Heavy metal contamination of roadside soil along Ljubljana – Obrežje highway

Onesnaženje tal s težkimi kovinami vzdolž avtoceste Ljubljana – Obrežje

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Abstract: In July 2004 28 soil samples were collected at 5 localities along Ljubljana – Obrežje highway with the intention to investigate heavy metal contamination in roadside soils. Samples were taken from the upper 5 cm of soil at different distances from the highway. Mineralogical and chemical compositions were determined. Heavy metals could be adsorbed by clay minerals, Fe-oxides and/or organic matter. They could be also incorporated in the structures of carbonates. Lead (values up to 516 ppm) can be confirmed as the most distinctive heavy metal from road traffic pollution, though levels of zinc (up to 873 ppm), copper (up to 74 ppm) and cadmium (up to 1.9 ppm) in roadside soil can also be connected with road traffic. The levels of these metals decreased with distance from the road at road sections, which were not included in highway reconstruction works in years after the first sampling. The reconstruction works seemingly changed this trend of heavy metal distribution. Direct comparison between profiles from the years 1993 (first sampling) and 2004 (second sampling) was not possible.

Izvleček: V juliju 2004 sem odvzel 28 vzorcev, vzdolž avtoceste Ljubljana – Obrežje, z namenom raziskati onesnaženje tal s težkimi kovinami. Tla sem vzorčil na petih lokacijah, na različnih oddaljenostih od ceste, v globini 0 – 5 cm. Vzorcem smo določili mineralno in kemijsko sestavo. Težke kovine so se v preiskanih vzorcih lahko absorbirale na glinene minerale, Fe-okside in /ali organsko snov. Lahko so se vključile v kristalne rešetke karbonatov. Lahko potrdimo, da je cestni promet prispeval k onesnaženju tal predvsem s svincem (vsebnosti do 516 ppm), vendar tudi vsebnosti cinka (do 873 ppm), bakra (do 74 ppm) in kadmija (do 1.9 ppm) lahko povežemo s cestnim prometom. Vsebnosti teh kovin upadajo z oddaljenostjo od ceste, vendar le ob cestnih odsekih, ki jih niso zajela avtocestna razširitvena dela v letih po prvem vzorčenju tal. Razširitvena dela so navidezno spremenila trend upadanja vsebnosti z oddaljenostjo od ceste. Neposredna primerjava med profili iz leta 1993 (prvo vzorčenje) in 2004 (drugo vzorčenje) ni mogoča.

Keywords: heavy metals, traffic, soil, contamination.
Ključne besede: težke kovine, promet, tla, onesnaženje.

Short scientific paper

INTRODUCTION

The first systematic research of heavy metal roadside contamination in Slovenia was carried out in the year 1993 by ZUPANČIČ (1994, 1997). The samples were taken along former two–laned Ljubljana – Obrežje high-speed road (now mostly reconstructed into four–laned highway), which connects Ljubljana with Dolenjsko and the Obrežje border crossing (Croatia). The reasons for new sampling along this highway in the year 2004 were: increasing traffic density along this highway (from the year 1993 till 2004 it increased for approximately 80 %), progressive introduction of unleaded petrol in Slovenia since 1988, prohibition of selling leaded petrol in Slovenia from the year 2001 and effects of highway reconstruction works to the level of contamination. The position of profiles was as near as possible to those ones from the year 1993. The objective of the present investigation was to determine the changes in heavy metal contamination (with Pb, Zn, Cu, Cd, Ni), which occured in the last 10 years.

Contamination of roadside soil due to road traffic

The main processes by which vehicles spread heavy metals (Pb, Zn, Cu, Cd, Ni) into the environment are combustion processes, the wear of cars (tires, brakes, engine), leaking of oil and corrosion. Lead is released in combustion of leaded petrol, zinc is derived from tire dust, copper is derived from brake abrasion and corrosion of radiators, the other heavy metals have mixed origins (DAVIS ET AL., 2001; VAN BOHEMEN AND VAN DE LAAK, 2003). Heavy metals are also released due to weathering of road surface asphalt and corrosion of crash barriers and road signs (VAN BOHEMEN AND VAN DE LAAK, 2003).

The general decrease in concentrations of heavy metals with distance from the road, with depth in the soil profile, and the general increase in concentrations with traffic density indicates their relation to traffic (MARKUS AND MCBRATNEY, 2001; OLAJIRE IN AYODELE, 1997; VIARD ET AL., 2004; ZUPANČIČ, 1997).

Background concentrations of heavy metals in soils

The soil contamination with an element can be estimated only if its background concentration is known. The natural heavy metal content of soil is strongly related to the composition of the parent rock. The mean lead (Pb) concentration for surface soil on the world scale could be estimated as 25 ppm. The upper limit for the Pb content of a normal soil could be established as 70 ppm. Mean total zinc (Zn) contents in surface soils of different countries range from 17 to 125 ppm. The mean levels for copper (Cu) in surface soils of different countries vary from 6 to 60 ppm. Soils throughout the world contain nickel (Ni) within the broad range of from 1 to around 200 ppm. The calculated mean of nickel for world soils is 20 ppm. The background cadmium (Cd) levels in soils do not exceed 0.5 ppm (KABATA-PENDIAS AND PENDIAS, 1986).

The boundary, the alerting and the chritical values of heavy metals in soils

The boundary, the alerting and the chritical values of heavy metals in soil, according to Slovenian legislation (Official Gazette, 68/96), can be found in Table 1.

Table 1. The boundary, the alerting and the chritical values of heavy metals in soil (Official Gazette, 68/96). Values are in mg/ kg of dry soil or in ppm.
Tabela 1. Mejne, opozorilne in kritične vrednosti težkih kovin v tleh (Uradni list RS, 68/96). Vrednosti so podane v mg/ kg suhih tal oziroma v ppm.

