxae concentrations in the atmosphere or even after ceasing the work of industries generators of the environment pollution. The results obtained after these investigations represent an alarm for the stages that should be done even when the activity of those factories ceased.

A high necessity appears in the idea to keep on the investigations especially in the areas with non-ferrous metals metallurgy (Copșa Mică, Zlatna, Baia Mare) in order to find the most indicated technology to detoxicate the polluted soils and their efficient utilization in the benefit of the society. This thing is possible by developing some interdisciplinary research in a research project financed by EU. It is known that the recordings by satellite in 1986 indicate that Copșa Mică was the most polluted area in the world (Ianculescu et al, 2009).

Considering this improvement, visible by satellite images, the results of the investigations presented in this paper are highly alarming for people and animals' health representing a high risk for food security.

CONCLUSIONS

The investigations carried out in the stand under the influence of air pollution in Copșa Mică area pointed out high accumulations of heavy metals (Cd, Pb, Cu, Zn) within time, in A₀ horizon of soils. Even if during the latest period of time after 1990 there have been reduced the industrial capacities generators of excessive pollution – existing for more than 25 years, at present the situation in Copșa Mică area is extremely alarming for food biosecurity of the population in the area. The extension of the investigations out of the forest stock in the population's vegetables gardens from 11 localities in Copșa Mică area pointed out the contents of heavy metals (Cd, Pb, Cu, Zn), much higher than the admitted limits by EU Directive nr. 86/278/EEC and EC Regulation No. 1881/ 2006 both in the superior horizon of soils and in the vegetables cultivated for ensuring the population's food.

Taking into account the extremely serious situation at present, it is necessary to draw the attention to both local decisional factors (from

townhalls, prefect's offices, authorities responsible for the environment, silviculture, agriculture and food industry) and those at the central level (Presidency, Parliament, Government) to take all measures recommended after the complex investigations in such areas in order to detoxicate the soils which are not proper for obtaining the food for the population's nutrition.

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