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Heavy metal dispersion detected in soils and plants alongside roads in Hungary

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ABSTRACT

Soil and plant samples from roadside areas contaminated with toxic by-products of vehicular traffic (for example, lead compounds from leaded fuel, metal contaminants derived from spare parts) were analysed in the case of five Hungarian sampling sites. The analyses were carried out with a new analytical method which is based on the use of a high performance sample introduction unit in the conventional ICP spectrometry. By using this system, significantly lower detection limits were obtained related to the conventional ICP–AES method. The analysis results showed a close correlation between the daily mean volume of traffic and metals accumulated in the soil samples. On the other hand, using this method similar heavy metal profiles were gained, *i.e.* the total and plant-available heavy metal concentrations were the highest in the 0–1 m region from the road, and the further sampling points (0–90 m) showed significantly lower and lower heavy metal contents until at about 90 m distance from the road, where heavy metal contamination reached such a low value that can be considered to be the environmental background level. A similar trend was also recognised in plants; the plants very near to the road surface accumulated very high amounts of lead, cadmium and other heavy metals derived from vehicular traffic. The results revealed different behaviours of contaminants in different conditions inside the roadside areas (soil pH and heavy metal content, species and abundance of flora at the different sampling points, *etc.*). For example, total and available concentrations were apparently low only in one sampling site where each sampling point of the cross-section showed acid soil medium. Generally, acid pH means higher mobility of heavy metals, which was verified in our study too, as low concentrations in soil are associated with high accumulation factors in plants. Both single- and two-factor ANOVA results showed that total and available lead contents were not significantly different between sampling sites, therefore they were considered as traffic as the main contaminating source produced near the same lead content in different conditions. Single-factor ANOVAs showed significant differences, meanwhile two-factor ANOVAs did not show significant differences among mean lead contents detected in plants sampled in the five sites. These different results for lead showed that with the increase of variables (from one to two factors) the mathematical differences between lead contents became less and less significant. It calls for paying attention to the evaluation of bioindication and requires use of all the available variables in order to gain precise results from sampled areas. Some plants such as *Echinocloa crus-galli*, *Clematis recta*, *Artemisia vulgaris*, *Crepis biennis*, *Portulaca oleracea* and *Lathyrus pratensis* were found to be good indicators, as they accumulated three or four different heavy metals in high concentrations. In addition to their sensitive indicating ability, the majority of these plants are weeds of cereals in arable lands, and so are easily available in roadside areas.

Keywords: Contamination, roadside area, heavy metals, leaded fuel, soil-plant relationship, HHPN–ICP.

INTRODUCTION

Highway and motorway systems in western countries are more developed than in the eastern part of Europe, therefore studies of heavy metal – especially lead – contamination has started earlier, in the 1960s. Hamilton and Harrison (1991) dealt with problems of highway pollution in detail, and in their book they summarised the results published so far and cited many authors who dealt with this field of environmental science. They pointed out that traffic of over 25,000 vehicles/day led to detectable heavy metal contamination and increasing accumu-

lation. In Hungary researchers of the Hungarian Environmental Protection Bureau mapped lead contamination in roadside areas along the highways and motorways of Hungary during the period 1981–1985 (Árkosi and Buna, 1990). For sampling 50 m × 30 m rectangle-shaped areas were selected along local roadsides, and in these areas 36 sampling points were sampled. Figure 1 illustrates high contamination levels (1,000–2,000 mg kg⁻¹) in industrial regions, meanwhile contamination in the agricultural regions reached medium or low levels. This survey also verified a linear correlation between lead loading and traffic volume. Csathó (1994) and

Kádár (1995) compared Hungarian and foreign results, and concluded that lead contamination in Hungarian roadside areas increases continuously. The situation is likely to improve with introduction of unleaded fuel and the continuous reduction in the lead content of leaded fuel in Hungary in 1997. However, vehicles from Eastern Europe, which still operate on leaded fuel, will contaminate the Hungarian roadsides for some time to come. Since our sampling was carried out in 1994, so the results are outcome of higher lead content of fuel than that of today. Simon *et al.* (1996) reported that some plants (*e.g.* chicory, dandelion, *etc.*) as having a phytoindicator ability of cadmium.

Berndt (1988) introduced the use of hydraulic high pressure nebulization (HHPN) in ICP analysis. As aerosol yield is about 50 % (conventional pneumatic nebulization: 1–2 %, ultrasonic nebulization: 20–30%), the HHPN method became an outstanding choice for more effective sample introduction in the analytical chemistry of the 1990s. Introducing a high amount of sample into plasma the detection limit of contaminants became lower and the sensitivity of analysis higher. These advantageous changes led to more reliable detection in ppb level and reproducible detection of the concentrations which were near to detection limit of pneumatic nebulizer – ICP system.

MATERIAL AND METHODS

Sampling

The sampling sites are illustrated in Figure 1. The volumes of traffic in those sites significantly differ: Bánk – 3700; Józsa – 9500; Mályi – 25000; Gyöngyös – 17800 vehicles per day. Rudabánya is an unused open iron-mine, where industrial traffic (trailers, dumpers,

etc.) is usual. Gyöngyös and Mályi are located very near to the M3 motorway, Józsa to Highway No. 35, Bánk to the low-traffic Road 121, meanwhile there is industrial traffic only in Rudabánya.

Soil samples were taken from the 10 cm topsoil, the average depth of the root zone of the weed plants sampled. For plant sampling 1 m² area fields were designed, and the selected plants were cut at a height of 2 cm above the soil surface. With the use of this sampling we avoided sampling of the lowest part of plants, which is frequently contaminated with soil. Toxic metals can be translocated in the plant body, or can penetrate as organometallic compounds (*e.g.* tetraalkyl-lead) through leaves. Grazing animals (*e.g.* cattle), or the farmers collecting fodder for small domestic animals (rabbits, poultry, *etc.*) use the soil-free part for animal feeding. Animals feeding on contaminated fodder can that have direct and indirect effects on both animals and human beings. The plants sampled were washed with de-ionised water and separated on the basis of their species.

Sample preparation

Preparation of soil samples

28 soil samples were dried for 24 hours at 40°C. The plant remains and roots were removed; the soil was homogenised and passed through sieves with 2 mm apertures. The total element contents of the soils were measured after wet total digestion (Kovács *et al.*, 1996 a), using 65% HNO₃–30% H₂O₂ digestion. The plant-available amount of metals were measured in Lakanen-Erviö extracts (Lakanen and Erviö, 1980).