	The boundary value Mejna vrednost	The alerting value Opozorilna vrednost	The chritical value Kritična vrednost
Pb	85	100	530
Zn	200	300	720
Cu	60	100	300
Ni	50	70	210
Cd	1	2	12

Mobility of heavy metals in soils

Mobilized heavy metals become readily available to plants and with this they enter into the food chain. They can also migrate to groundwater. Specific soil properties, mainly its pH, Eh, cation exchange capacity, amount of organic matter in soil, amount of clay minerals and amount of iron, manganese and aluminum oxides, control the rates of heavy metal migration in the profiles. Clay minerals, Fe-, Mn-, Al- oxides and organic matter are the most important groups for the sorption of heavy metals. Heavy metals can be also incorporated in the structures of carbonates, phosphates and sulfides (Kabata-Pendias and Pendias, 1986).

Materials and Methods

Sampling and preparation for analysis

In July 2004, soil contamination along Ljubljana – Obrežje highway was monitored at 5 localities – Krakovski gozd, Družinska vas, Dob, Ivančna Gorica and Dedni dol. The shortest distance between two profiles was approximately 4 km and the longest approximately 43 km. Selected profiles lied along road sections, which have been opened to traffic for different periods of time. The position of profiles was as near as possible to those ones from the year 1993. In 1993 the profiles lied mostly along two-laned high-speed road (except profile Dedni dol, which lied at highway section).

Twenty-seven soil samples (including four sampling replicants) were taken from the upper 5 cm of soil, at 1 m, 5 m and 10 m distances from the road. The research from the year 1993 (Zupančič 1994, 1997) showed that the highest values are limited to topsoil (0 - 5 cm) and distances less than 10 m from the road. Samples and sampling replicants (14 and 15, 21 and 22, 31 and 32, 41 and 42) were taken 1 m apart.

At sites, soil was sampled somewhere on the right side of the road (Ivančna Gorica and Dedni dol), somewhere on the left side of the road (Družinska vas) and somewhere on both sides of the road (Krakovski gozd and Dob). „Right“ and „left“ represents the position according to the direction of driving from Obrežje to Ljubljana. It was concluded in the previous research (Zupančič, 1997) that along this highway prevailing wind direction as well as the traffic direction and

Table 2. Description of samples.
Legend: Sample – sample number; Profile – locality name (Kr. g. – Krakovski gozd, Dr. v. – Družinska vas, Dob – Dob; Iv. G. – Ivančna Gorica, De. d. – Dedni dol); Distance – distance from the road in m; Position – position according to Obrežje – Ljubljana direction (l –left, r – right; Dob: i.t.s – in front of the toll station, b.t.s. – behind the toll station); Morphology – road cut, mound, plane, slope; Parent rock – clay, Q –sand, limestone, dolomite; Vegetation – grass, bush, absent; Bedrock fragments – amount of bedrock fragments in %.
Tabela 2. Opis vzorcev tal.
Legenda: Vzorec – številka vzorca; Profil – ime lokacije (Kr. g. – Krakovski gozd, Dr. v. – Družinska vas, Dob – Dob; Iv. G. – Ivančna Gorica, De. d. – Dedni dol); Razdalja – oddaljenost od ceste v m; Položaj – glede na smer vožnje od Obrežja proti Ljubljani (l – levo, r– desno; Dob: i.t.s – pred cestninsko postajo, b.t.s. – za cestninsko postajo); Oblikovanost – usek, nasip, ravnina, pobočje; Matična kamnina – glina, kremenov pesek, apnenec, dolomit; Poraslost – trava, grmovje, odsotna; Skelet – količina skeleta v %.

Sample	Profile	Distance	Position	Morphology	Parent rock	Vegetation	Bedrock fragments
Vzorec	**Profil**	**Razdalja**	**Položaj**	**Oblikovanost**	**Matična kamnina**	**Poraslost**	**Skelet**
11	Kr. g.	1	l	road cut (0.8 m)	clay	grass	50
12	Kr. g.	5	l	plane	clay	bush	25
13	Kr. g.	10	l	plane	clay	bush	5
14, 15	Kr. g.	1	r	road cut (0.8 m)	clay	grass	50
16	Kr. g.	5	r	plane	clay	absent	10
17	Kr. g.	10	r	plane	clay	absent	1
21, 55, 22	Dr. v.	1	l	mound (0.7 m)	Q sand	absent	25
23	Dr. v.	5	l	mound (0.7 m)	Q sand	absent	25
24	Dr. v.	10	l	plane	Q sand	grass	5
31, 32	Dob	1	l (i.t.s.)	mound (2 m)	limestone	grass	30
33	Dob	5	l (i.t.s.)	mound (2 m)	limestone	grass	30
34	Dob	10	l (i.t.s.)	plane	limestone	grass	5
35	Dob	1	r (i.t.s.)	mound (1 m)	limestone	grass	30
36	Dob	5	r (i.t.s.)	mound (1 m)	limestone	grass	30
37	Dob	1	r (b.t.s.)	plane	limestone	grass	30
38	Dob	5	r (b.t.s.)	plane	limestone	grass	35
39, 57	Dob	10	r (b.t.s.)	plane	limestone	grass	35
41, 56, 42	Iv. G.	1	r	road cut (8 m)	limestone	grass	10
43	Iv. G.	5	r	road cut (8 m)	limestone	grass	5
44	Iv. G.	10	r	road cut (8 m)	limestone	grass	5
51	De. d.	1	r	plane	dolomite	grass	50
52	De. d.	5	r	plane	dolomite	grass	30
53	De. d.	10	r	slope	dolomite	grass	40

consequently the side of the road have little effect on the contamination pattern.