Preparation of plant samples

128 plant samples were dried at 105°C, and ground with a Retsch SK 1 hammer mill using a 1 mm sieve. 1 g

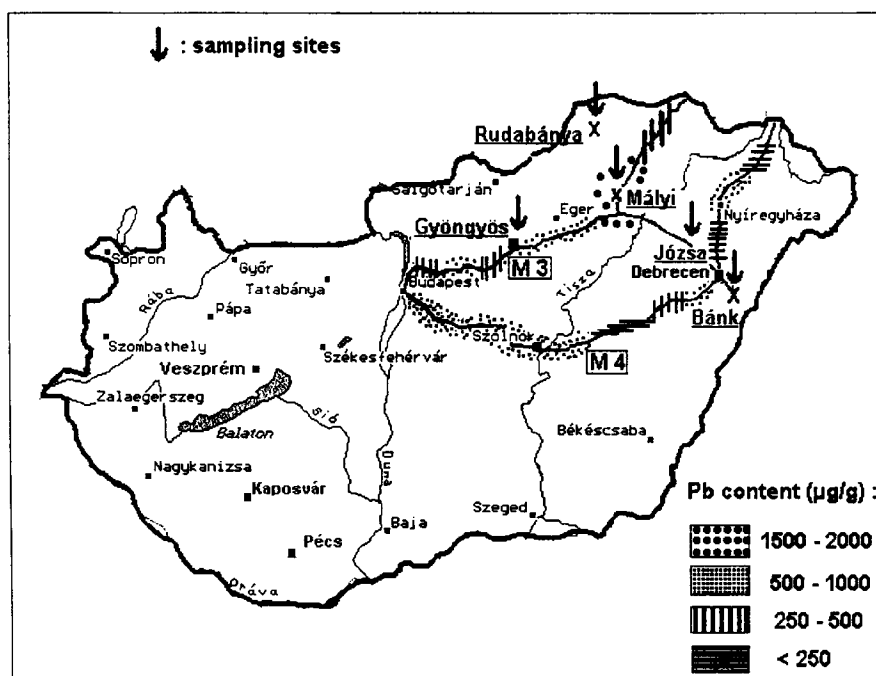


Figure 1 Location of the sampling sites in Hungary. The lead content in soils in roadside areas are illustrated based on national survey of Hungarian Environmental Protection Bureau (source: Árkosi and Buna, 1990.)

sample was digested with wet 65 % HNO₃–30% H₂O₂ digestion (Kovács *et al.*, 1996 b.) method.

Digestion

1 g homogenised soil samples were balanced for wet digestion. LABOR MIM OE-718/A electronic block digestion was used for this purpose. The digestion consisted of two stages: (1) Pre-digesting: 10 mL 65% HNO₃ was added to 1 g soil and was kept at 60°C for 30 minutes. (2) Digestion: 3 mL: 30 % H₂O₂ was added to the mixture at 120°C for 1.5 hours. The digested sample was filled up to 50 mL with de-ionised water and then filtered through MN (Macherey-Nagel) 640W filter paper. The filtered solution was analysed with HHPN-ICP system.

Soil extraction method

50 mL Lakanen-Erviö solution was added to 5 g soil placed into a 100 mL plastic vessel. The extraction was carried out in a soil extractor apparatus for 1 hour. The Lakanen-Erviö solution is a buffered EDTA solution (0.02 M Na₂EDTA, 0.5 M ammonium acetate, 0.5 M acetic acid; the pH was adjusted to 4.65 with either ammonia or acetic acid) and it is used to estimate the available element content for plants.

Detection

Significant improvements in sensitivity and detection limits were obtained for most of the elements related to conventional ICP–AES method (Table 1). Detection limits of ICP and HHPN–ICP were calculated based on counts of ICP detector in analysis of verified composition BCR soil standards (CRM 141R - calcereous loam soil, CRM 142R–trace elements in a light sandy soil, CRM 281–rye grass). The signals from the samples were detected with computer, then the detected (signal–baseline–signal)_n continuum were printed on paper, and were evaluated on the basis of peak height method. The peak heights of the samples were measured relating to the baseline. Calibration curves were plotted based on the measured peak heights of the standards, and concentration of the samples were calculated by their peak heights and the related concentration–height calibration curves.

pH measures

5 g air-dried soil samples were mixed with 12.5 mL de-ionised water (conductivity, 0.1 microSiemens). The pH of the soil – water system was measured with a pH-meter (Table 2.).

Table 1 Detection limits of ICP-AES and HHPN-ICP calculated by results of verified soil standards

Elements	ICP [µg/kg]	HHPN-ICP [µg/kg]
Cd, Zn	1	0.2
Ni	2	0.2
B, Co, Cr, Cu	3	0.1
Al, As, Hg, P, Pb, S	20	5.0
Se	25	4.0
Mo	50	12.0

Table 2 Mean pH values of soil samples from cross-section of roadside areas

Bánk	0 m	1 m	4 m	10 m	20 m		
	6.31	6.35	5.94	6.34	5.96		
Józsa	1 m	2 m	5 m	10 m	20 m	40 m	90 m
	8.1	7.65	7.57	7.68	6.91	7.72	7.46
	0 m	1 m	2 m	5 m	10 m	20 m	40 m
	7.86	7.10	6.80	6.43	6.52	6.34	5.73
Mályi	0 m	2 m	5 m	10 m	20 m	40 m	90 m
	7.86	7.10	6.80	6.43	6.52	6.34	5.73
Gyöngyös	0 m	2 m	5 m	10 m			
	9.24	8.31	7.94	8.20			
Rudabánya	0 m	5 m	20 m	40 m			
	7.74	8.00	8.02	8.35			

RESULTS

Concentrations measured in soil and plant samples

Total and available concentrations of the heavy metals in the soils and plant samples are presented in Table 3. It is remarkable that both the total and the available concentrations are apparently lower in Bánk, which is the only sampling site, where each soil sample is acid (Table 2). Bánk is located in a contaminated industrial region, where both acid sulfur and lead emissions are high (see the results of the national survey in Figure 1). Mean values of both the total and the plant-available concentrations of heavy metals in the soil decreased reasonably in roadside area moving away at right angles to the road. Similarly to the soil, the mean heavy metal concentration values in plants also decrease reasonably moving away the road (Figure 2 and 3). Thus the precipitation of heavy metal containing colloid and fuel droplet emission from vehicular traffic is highest very near to the road surface (0–1 m), and moving away the road impact of precipitation is less and less detectable, until (at ~90–100 m from the road) the detected concentration decreases to the level of background concentration. It is known that cars and other vehicles emit: (1) colloidal lead and uncombusted fuel droplets containing tetraalkyl-lead; (2) cadmium from filling material of tyres; and (3) zinc, copper, manganese, chromium and other heavy metals resulting from the abrasion of metal parts of the vehicles (Kádár, 1995). Thus, the concentrations of all heavy metals are expected to be highest at a distance of 0–1 m from roadsides. The statistical analysis showed a close correlation between the volume of daily traffic in the sampling sites and heavy metal concentration detected in 0–1 m zone from the road (Figures 4–6), particularly for the Bánk, Józsa and Gyöngyös. Exact traffic volume in Rudabánya is unknown and Mályi, the site with the highest traffic volume, did not fit in the pattern of the trend of Bánk–Józsa–Gyöngyös.

Correlation of different measured parameters

The following correlations were calculated: (1) between concentration of heavy metals in soil and plant and (2) between pH and concentration of heavy metals taken up by all the plants sampled.

Table 3 The mean heavy metal content in soil and plant samples derived from different points of roadsides

	Cd, total	Cd, avail	Cd, plant	Cr, total	Cr, avail	Cr, plant	Cu, total	Cu, avail	Cu, plant	Mn, total	Mn, avail	Mn, plant	Ni, total	Ni, avail	Ni, plant	Pb, total	Pb, avail	Pb, plant	Zn, total	Zn, avail	Zn, plant
Bank																					
0 m (3)	0.12	0.03	0.87	4.85	0.23	5.37	7.60	1.24	5.10	81.1	22.0	232.9	2.65	0.27	1.48	6.60	5.60	9.52	14.0	4.5	34.7
1 m (1)	0.10	0.01	0.99	4.30	0.08	7.00	3.92	0.53	5.94	96.1	26.5	97.0	3.75	0.24	1.53	4.92	4.41	8.68	12.2	2.4	23.4
4 m (2)	0.10	0.01	0.66	4.24	0.06	4.39	4.34	0.46	2.19	99.3	30.6	304.4	3.96	0.11	1.45	1.88	1.12	7.35	11.0	1.8	14.1
10 m (2)	0.06	0.02	0.57	3.81	0.03	5.35	3.63	0.45	3.48	96.0	38.3	80.1	3.21	0.16	1.01	1.82	0.81	7.04	9.4	1.0	15.8
Józsa																					
1 m (5)	1.32	0.60	0.18	44.6	1.04	0.78	24.4	8.9	2.98	439.0	231.2	30.2	18.94	2.87	0.36	202.11	190.05	5.24	117.2	46.6	20.0
2 m (7)	1.41	0.52	0.54	30.1	0.53	19.39	24.2	9.3	2.77	477.2	318.3	37.2	20.21	4.00	0.43	102.08	109.01	7.31	116.1	36.4	19.8
5 m (8)	1.35	0.37	0.17	26.1	0.18	0.57	27.0	9.6	2.84	890.1	650.3	50.3	25.38	6.00	0.51	48.92	31.80	2.92	106.1	30.9	10.6
10 m (16)	1.24	0.21	0.22	27.0	0.17	16.52	22.9	7.2	3.05	790.2	540.1	47.3	27.62	6.30	0.48	19.81	15.10	3.24	77.5	14.0	12.1
20 m (9)	1.24	0.16	0.29	28.3	0.07	13.80	23.6	4.3	2.41	890.3	445.2	45.6	27.44	4.29	0.50	19.80	10.50	2.75	79.0	8.1	12.3
40 m (11)	1.27	0.19	0.63	25.6	0.63	52.24	15.3	5.3	5.03	770.3	520.5	45.0	22.72	2.97	1.20	12.85	8.92	5.87	51.4	6.9	22.5
90 m (11)	2.17	0.35	0.70	35.2	0.03	24.89	29.6	11.9	4.85	3310.2	780.7	52.9	34.83	7.90	1.32	26.50	24.32	6.18	92.2	12.0	15.7
Gyöngyös																					
0 m (2)	2.82	0.55	1.54	43.8	1.57	5.12	56.0	10.8	6.38	920.1	242.4	54.7	22.41	2.49	2.18	670.22	520.05	40.93	186.0	66.0	50.8
2 m (7)	1.71	0.17	1.06	35.1	0.64	3.07	27.5	3.35	6.16	471.4	124.5	104.7	34.55	0.77	1.43	88.14	57.01	23.03	83.0	6.1	40.9
5 m (3)	1.96	0.14	1.17	33.5	0.72	7.31	31.8	4.76	7.75	610.3	93.0	167.5	36.24	0.65	2.08	45.63	20.72	20.31	72.0	3.5	29.0
10 m (3)	1.80	0.10	0.57	32.5	1.02	0.21	35.4	6.00	14.42	550.0	71.2	61.9	33.00	0.30	0.89	14.12	4.06	9.94	57.0	2.1	19.7
Mályi																					
0 m	1.45	0.46	2.11	56.0	1.55	20.28	23.3	6.91	4.75	610.2	155.1	64.3	15.60	1.58	2.61	200.01	130.12	26.48	102.0	37.5	46.6
1 m (4)	1.54	0.26	1.16	32.9	0.31	8.30	20.5	6.52	3.38	600.5	335.2	46.4	23.91	5.00	2.11	34.62	19.14	17.31	70.0	9.8	25.9
2 m (5)	1.44	0.22	0.96	30.1	0.38	4.39	23.8	5.70	2.54	670.1	277.0	46.1	24.24	4.96	1.37	31.07	14.81	7.95	70.0	6.7	21.9
5 m (5)	1.38	0.15	1.56	27.8	0.03	0.54	18.5	4.07	3.24	660.4	197.7	48.7	25.60	4.51	2.44	19.42	5.62	4.84	58.0	2.2	24.1
10 m (3)	1.40	0.19	1.36	26.0	0.10	1.18	18.1	4.56	2.20	680.1	195.5	87.7	25.02	4.23	2.98	20.73	5.12	4.64	57.0	2.2	23.1
20 m (3)	1.23	0.21	0.74	25.1	0.12	0.0	17.7	4.24	3.35	700.7	205.2	76.4	24.41	4.63	3.14	21.04	5.81	5.17	58.0	2.5	17.9
40 m (2)	1.15	0.15	0.62	25.2	0.15	0.0	16.1	4.74	2.37	680.2	203.2	132.6	23.32	4.67	3.18	17.81	5.15	4.34	55.0	3.4	22.2
90 m (2)	1.61	0.22	0.80	28.7	0.25	0.54	18.1	5.00	1.99	580.5	237.1	92.1	27.35	5.23	2.42	17.33	5.92	5.33	69.0	4.2	25.6
Rudabány																					
0 m (1)	2.40	0.52	0.75	8.5	0.49	5.67	113.1	38.8	5.58	2550.4	620.4	50.0	8.80	3.53	1.26	65.02	59.07	9.21	262.0	92.2	25.6
5 m (1)	1.95	0.50	1.18	12.4	0.24	8.92	77.2	16.4	2.49	2040.1	670.2	60.5	13.21	2.45	0.91	120.10	65.01	5.52	397.0	93.1	20.2
10 m (5)	3.95	0.71	0.48	10.3	0.45	5.74	227.0	23.0	3.44	3470.5	900.5	52.1	11.65	3.82	0.93	177.01	83.06	6.16	570.0	113.0	26.8
40 m (3)	3.79	0.53	0.64	9.1	0.83	5.68	305.0	63.0	3.01	3240.8	600.3	29.8	9.70	3.04	1.03	127.04	89.11	7.89	354.0	105.1	20.9

The number of soil samples is constant: 1 sample/sampling point. The number of plant samples are presented in parentheses.