To evaluate if terrain features close to the road affected contamination levels, sampling localities were chosen in the way that all possible morphologies (road-cut, mound, plain, slope) were included. Additionally the parent rock, vegetation type and the percent of bedrock fragments were also determined (Table 2).

The soil samples were air dried. About 1 kg of every sample was ground, split and sieved to a 20 g sample with grain size under 0.063 mm. Before and between sieving we tried to remove as much bedrock fragments as possible, but we did not succeed entirely.

Selected localities

Profile Krakovski gozd

The profile Krakovski gozd was the most distant from Ljubljana. In the time of sampling (July 2004) the traffic was still running on old two-laned high-speed road H1 along which on the right side (the position according to the direction of driving from Obrežje to Ljubljana) half part of the highway was under construction. In July 2004 the soil was just ploughed up and cloddy leveled on the right side of the road at 5 m and 10 m distances and it waited for further highway construction works. Compared to the year 1994, the traffic on this road section grew by 82 % in the year 2003. In the year 2003 the average daily traffic was 14000 vehicles per day (Ministry of Transport of the Republic of Slovenia, Directorate of the Republic of Slovenia for Roads, 1995, 2004).

Profile Družinska vas

The distance between the profile Družinska vas and the profile Krakovski gozd was approximately 15 km. The profile lied at four-laned highway section Kronovo - Smednik, which was opened to traffic on 02.07.2004, just 14 days before sampling. The highway was reconstructed from two-laned high-speed road (web site Dars d.d.). In the year 2003 there were 13000 vehicles per day on this road section (Ministry of Transport of the Republic of Slovenia, Directorate of the Republic of Slovenia for Roads, 2004).

Profile Dob

The profile Dob was approximately 43 km distant from the profile Družinska vas. The profile lied at the toll station Dob and was divided in two parts; on profile in front of the toll station and profile behind the toll station (the position according to the direction of driving from Obrežje to Ljubljana). Profiles were 140 m apart. The profile Dob was located at highway section Višnja Gora-Bič, which was opened to traffic in the year 2000 as reconstruction of two-laned high-speed road into four-laned highway (web site Dars d.d.). Compared to 1994, the traffic on this road section grew by 84 % in 2003. Compared to the year 2000, when this highway section was opened, the traffic grew by 3% in the year 2003. In the year 2003 the average daily traffic was 15424 vehicles per day (Ministry of Transport of the Republic of Slovenia, Directorate of the Republic of Slovenia for Roads, 1995, 2001, 2004).

Profile Ivančna Gorica

The profile Ivančna Gorica and the profile Dob were 4 km apart. The profile Ivančna Gorica also lied at four-laned highway section Višnja Gora - Bič, which was opened to traffic in the year 2000 (web site Dars d.d.). Compared to 1994, the traffic on this road section grew by 87 % in 2003. Compared to the year 2000, when this road section was opened, the traffic grew by 23 % in the year 2003. In the year 2003 the average daily traffic was 28494 vehicles per day (Ministry of Transport of the Republic of Slovenia, Directorate of the Republic of Slovenia for Roads, 1995, 2001, 2004).

Profile Dedni dol

The distance between profiles Dedni dol and Ivančna Gorica was approximately 10 km.

The profile lied at two-laned highway section, which leads to Ljubljana. This highway section was newly constructed and opened to traffic in the year 1989. In the time of the first sampling in the year 1993 there were about 8000 vehicles per day on this highway section and in the time of the second sampling there were more than 14000 vehicles per day (Ministry of Transport of the Republic of Slovenia, Directorate of the Republic of Slovenia for Roads, 1995, 2004).

Mineralogical analysis

The powder X-ray diffraction measurements were made on Faculty of Natural Sciences and Engineering in Ljubljana using Philips diffractometer, equipped with a CuK_α radiation. The X-ray radiation was generated with a tension of 40 kV and a current of 30 mA and was filtered with a graphite monochromator. Data were recorded in the range $2° \leq 2\Theta \leq 70°$. For mineralogical analysis, 10 samples were prepared - two from each profile (from 1 m and 10 m distances from the road). Mineralogical composition of 10 selected samples is listed in Table 3.

Chemical analysis

Chemical composition of the samples was determined in the ACME Analytical Laboratories in Canada. Concentrations of major elements and heavy metals were determined by inductively coupled plasma (ICP). For major elements analysis a 2 g sample was digested in 5% HNO_3 after $LiBO_2$ fusion. The concentrations of major elements (expressed as common oxides) were determined by inductively coupled plasma (ICP – ES) method. Carbon and sulphur were determined by Leco method. For determination of heavy metal concentrations a 0.5 g sample was leached with 3 ml $HCl – HNO_3 – H_2O$ (2: 2: 2) mixture at 95 °C for one hour, then diluted to 10 ml and analysed by inductively coupled plasma (ICP – MS) method. For chemical analysis 30 samples were prepared (including three analytical replicants). Objectivity was assured through the use of neutral laboratory numbers. According to added analytical replicants (samples 55, 56, 57) the analytical quality is estimated as satisfactory. The difference between analytical replicants (samples 21 and 55, 39 and 57, 41 and 56) is

Table 3. Mineralogical composition of samples.
Legend: Sample – sample number; C + V + M – Chlorite + Vermiculite + Montmorillonite.
Tabela 3. Mineralna sestava vzorcev.
Legenda: Vzorec – številka vzorca; C + V + M – kloritova + vermikulitova + montmorillonitova skupina.

Sample Vzorec	C + V + M C + V + M	Kaolinite Kaolinit	Illite Illit	Quartz Kremen	Dolomite Dolomit	Calcite Kalcit	Feldspars Glinenci	Fe oxides Fe oksidi
11	*	*	*	*	*	*	*	*
13	*	*	*	*			*	*
21	*	*	*	*	*		*	*
24	*	*	*	*	*		*	
31	*	*	*	*	*		*	*
34	*	*	*	*			*	*
41	*		*	*	*		*	*
44	*	*	*	*			*	*
51	*		*	*	*		*	
53	*		*	*	*		*	*

Table 4. Concentrations of major elements, loss on ignition, carbon and sulphur in %.
Legend: Sample – sample number (v – sampling replicant, r – analytical replicant); LOI – loss on ignition; Tot/C – total C; Tot/S – total S.
Tabela 4. Vsebnosti glavnih prvin, žaroizgube, ogljika in žvepla v %.
Legenda: Sample – številka vzorca (v – vzorčna ponovitev, r – analitska ponovitev); LOI – žaroizguba; Tot/C – totalni C; Tot/S – totalni S.

Sample	SiO2	Al2O3	Fe2O3	MgO	CaO	Na2O	K2O	TiO2	P2O5	MnO	Cr2O3	LOI	Tot/C	Tot/S
11	28, 22	4, 75	2, 61	8, 98	14, 95	0, 49	0, 55	0, 39	0, 17	0, 07	0, 012	38, 6	12, 77	0, 13
12	62, 48	9, 41	3, 60	2, 44	3, 61	0, 59	1, 20	0, 83	0, 10	0, 07	0, 026	15, 5	4, 70	0, 07
13	53, 57	12, 99	4, 27	1, 25	1, 12	0, 68	1, 69	1, 07	0, 15	0, 13	0, 011	23, 0	7, 70	0, 08
14	22, 52	3, 30	1, 95	9, 92	17, 91	0, 43	0, 46	0, 26	0, 14	0, 06	0, 008	43, 0	14, 33	0, 07
15 v	21, 76	3, 24	1, 92	10, 52	18,42	0, 43	0, 46	0, 27	0, 14	0, 05	0, 009	42, 4	13, 29	0, 02
16	58, 16	14, 92	5, 37	1, 26	0, 92	0, 77	1, 90	1, 18	0, 12	0, 11	0, 013	15, 1	3, 52	0, 05
17	59, 93	15, 82	6, 07	1, 22	0, 49	0, 85	1, 99	1, 25	0, 13	0, 10	0, 013	11, 9	2, 38	0, 04
21	59, 80	11, 24	4, 56	2, 57	4, 67	0, 45	1, 26	1, 04	0, 14	0, 09	0, 019	14, 0	2, 51	0, 03
55 r	59, 45	11, 14	4, 69	2, 56	4, 74	0, 43	1, 41	1, 03	0, 16	0, 09	0, 020	14, 1	2, 45	0, 03
22 v	58, 17	12, 03	5, 10	2, 18	4, 27	0, 44	1, 16	1, 00	0, 14	0, 09	0, 020	15, 3	2, 43	0, 03
23	59, 26	11, 97	4, 91	2, 37	3, 53	0, 46	1, 22	1, 01	0, 13	0, 09	0, 019	14, 9	2, 51	0, 03
24	58, 02	10, 29	4, 52	2, 13	2, 54	0, 47	1, 25	1, 11	0, 24	0, 11	0, 015	19, 1	5, 69	0, 07
31	58, 62	15, 13	6, 77	1, 33	1, 11	0, 54	1, 72	1, 56	0, 36	0, 22	0, 024	12, 4	2, 17	0, 04
32 v	58, 15	15, 04	6, 60	1, 34	1, 14	0, 55	1, 51	1, 57	0, 36	0, 22	0, 024	13, 1	2, 15	0, 04
33	54, 87	15, 53	7, 25	1, 72	1, 70	0, 32	1, 27	1, 57	0, 44	0, 23	0, 024	14, 8	2, 57	0, 02
34	57, 04	15, 79	7, 12	0, 73	0, 53	0, 24	1, 39	1, 65	0, 64	0, 25	0, 025	14, 4	2, 41	0, 04
35	51, 53	13, 57	5, 55	3, 80	5, 00	0, 55	1, 43	1, 42	0, 36	0, 13	0, 017	16, 4	3, 23	0, 03
36	40, 53	11, 05	4, 93	2, 76	15, 24	0, 31	1, 29	0, 79	0, 22	0, 20	0, 019	22, 5	4, 73	0, 02
37	63, 31	13, 84	5, 71	0, 69	0, 63	0, 36	0, 98	0, 96	0, 50	0, 13	0, 016	12, 7	2, 16	0, 03
38	55, 14	12, 94	5, 13	2, 48	3, 63	0, 26	0, 97	0, 94	0, 49	0, 12	0, 014	17, 6	4, 13	0, 05
39	49, 24	11, 26	4, 40	4, 36	6, 40	0, 31	1, 05	1, 04	0, 42	0, 15	0, 014	21, 2	5, 14	0, 04
57 r	48, 96	10, 97	4, 50	4, 68	6, 81	0, 26	1, 00	1, 01	0, 36	0, 14	0, 014	21, 0	5, 08	0, 04
41	59, 01	13, 02	5, 19	2, 45	3, 03	0, 72	1, 66	1, 15	0, 29	0, 15	0, 012	13, 1	2, 90	0, 03
56 r	57, 57	12, 50	5, 12	2, 67	3, 66	0, 66	2, 13	1, 09	0, 30	0, 15	0, 013	14, 4	3, 37	0, 03
42 v	59, 41	12, 58	5, 06	2, 31	2, 98	0, 74	1, 58	1, 14	0, 30	0, 16	0, 012	13, 5	3, 10	0, 04
43	36, 58	23, 83	9, 48	1, 41	1, 56	0, 24	1, 35	1, 31	1, 40	0, 29	0, 022	22, 4	3, 86	0, 06
44	41, 59	19, 72	8, 02	1, 13	1, 05	0, 29	1, 51	1, 46	1, 08	0, 28	0, 022	23, 6	5, 67	0, 07
51	16, 79	3, 53	2, 66	13, 43	24, 58	0, 40	0, 42	0, 34	0, 12	0, 04	0, 017	37, 4	10, 59	0, 05
52	32, 96	12, 48	5, 01	8, 38	11, 08	0, 37	1, 18	0, 80	0, 11	0, 10	0, 013	27, 3	6, 62	0, 03
53	24, 54	10, 28	3, 97	10, 65	14, 37	0, 28	1, 22	0, 57	0, 14	0, 07	0, 010	33, 9	8, 89	0, 03

mostly less than 10 %. Concentrations of major elements are presented in Table 4 and concentrations of heavy metals are presented in Table 5.