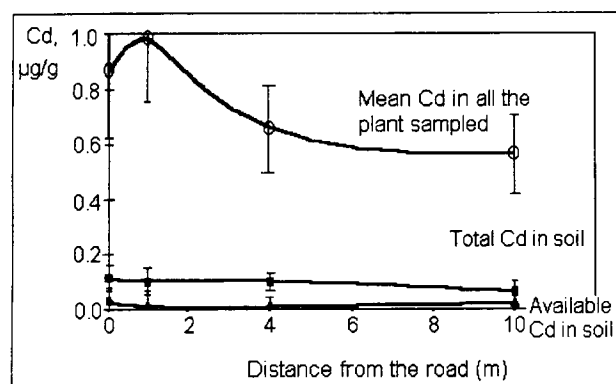


Figure 2 Cd accumulation in plants grown in roadside area in Bánk.

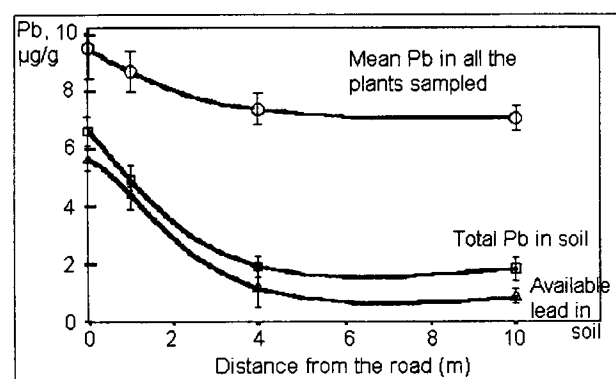


Figure 3 Pb accumulation in roadside area in Bánk.

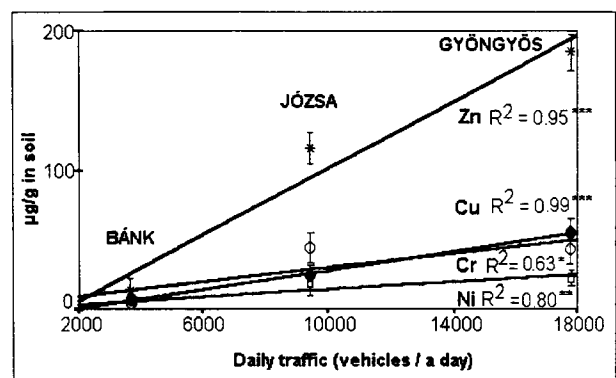


Figure 4 Correlation of volume of daily traffic and total Zn, Cu, Cr and Ni concentration in 0-1 m soil samples.

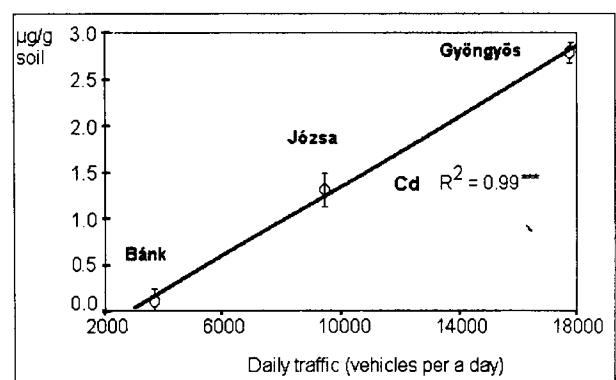


Figure 5 Correlation of volume of daily traffic and total Cd concentration detected in 0-1 m soil samples.

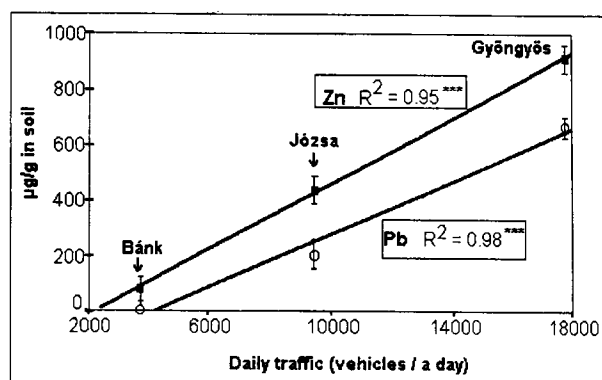


Figure 6 Correlation between volume of daily traffic and total Pb and Zn content in 0-1 m from the road.

Close correlations were found between total concentrations of heavy metals in soil and plants sampled in Mályi (Table 4) for which the sample number was the highest. The results reflected a close correlation between each combination of Pb and Zn content in soil and plant. Cross-effects were found between Pb, Cd and Zn concentrations in the plant – soil system. Although these results are expressive parameters of the description of soil – plant relation, the calculation of the correlation depends remarkably on the sample number. Therefore calculation of accumulation factors, which does not depend on sample number (see Table 5), seem to be a more characteristic, descriptive and reliable method than that of the correlation coefficients.

Close correlation was found between pH and total Pb in the sampling points at Mályi, Józsa and Gyöngyös (Figures 7 and 8). It verified the direct relation between pH and accumulation of heavy metals.

Close correlation between the pH and element content (Al, Cd, Cu, Cr, Fe, Pb and Zn) was found between plant samples derived from different points in roadsides of Mályi, Józsa and Gyöngyös. Although aluminium is not heavy metal it is represented in Figure 9-11, because aluminium is also a phytotoxic element and may increase the adverse effects of heavy metals on plants.