Results and Discussion

The samples are composed in general of clay minerals, quartz, carbonates, feldspars and iron oxides. In accordance with data reported by Kabata-Pendias and Pendias (1986) heavy metals could be adsorbed in investigated samples by clay minerals, Fe-oxides and/or organic matter. They could also be incorporated in the structure of carbonates.

The results of first sampling in the year 1993 along the two-lane high-speed road Ljubljana-Obrežje (Zupančič, 1994, 1997)

Table 5. Heavy metal concentrations in ppm.
Legend: Sample – sample number (v – sampling replicant, r – analytical replicant); Profile – locality name (Kr. g. – Krakovski gozd, Dr. v. – Družinska vas, Dob – Dob; Iv. G. – Ivančna Gorica, De. d. – Dedni dol); Distance – distance from the road in m; Position – position according to Obrežje – Ljubljana direction (l –left, r – right; Dob: i.t.s – in front of the toll station, b.t.s. – behind the toll station).

Tabela 5. Vsebnosti težkih kovin v ppm.
Legenda: Vzorec – številka vzorca (v – vzorčna ponovitev, r – analitska ponovitev); Profil – ime lokacije (Kr. g. – Krakovski gozd, Dr. v. – Družinska vas, Dob – Dob; Iv. G. – Ivančna Gorica, De. d. – Dedni dol); Razdalja – oddaljenost od ceste v m; Položaj – glede na smer vožnje od Obrežja proti Ljubljani (l – levo, r– desno; Dob: i.t.s – pred cestninsko postajo, b.t.s. – za cestninsko postajo).

Sample Vzorec	**Profile Profil**	**Distance Razdalja**	**Position Položaj**	**Pb**	**Zn**	**Cu**	**Ni**	**Cd**
11	Kr. g.	1	l	481, 4	321	73, 9	23, 1	0, 9
12	Kr. g.	5	l	267, 4	157	41, 3	18, 0	0, 6
13	Kr. g.	10	l	149, 6	95	19, 4	20, 6	0, 4
14	Kr. g.	1	r	516, 0	179	42, 3	19, 3	0, 5
15 v	Kr. g.	1	r	446, 7	179	42, 4	18, 8	0, 6
16	Kr. g.	5	r	89, 8	80	16, 7	20, 8	0, 2
17	Kr. g.	10	r	35, 2	60	13, 9	21, 8	0, 1
21	Dr. v	1	l	30, 4	51	15, 3	26, 8	0, 6
55 r	Dr. v	1	l	28, 3	47	14, 0	25, 3	0, 6
22 v	Dr. v	1	l	31, 3	53	17, 8	29, 6	0, 8
23	Dr. v	5	l	40, 6	63	18, 2	30, 1	0, 8
24	Dr. v	10	l	52, 2	66	16, 2	17, 8	0, 7
31	Dob	1	l (i.t.s.)	32, 6	84	23, 7	27, 3	1, 8
32 v	Dob	1	l (i.t.s.)	36, 1	73	23, 7	27, 2	1, 9
33	Dob	5	l (i.t.s.)	32, 6	71	23, 1	24, 3	1, 7
34	Dob	10	l (i.t.s.)	35, 6	81	27, 2	27, 4	1, 1
35	Dob	1	r (i.t.s.)	35, 0	71	20, 4	22, 3	1, 2
36	Dob	5	r (i.t.s.)	18, 9	60	24, 5	45, 7	1, 4
37	Dob	1	r (b.t.s.)	28, 4	76	34, 8	22, 8	0, 8
38	Dob	5	r (b.t.s.)	32, 2	65	30, 0	21, 1	0, 8
39	Dob	10	r (b.t.s.)	24, 4	61	20, 1	17, 5	0, 8
57 r	Dob	10	r (b.t.s.)	25, 0	62	20, 9	21, 2	0, 9
41	Iv. G.	1	r	35, 4	105	22, 2	22, 1	1, 2
56 r	Iv. G.	1	r	37, 5	85	22, 7	22, 4	1, 1
42 v	Iv. G.	1	r	39, 0	90	24, 2	23, 4	1, 3
43	Iv. G.	5	r	130, 6	170	50, 4	48, 7	4, 9
44	Iv. G.	10	r	104, 9	130	37, 7	31, 2	1, 2
51	De. d.	1	r	226, 7	873	54, 7	21, 2	0, 7
52	De. d.	5	r	45, 1	57	15, 3	21, 8	0, 6
53	De. d.	10	r	24, 0	44	11, 4	18, 2	0, 4

showed that Pb, Zn, Cu and Ni concentrations decreased with distance from the road (Table 6). The reconstruction of two-lane high-speed road Ljubljana – Obrežje into four-lane highway in the years after the first sampling seemingly changed this trend of heavy metal distribution in some profiles. Last year this trend was confirmed only in two profiles (in Dedni dol and partly in Krakovski gozd – only on the left side), where circumstances stayed unchanged since the first sampling in 1993. In these two profiles the levels of Pb, Zn, Cu and Cd decreased with distance from the highway last year (Table 6). At the profile Dedni dol the highway was already constructed in 1989. At the profile Krakovski gozd the initial construction works have just started in July 2004 on the right side of the old two-lane high-speed road.

Most likely the heavy metal concentrations in profiles Krakovski gozd and Dedni dol are higher at 1 m distance from the road. In the profile Dedni dol also at 10 m distance. Insufficient removal of small sized bedrock fragments (mostly dolomite) from these samples at sieving (because of its large quantity) influenced the reduction in heavy metal concentrations. High amount of dolomite fragments in these sampling spots was already recognized during sampling and sieving (Table 2). X-ray diffraction also showed relatively higher amount of dolomite in Krakovski gozd at 1 m distance (left and right) and in Dedni dol at 1 m and 10 m distance (Figure 1). At 1 m distance from the road in the profile Krakovski gozd dolomite

Table 6. Comparison of heavy metal concentrations (in ppm), measured in the years 1993 and 2004.
Legend: Profile – locality name (Kr. g. – Krakovski gozd, Dr. v. – Družinska vas, Dob – Dob; Iv. G. – Ivančna Gorica, De. d. – Dedni dol); Distance – distance from the road in m; Position – position according to Obrežje – Ljubljana direction (l – left, r – right; Dob: i.t.s – in front of the toll station).
Tabela 6. Primerjava vsebnosti težkih kovin (v ppm), izmerjenih leta 1993 in 2004.
Legenda: Profil – ime lokacije (Kr. g. – Krakovski gozd, Dr. v. – Družinska vas, Dob – Dob; Iv. G. – Ivančna Gorica, De. d. – Dedni dol); Razdalja – oddaljenost od ceste v m; Položaj – glede na smer vožnje od Obrežja proti Ljubljani (l – levo, r – desno; Dob: i.t.s – pred cestninsko postajo).

Profile	Distance	Position	Pb		Zn		Cu		Ni	
Profil	Razdalja	Položaj	1993	2004	1993	2004	1993	2004	1993	2004
Kr. g.	1	l	457, 5	481, 4	236	321	67, 0	73, 9	43, 5	23, 1
Kr. g.	5	l	83, 0	267, 4	84	157	18, 0	41, 3	22, 0	18, 0
Kr. g.	10	l	53, 0	149, 6	58	95	12, 0	19, 4	19, 0	20, 6
Kr. g.	1	r	23, 0	481, 4	20	179	13, 0	42, 3	25, 0	19, 1
Kr. g.	5	r	17, 5	89, 9	57	80	12, 0	16, 7	24, 5	20, 8
Kr. g.	10	r	34, 0	35, 2	61	60	11, 0	13, 9	23, 5	21, 8
Dr. v.	1	l	375, 0	30, 0	326	50	89, 0	15, 7	55, 0	27, 2
Dr. v.	5	l	59, 0	40, 6	79	63	20, 0	18, 2	27, 0	30, 1
Dr. v.	10	l	37, 0	52, 2	70	66	17, 0	16, 2	27, 0	17, 8
Dob	1	l (i.t.s.)	398, 0	34, 4	233	79	51, 0	23, 7	33, 0	27, 3
Dob	5	l (i.t.s.)	45, 0	32, 6	64	71	17, 0	23, 1	22, 0	24, 3
Dob	10	l (i.t.s.)	27, 0	35, 6	54	81	17, 0	27, 2	21, 0	27, 4
Dob	1	r (i.t.s.)	374, 0	35, 0	191	71	35, 0	20, 4	40, 0	22, 3
Dob	5	r (i.t.s.)	58, 0	18, 9	74	60	18, 0	24, 5	23, 0	45, 7
Iv. G.	1	r	765, 0	37, 3	420	93	104, 0	23, 0	61, 0	22, 6
Iv. G.	5	r	120, 0	130, 6	143	170	38, 0	50, 4	40, 0	48, 7
Iv. G.	10	r	28, 0	104, 9	78	130	27, 0	37, 7	27, 0	31, 2
De. d.	1	r	381, 0	226, 7	617	873	52, 0	54, 7	47, 0	21, 2
De.d.	5	r	24, 0	45, 1	52	57	16, 0	15, 3	45, 0	21, 8
De. d.	10	r	34, 0	24, 0	210	44	20, 0	11, 4	56, 0	18, 2

was metalled at the bottom of only 4 cm deep soil profile. In the profile Dedni dol dolomite is parent rock. Elevated contents of MgO, CaO and tot C (Table 4) in these sampling spots also indicates elevated content of carbonates in soil. The values of Pb, Zn and Cu in these two profiles have increased since the year 1993, but direct comparison between samples from the years 1993 and 2004 is not possible due to the reduction of concentrations in mentioned sampling spots.

The other three profiles (Družinska vas, Dob and Ivančna Gorica) were laid in the year 1993 at two-lane high-speed road and in the year 2004 at four-lane highway. In the year 1993 heavy metal content in these three profiles also decreased with distance from the road. During widening of the high-speed road into the highway the "old" roadside topsoil, contaminated with heavy metals, was affected by construction works. This led to the exchange of soil at the top. "New" topsoil close by the highway contained only background concentrations. Sampling of this "new" roadside soil at 1 m and somewhere at 5 m distance from the road and sampling of "old" soil at longer distances in the same profile changed the trend of heavy metal distribution, which was recognized in 1993. In the profiles Družinska vas, Dob and Ivančna Gorica concentrations of Pb, Zn, Cu and Ni at 1 m distance were lower than at longer distances from the road although in the year 1993 were higher (Table 6). Lower concentrations in the "new" soil along reconstructed highway sections (in profiles Družinska vas, Dob and Ivančna Gorica) could be connected with a short period of time, when the soil was exposed to traffic. Low correlation coefficients between distance from the road and last year Pb (-0.352) and Zn (-0.351) concentrations (Figures 2 and 3) corroborate upper statements. Direct comparison between samples from the years 1993 and 2004 is also not possible in these three profiles, because of reconstruction works.