Soil-plant accumulation factors

Acid pH of soils means a higher availability of the vast majority of heavy metals for plants. It was verified by the calculation of soil / plant accumulation factors in our study too (Table 5), because low concentrations in soil and high accumulation factors of all the metals in plants were recorded only in the case of acid soils. Therefore, high concentrations of heavy metals in soils and low accumulation factors in plants in the case of alkali soil samples is also understandable, *i.e.* in this medium heavy metals do not enter the soil solution and the originally dissolved forms will precipitate. Accumulation factors of a sampling point in the cross-section of the roadside were calculated as the concentration of a selected heavy metal in all the plants sampled at that point (and digested together as one sample), divided by concentration of the (1) total or (2) plant-available form of heavy metal in the soil sample. Both the calculated factors (accumulation factors of total and available concentrations) are illustrated

Table 4 Correlation of Pb, Cd and Zn content in soil and plant samples in Mályi

	Soil Pb	Plant Pb	Soil Cd	Plant Cd	Soil Zn	Plant Zn
Soil Pb	1					
Plant Pb	0.98***	1				
Soil Cd	0.21	0.18	1			
Plant Cd	0.78**	0.73*	0.08	1		
Soil Zn	0.96***	0.96***	0.39	0.69	1	
Plant Zn	0.97***	0.93***	0.40	0.81**	0.95***	1

(* P < 5 %, ** P < 1 %, *** : P < 0.1)

in Table 5. The factors which are higher than 1, showed higher absolute values than the mean plant/soil factors published so far (e.g. Lisk, 1972 : plant/soil for Cd 5.3 and for Pb 2.3). High accumulation factors were found in acid Bánk soil samples (Cd: 22.8–99, Cr: 68–178.3, Cu: 2.9–12.7, Mn: 3.2–8.8, Ni: 6.2–14.2, Pb: 2.2–17.9, Zn: 9.3–14.5) in the case of both accumulation factors, however the factor calculated with plant-available concentration is significantly higher than that calculated with the total concentration. Concerning behaviour of the elements, the ranges of accumulation factors reflected outstanding accumulation ability of cadmium and chromium. Chromium accumulated even

in those plants which were grown in alkali soils, so chromium seems to be the most easily available heavy metal. Probably the reason of this result is the behaviour of chromium: one of its chemical form is essential cationic Cr (III), the other one is toxic anionic Cr (VI) and both of them are plant-available. The others are bivalent cations, thus they are available as the cations. Accumulation of heavy metals in plants is detected remarkably less frequently in the case of alkali soil samples than in acid soil samples. Although Cd and Cr also seemed to be accumulated in plants grown in alkali soils, their accumulation factors are apparently lower than in acid soils.

Table 5 Accumulation factors of the studied heavy metals

	Cd,t.	Cd,a.	Cr,t.	Cr,a.	Cu,t.	Cu,a.	Mn,t.	Mn,a.	Ni,t.	Ni,a.	Pb,t.	Pb,a.	Zn,t.	Zn,a.
Bánk														
0 m	8.8	72.5	1.2	68.0	1.3	9.6	2.4	8.8	0.4	6.2	1.9	2.2	2.8	14.5
1 m	9.8	99.0	1.7	120.7	1.4	12.7	1.0	3.2	0.4	14.2	4.6	7.8	2.1	13.0
4 m	10.8	28.7	1.2	146.3	0.6	4.8	3.2	7.9	0.5	8.8	4.0	9.1	1.5	13.3
10 m	11.4	22.8	1.3	178.3	1.1	2.9	1.2	4.2	0.3	10.7	7.0	17.9	1.7	9.3
Józsa														
1 m	0.1	0.3	0.0	0.8	0.1	0.3	0.1	0.1	0.0	0.1	0.0	0.0	0.2	0.4
2 m	0.4	1.0	0.6	36.6	0.1	0.3	0.1	0.1	0.0	0.1	0.1	0.1	0.2	0.5
5 m	0.1	0.5	0.0	3.1	0.1	0.3	0.1	0.1	0.0	0.1	0.1	0.1	0.1	0.3
10 m	0.2	1.0	0.6	96.0	0.1	0.4	0.1	0.1	0.0	0.1	0.2	0.2	0.2	0.9
20 m	0.2	1.8	0.5	191.7	0.1	0.6	0.1	0.1	0.0	0.1	0.1	0.3	0.2	1.5
40 m	0.5	3.3	2.0	82.9	0.3	0.9	0.1	0.1	0.1	0.4	0.5	0.7	0.4	3.3
80 m	0.3	2.0	0.7	829.7	0.2	0.4	0.0	0.1	0.0	0.2	0.2	0.3	0.2	1.3
Gyöngyös														
0 m	0.6	2.8	0.1	3.3	0.1	0.6	0.1	0.2	0.1	0.9	0.1	0.1	0.3	0.8
2 m	0.6	6.1	0.1	4.8	0.2	1.8	0.2	0.8	0.0	1.9	0.3	0.4	0.5	6.7
5 m	0.6	8.0	0.2	10.2	0.2	1.6	0.3	1.8	0.1	3.2	0.4	1.0	0.4	8.1
8 m	0.3	5.8	0.0	0.2	0.4	2.4	0.1	0.9	0.0	3.0	0.7	2.4	0.3	9.2
Mályi														
0 m	1.5	4.6	0.4	13.1	0.2	0.7	0.1	0.4	0.2	1.7	0.1	0.2	0.5	1.2
1 m	0.8	4.4	0.3	26.7	0.2	0.5	0.1	0.1	0.1	0.4	0.5	0.9	0.4	2.6
2 m	0.7	4.3	0.1	11.4	0.1	0.4	0.1	0.2	0.1	0.3	0.3	0.5	0.3	3.3
5 m	1.1	10.3	0.0	18.0	0.2	0.8	0.1	0.2	0.1	0.5	0.2	0.9	0.4	10.8
10 m	1.0	7.3	0.0	11.8	0.1	0.5	0.1	0.4	0.1	0.7	0.2	0.9	0.4	10.4
20 m	0.6	3.5	0.0	0.0	0.2	0.8	0.1	0.4	0.1	0.7	0.2	0.9	0.3	7.2
40 m	0.5	4.0	0.0	0.0	0.1	0.5	0.2	0.7	0.1	0.7	0.2	0.9	0.4	6.5
90 m	0.5	3.7	0.0	2.1	0.1	0.4	0.2	0.4	0.1	0.5	0.3	0.9	0.4	6.1
Rudabánya														
0 m	0.3	1.4	0.7	11.7	0.0	0.1	0.0	0.1	0.1	0.4	0.1	0.2	0.1	0.3
4 m	0.6	2.4	0.7	37.5	0.0	0.2	0.0	0.1	0.1	0.4	0.0	0.1	0.1	0.2
30 m	0.1	0.7	0.6	12.8	0.0	0.1	0.0	0.1	0.1	0.2	0.0	0.1	0.0	0.2
50 m	0.2	1.2	0.6	6.8	0.0	0.0	0.0	0.0	0.1	0.3	0.1	0.1	0.1	0.2

> 1 are in bold, t. = total conc., a. = plant-available conc.