This research confirmed that road traffic was an important source of lead contamination in soil (ZUPANČIČ, 1997), but introduction of unleaded petrol and prohibition of selling leaded petrol in Slovenia from the year 2001 has diminished its influence. High concentrations of Pb (Table 5) were measured along the road sections, which have been opened to traffic for longer time (profiles Krakovski gozd and Dedni dol). Values were up to 516 ppm. According to Slovenian legislation, the alerting concentration (Official Gazette, 68/96) of Pb in soil is 100 ppm and the chritical concentration (Official Gazette, 68/96) is 530 ppm. As stated above the concentrations of heavy metals in these two profiles at 1 m distance from the road are most likely higher and were reduced due to insufficient removal of small sized bedrock fragments from samples at sieving. In accordance with previous studies (MARKUS AND MCBRATNEY, 2001; OLAJIRE AND AYODELE, 1997; VIARD, 2004; ZUPANČIČ, 1997) lead concentrations decrease with distance from the road, indicating their relation to road traffic. The roadside distribution of Pb is ascribed to combustion of leaded petrol by automobiles (VAN BOHEMEN AND VAN DE LAAK, 2003). Roadside soils along the road sections, which have been opened to traffic for shorter time – from the year 2000 onwards (profiles Družinska vas, Dob and Ivančna Gorica) contained lower Pb concentrations. During widening the high-speed road into the highway, highly Pb contaminated topsoil was replaced on the surface with soil, which contained only background values. Due to prohibition of selling leaded petrol in Slovenia from the year 2001 the Pb values stayed mainly unchanged. In profiles Družinska vas, Dob and Ivančna Gorica Pb concentrations at 1 m distance from the road do not exceed 40 ppm.

Concentrations, higher than the alerting value (Official Gazette, 68/96) 300 ppm were also measured for zinc (Table 5), but only at 1 m distance from the road in the profiles Dedni dol and Krakovski gozd. In Dedni dol the values were up to 873 ppm, which exceeds also the critical value (Official Gazette, 68/96) of Zn 720 ppm. In Krakovski gozd the highest value was 321 ppm. Zinc concentrations in these two profiles can be connected with road traffic because they decrease with distance from the road. This is in accordance with previous studies (Olajire and Ayodele, 1997; Viard, 2004). Zinc is mainly derived from tire dust (to automobile tires is added as a filler and is released by tire wear) (Davis et al., 2001; Van Bohemen and Van de Laak, 2003).

Concentrations of copper (Table 5) do not exceed the alerting value (Official Gazette, 68/96) 100 ppm in any analysed soil sample. The highest value (73.9 ppm) was measured in profile Krakovski gozd at 1 m distance from the road. Decrease in copper concentrations with distance from the road in profiles Krakovski gozd and Dedni dol can be connected with road traffic. Lower copper concentrations indicate that road traffic is minor Cu contributor to soil contamination. Copper is mainly derived from the corrosion of radiators and brakes (Davis et al., 2001; Van Bohemen and Van de Laak, 2003).

The highest cadmium concentrations (Table 5) were observed on the left side of the profile Dob in front of the toll station (the position according to the direction of driving from Obrežje to Ljubljana). The highest value (1.85 ppm) was measured at 1 m from the highway. On the right side of the profile Dob in front of the toll station as well as behind the toll station and also in other profiles the values were lower at least for 30% than the highest value measured. In profiles Dedni dol and Krakovski gozd Cd concentrations decrease with distance from the road. This distribution trend agrees with data reported by Olajire and Ayodele (1997) and Viard et al. (2004). The alerting value (Official Gazette, 68/96) for Cd is 2 ppm. The value 4.9 ppm, which was measured in the profile Ivančna Gorica at 5 m distance from the highway, was most likely an analytical error. The road traffic is in view of low Cd concentrations only minor contributor to soil contamination with cadmium. Cadmium is in small quantities derived from exhaust, tires and brakes (Davis et al., 2001; Van Bohemen and Van de Laak, 2003).

The nickel levels (Table 5) were in all analysed soil samples lower than the boundary value (Official Gazette, 68/96) 50 ppm. The values range between 17 ppm and 49 ppm. Nickel did not show decrease in values with distance from the road in any profile although this trend could be masked in our case. As stated above the concentrations of heavy metals in profiles Dedni dol and Krakovski gozd at 1 m distance from the road are most likely higher and were reduced due to insufficient removal of small sized bedrock fragments from samples at sieving. In the year 1993 the nickel values decreased with distance from the road (Table 6) (Zupančič, 1994). In any case the road traffic could be only minor Ni contributor to soil contamination.

It has not passed enough time since the opening of the toll station Dob in the year 2000 so it is difficult to conclude to what extent the toll station contributes to enhancement of heavy metal content in soil. From Table 5 it is evident that none of heavy metals exceed the alerting value (Official Gazette, 68/96) Only cadmium exceeds the boundary value (Official Gazette, 68/96).

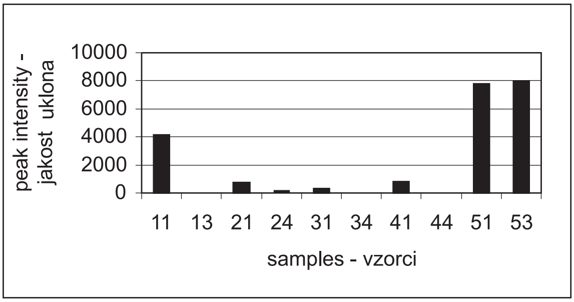

Figure 1. Peak intensities (d = 0.289 nm) of dolomite.
Slika 1. Jakosti uklona (d = 0.289 nm) dolomita.