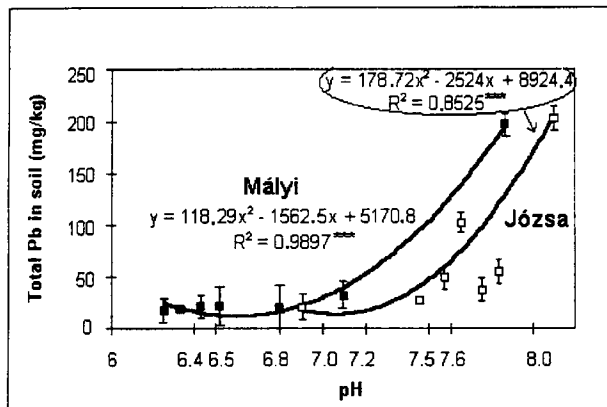


Figure 7 Correlation of total Pb and pH in samples from Mályi and Józsa (the points of curves represent related data, but do not represent cross-section of roadside areas).

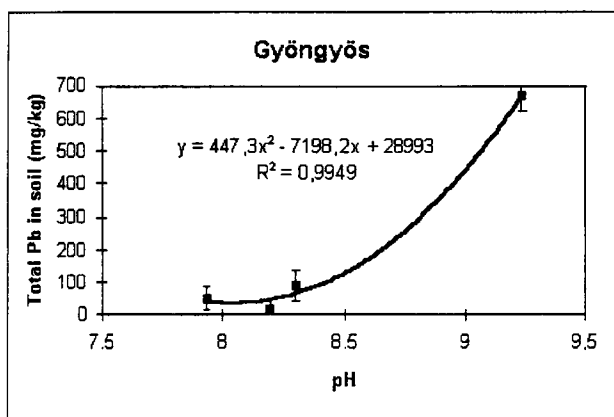


Figure 8 Correlation of total Pb and pH in samples from Gyöngyös (the points of curves represent related data, but do not represent cross-section of roadside areas).

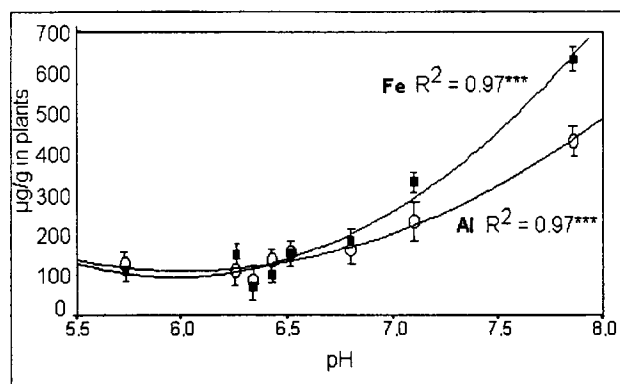


Figure 9 Correlation between pH and elements (Al, Fe) in all the plants sampled in different sampling points Mályi (the points of curves represent related data, but do not represent cross-section of roadside areas).

Statistical analysis

The element contents measured in plant and soil samples were analysed with one-factor and two factor ANOVA methods in order to evaluate whether the location of the samples sites and the distance from the road have got effects on the detected heavy metal concentrations in soil and plant samples.

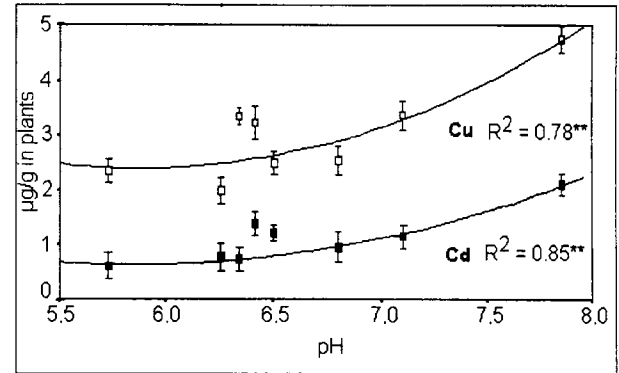


Figure 10 Correlation between pH and heavy metals (Cd, Cu) in all the plants sampled in different sampling points of Mályi (the points of curves represent related data, but do not represent cross-section of roadside areas).

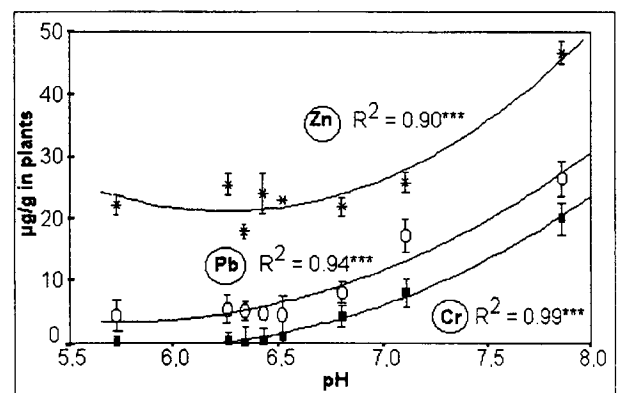


Figure 11 Correlation between pH and heavy metals (Zn, Pb, Cr) in all the plants sampled in different sampling points of Mályi (the points of curves represent related data, but do not represent cross-section of roadside areas).

ANOVA results of soil and plant samples from different sampling sites

pH values were significantly different between sampling sites ($F = 20.36$, $p < 0.1\%$, $F_{crit} : 2.79$), so it verified the original supposition, i.e. comparison of different sampling sites based on really different sampling conditions. It also verified differences between conditions affecting heavy metal mobility and uptake.

Single-factor ANOVA

The differences were not significant neither between total Pb contents, nor between available Pb contents measured in the different sampling sites (Table 6). Probably the reason why lead contents did not differ significantly is that high values were detected in each sampling site. However, lead contents detected in all the plants grown showed significant differences in the results of the different sampling points. It is also understandable, because it is predictable that heterogeneous plant populations in different conditions cannot accumulate equal amounts of lead. On the other hand, the results of Cr and Zn showed a reversed tendency to that of Pb, because in the cases when Cr and Zn showed significant differences, Pb was not significant, and vice versa. The reason for this can be continuous lead emission in

Table 6 F-values of single-factor and two-factor ANOVA different soil and plant samples from the five sampling sites

F-values of ANOVAs	F-crit.	Mean values of heavy metals in the sampling sites						
		Cd	Cr	Cu	Mn	Ni	Pb	Zn
Total concentration								
Single-factor, factor: sampling sites	2.79	25.92***	21.37***	15.46***	14.75***	37.61***	1.78	34.47***
Two-factor, 1st factor: distance from the road (0–90 m)	2.36	0.95	0.67	1.06	0.79	1.25	1.29	1.09
Two-factor, 2nd factor: sampling sites	2.71	2.85*	8.99**	3.25*	3.00*	8.67***	0.81	3.56*
Plant-available concentration								
Single-factor, factor: sampling sites	2.79	10.29***	3.36*	13.63***	26.63***	15.76***	1.51	28.22***
Two-factor, 1st factor: distance from the road (0–90 m)	2.36	0.77	1.46	0.86	0.81	0.70	1.31	1.17
Two-factor, 2nd factor: sampling sites	2.71	3.15*	1.37	2.96*	5.60**	13.84***	0.79	3.95*
Mean concentration in plants								
Single-factor, factor: sampling sites	2.81	4.80**	2.33	7.49***	5.84**	16.11***	5.39**	3.52*
Two-factor, 1st factor: distance from the road (0–90 m)	2.36	1.96	0.25	1.08	1.08	1.01	1.43	0.90
Two-factor, 2nd factor: sampling sites	2.71	5.11**	3.02*	0.94	1.44	12.16***	1.82	1.62

* $p = 5\%$; ** $p = 1\%$; *** $p = 0.1\%$.

contrast to random chromium and zinc contamination, which leads to deviations. Various forms of an element can show all the possible variations in this ANOVA analysis, as it was in the first case. The difference (1) between total chromium contents was significant at $p < 0.1$ level, meanwhile (2) between available Cr contents was significant at $p < 5\%$ level only; and (3) between plant concentrations was not significant. Thus the difference becomes smaller and smaller as we get closer and closer to the final stage in soil–plant system, *i.e.* to the concentration data of metals taken up by plants.

The second remarkable case is zinc. Total and available zinc was significant at $p < 0.1$ level, but plant was significant at $p < 5\%$ level only. It is similar to the trend seen in the case of chromium: uptake by plant decreases the differences between the related samples.

Two-factor ANOVA

Two-factor ANOVA was needed to get more information as to which factor is really important and which is not. The factors were, distance from the road and location of sampling sites (Table 6). The first factor, distance from the road produced only not significantly differing data. The second factor, difference between mean concentrations in the sampling sites were significant in the majority of the samples although some cases did not show significant differences. Such cases are: (1) total lead content in soil; (2) plant-available lead and chromium content in soil; and (3) copper, manganese, lead, zinc content in plants. These cases were not significantly different with any of the factors. Lead is the most remarkable element among heavy metals, as lead was found in the soil and plant samples in a well-determined concentration range.

ANOVA results of plant samples from roadside cross-section

Single-factor ANOVA

In the former paragraph ANOVA of soil and plant samples from the sampling sites were calculated and

here the ANOVA results of all the plant samples from roadside cross-section are presented in Table 7. Pb and Zn contents differed significantly in the high traffic sites only, meanwhile low traffic sites did not show significant differences. It means that the response of the flora is detectable in highly contaminated sites only. On the other hand, the higher the sample number, the higher the significant F-values. In sampling points at Rudabánya there were no elements revealing significant differences.

DISCUSSION

Relationship between botanical and chemical results

The soils near the road surface are mainly alkali as a result of alkali by-products of road construction, and this condition leads to long-time precipitation and accumulation of heavy metals in the vicinity of the roads. It resulted high lead and heavy metal concentration in both soils and plants. Short- or long-term acidification of contaminated soil makes heavy metals more and more easily available for plants. In addition, acidification is a necessary consequence of natural processes such as (1) root acid emission of plants, by which plants prepare elements for uptake, and (2) decomposition of plant residues, as well as the human effects (*e.g.* agricultural activities such as fertilisation near roads, or pollution with acid fuel fragments, acid rains of industrial origin). On the other hand, this is one of the reasons why we used acid EDTA solution to evaluate the plant-available amount of heavy metals. If the contaminated soils are used to produce plants – most frequently for grass and weed fodder production—adverse effects of Pb, Cd, *etc.* on plant growth, high lead accumulation in plants and other harmful effects will be observable and these effects will cause other problems in the higher members of food chain (domestic animals and human beings). In order to support this theory, we can only mention the case of the one acid soil sampling site (Bánk), which contained

Table 7 *F-values of single-factor ANOVA of plants sampled in 0–90 m roadside cross-sections (the single factor is distance from the road, the position between 0 and 90 m)*

Sampling sites	Mean values of heavy metals in the sampling sites							
	F crit.	Cd	Cr	Cu	Mn	Ni	Pb	Zn
Bánk F	4.07	1.10	1.97	0.95	5.75*	0.86	1.88	1.19
Józsa F	2.26	2.88*	1.29	10.11***	1.12	27.03***	13.69***	9.61***
Mályi F	2.66	1.04	2.78*	1.06	3.33*	1.72	6.13***	3.05*
Gyöngyös F	3.59	1.54	2.35	1.58	1.37	1.57	7.04**	3.81*
Rudabánya F	4.74	1.99	1.62	0.21	1.08	0.25	0.67	0.11

* $p = 5\%$, ** $p = 1\%$, *** $p = 0.1\%$.

apparently less lead than the alkali sites did. It is highly probable that the plant population took up the missing lead amount.

Concerning the results of single-factor ANOVA of plant samples, one reason for the behaviour of heavy metals detailed above is the existence of a biological threshold in plants which limits the amount of Cr and Zn entering into the plant's body, as they have smaller ionic radii than lead. Lead in animal and human toxicology (Butler, 1957, Pounds, 1991) is verified as an ion entering in the pathway of calcium on the basis of similar ionic radius and electric charge, therefore uptake of the calcium-imitating lead is possible in plant life processes too. It is understandable if we consider that each plant needs much calcium but a low amount of heavy metals for its growth and development. Another reason for the behaviour of heavy metals can be aerosol contamination of the immediate environment of roads. Aerosol droplets contain unreacted organic lead compounds such as tetraethyl - lead, which penetrates through plant cells or human skin very easily. As fine aerosol it sprays the roadside continuously, and can be available through leaves continuously.

In this study we considered as 'indicators' those plants which contained an element over its (mean + SD) value calculated for all the plant samples independently from sites. The plants that contained much higher concentration than that of the mean calculated according to the above mentioned, were considered 'accumulator plants'.

The greatest problem of plant sampling was availability of the good indicator plants in a selected roadside area. In our study *Echinochloa crus-galli*, *Cichorium inthybus*, *Sonchus arvensis* and *Artemisia vulgaris* were the element-rich plants that could be found in more than one site. Abundance of a plant strongly depends on actual conditions (e.g. soil, pH, nutrients, moisture, the actual dominant plant species in the flora), therefore existence of a required plant in the selected plant population is not guaranteed.

The results of plant – heavy metal concentrations are listed element by element in Table 8.

Echinochloa crus-galli (Cd, Ni, Pb, Zn), *Clematis recta* (Cu, Mn, Ni, Pb), *Artemisia vulgaris* (Cu, Mn, Zn), *Crepis biennis* (Cu, Pb, Zn), *Portulaca oleracea* (Cd, Pb, Zn) and *Lathyrus pratensis* (Cd, Cr, Cu) were considered as the most sensitive indicator plants, because they accumulated high concentrations of (at least) three heavy metals. There were other sensitive indicator plants which accumulated two heavy metals – *Artemisia absinthium* (Cd, Zn), *Hieracium caespitosum* (Cd, Cr), *Matricharia inodora* (Cd, Ni), *Cirsium arvense* (Cu, Zn). On the above basis, in chemical aspect, Cd seems to be the most easily monitorable contaminant.

Evaluating the results we tried to predict accumulation ability of plants on the basis of botanical (macroscopic) characteristics and found similarities between some plants. For example, similar trends were found between botanical and accumulative characteristics of *Lathyrus pratensis* and *Vicia tetrasperma*, because both of them (1) are leguminous weeds and (2) accumulate high amounts of chromium and copper. Concerning high accumulation detected in leguminous weeds, this good indicator characteristic could be considered general for leguminous plants. On the other hand, samples of two *Artemisia* (*Artemisia vulgaris* and *Artemisia absinthium*) species were analysed and the results showed that both of them accumulated high amounts of elements, especially heavy metals (Cd, Zn).

It is remarkable as regards biomonitoring that the samples of *Chenopodium album*, which is one of the most frequently found weeds in maize fields, did not accumulate heavy metals in high concentrations. Maybe the sample plants were offspring of such parental plants, that lived in well-fertilised cultivated areas, and as a consequence, the vitality of offspring – and consequently element uptake – is reduced ('assimilation effect'). This theory was based on the fact that other plants in the vicinity of *Chenopodium album* samples accumulated high concentrations of heavy metals.

Table 8 The highest detected concentration in the plant samples and the mean \pm SD value of the related species

Latin name	Cd	Cr	Cu	Mn	Ni	Pb	Zn
<i>Achillea millefolium</i>	–	–	–	–	–	20.5	–
<i>Agropyron repens*</i>	–	–	8.7	–	–	–	–
<i>Agrostys tenuis</i>	–	–	–	226	–	–	–
<i>Artemisia absinthium</i>	1.64	–	–	–	–	–	51
<i>Artemisia vulgaris</i>	1.41	–	9.5	335	–	–	67
<i>Carduus acanthoides</i>	–	–	–	–	–	33.1	–
<i>Chenopodium album*</i>	–	–	–	–	3.7	–	–
<i>Cichorium inthybus</i>	–	–	27	–	–	–	–
<i>Cirsium arvense*</i>	–	–	6.6	–	–	–	67.4
<i>Crepis biennis</i>	–	–	12	–	–	25.4	43
<i>Clematis recta</i>	–	–	7	322	3.5	36.8	–
<i>Dactylis glomerata</i>	–	53	–	–	–	–	–
<i>Echinochloa crus-galli*</i>	2.11	–	–	–	3.7	42.4	54
<i>Galium aparine*</i>	–	276	–	–	–	–	–
<i>Hieracium caespitosium</i>	1.55	100	–	–	–	–	–
<i>Lathyrus pratensis</i>	2.36	194	7.5	–	–	–	–
<i>Matricaria inodora*</i>	3.42	–	–	–	4.6	–	–
<i>Melilotus officinalis</i>	–	–	–	276	–	–	–
<i>Portulaca oleracea</i>	1.84	–	–	–	–	39.4	47
<i>Polygonum vonvulvulus</i>	–	–	–	–	–	17.9	–
<i>Populus alba</i>	–	–	–	–	–	–	65
<i>Salvia nemorosa</i>	–	–	6.6	–	–	–	–
<i>Sonchus arvensis</i>	2.6	69	–	–	–	–	–
<i>Taraxacum officinale</i>	–	81	–	–	–	–	–
<i>Urtica urens</i>	–	–	–	–	–	20.5	–
<i>Valerianella locusta</i>	–	77	–	–	–	–	–
<i>Vicia tetrasperma</i>	–	54	6.2	–	–	–	–
Mean of the species	0.7	12.7	4.0	66.4	1.2	7.9	21.2
Standard deviation	0.6	35.0	2.9	58.6	0.9	7.4	12.2

(– not significantly higher than mean; * the most frequently found weeds in wheat and maize fields.

Another interesting, although already mentioned result is the detection of high Cr and Cu concentrations in *Vicia tetrasperma*, which is a member of the *Vicia* family. Another member of this family – *Vicia faba* – is a cultivated fodder plant used in animal feeding. Although cultivation of *Vicia faba* decreases nowadays, because of its bitter alkaloid contents and low yields, it can be efficiently produced in the feed industry. If *Vicia faba* is also be verified as natural Cr and Cu source, it can be very useful in healing anaemia, which frequently appears in new-born piglets, and which is derived from disadvantageous iron-copper ratio in fodder.

CONCLUSIONS

As a result of this study a uniform trend was mapped in the field of roadside protection research. It reflected that mean concentrations of heavy metals decreased significantly in relation to moving away from the road shoulder. Soil contamination very near the hard shoulder was found to be a general trend for each sampling site. In the case of related plant samples similar decreasing concentration profiles were recorded. It was considered that these results were the consequences of the different behaviours of the contaminants in the different soil-plant system found in the roadside areas. For example, total and available concentrations were apparently low in the only one sampling site where each sampling point of the cross-section was acidic. Acidic pH means higher mobility

of heavy metals, which was verified by low concentrations in the soil and also by the calculated high accumulation factors. Both single- and two-factor ANOVA showed that total and available lead contents were not significantly different between sampling sites. Based on this result, lead was considered as the most dangerous contaminant that accumulated very effectively in the roadside areas. Both significant and not significant differences between mean lead contents in plants were given, depending on whether single – or two-factor ANOVA method has been applied. This uncertainty concerning impact of lead on plant population pointed out the heterogeneity of plant population, as two-factor ANOVA, which indicated significant difference, can more exactly show the combination of actual conditions in sampling points. Cadmium and chromium were also accumulated effectively in plants. Considering botanical aspects, some plants such as *Echinochloa crus-galli*, *Clematis recta*, *Artemisia vulgaris*, *Crepis biennis*, *Portulaca oleracea* and *Lathyrus pratensis* were found to be good indicators, as they accumulated three or four different heavy metals in high concentrations.

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