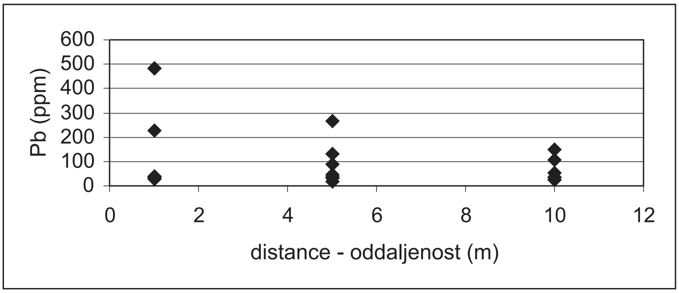

Figure 2. Variation of Pb content in topsoil with distance from highway.
Slika 2. Spreminjanje vsebnosti Pb v najzgornejšem delu tal z oddaljenostjo od avtoceste.

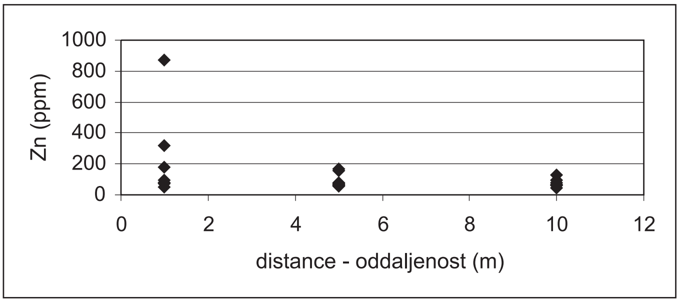

Figure 3. Variation of Zn content in topsoil with distance from highway.
Slika 3. Spreminjanje vsebnosti Zn v najzgornejšem delu tal z oddaljenostjo od avtoceste.

It was concluded in the previous study along this road (Zupančič, 1997) that near roadways terrain morphology affects the distribution of lead contamination. Higher lead values were found in road-cuts (probably as consequence of obstructed dispersion) and in slopes (connected with driving mode). Last year the samples were also taken along reconstructed highway sections with lower concentrations in the "new" soil (profiles Družinska vas, Dob and Ivančna Gorica). That is why it was impossible to compare the values from different morphologies (road-cuts, mounds, slopes and plains).

Conclusions

Lead can be confirmed as the most distinctive heavy metal from road traffic pollution, though levels of zinc, copper and cadmium in roadside soil can be also connected with road traffic. The levels of these metals in roadside soils decreased with distance from the road in the profiles (Dedni dol and Krakovski gozd) at road sections, which were not included in highway reconstruction works in years after the first sampling. The other three profiles (Ivančna Gorica, Dob and Družinska vas) were laid down in the year 1993 at two-lane high-speed road and in the year 2004 at four-lane highway. In the year 1993 Pb, Zn and Cu contents in these three profiles also decreased with distance from the road (Zupančič, 1994, 1997). The widening of two-lane high-speed road seemingly changed this trend of heavy metal distribution. The highly contaminated "old" roadside topsoil was at widening removed or covered with "new" topsoil, which contained only background values. Low heavy metal concentrations in the "new" soil along reconstructed highway sections (in profiles Družinska vas, Dob and Ivančna Gorica) could be connected with a short period of time, in which the soil was exposed to traffic. Low Pb content also reflects the prohibition of selling leaded petrol in Slovenia from the year 2001.

Direct comparison between profiles from the years 1993 and 2004 is not possible. Profiles Ivančna gorica, Dob and Družinska vas were laid in the year 1993 at two-lane high-speed road and in the year 2004 at four-lane highway. In profiles Krakovski gozd (at 1 m distance from the road) and Dedni dol (at 1 m and 10 m distance from the road) the values were most likely reduced due to insufficient removal of small sized bedrock fragments (mostly dolomite) at sieving (because of its large quantity).

Heavy metals could be adsorbed in investigated samples by clay minerals, Fe-oxides and/or organic matter. They could also be incorporated in the structures of carbonates.

It has not passed enough time since the opening of the toll station Dob in the year 2000 so it is difficult to conclude to what extent the toll station contributes to enhancement of heavy metal content in soil. None of the heavy metals exceed the alerting value (Official Gazette, 68/96), only cadmium exceeds the boundary value (Official Gazette, 68/96).

POVZETEK

Onesnaženje tal s težkimi kovinami vzdolž avtoceste Ljubljana – Obrežje

Namen raziskave je bil prikaz sprememb onesnaženja tal s težkimi kovinami (Pb, Zn, Cu, Cd, Ni), ki so nastale v obdobju zadnjih desetih let vzdolž bodočega dolenjskega avtocestnega kraka Ljubljana-Obrežje. Prvo vzorčenje tal je bilo izvedeno leta 1993 (ZUPANČIČ, 1994, 1997). Zaradi močnega povečanja gostote prometa vzdolž te trase v zadnjih desetih letih (za približno 80 %), večanja deleža prodaje neosvinčenega bencina in prepovedi prodaje osvinčenega bencina v Sloveniji v letu 2001 smo leta 2004 ponovili vzorčenje tal. Vzorčili smo v petih profilih (Krakovski gozd, Družinska vas, Dob, Ivančna Gorica in Dedni dol), ki so ležali čim bližje tistim iz leta 1993. Leta 1993 so bili profili večinoma locirani ob dvopasovni hitri cesti (razen profila Dedni dol, ki je ležal ob avtocesti). Dvopasovna hitra cesta je bila v letih po prvem vzorčenju večinoma razširjena v štiripasovno avtocesto, tako da so leta 2004 profili večinoma ležali ob njej. Na odseku, ob katerem je ležal profil Krakovski gozd, pa so potekala razširitvena dela.

Predhodna raziskava (ZUPANČIČ, 1994, 1997) je pokazala, da so najvišje vsebnosti težkih kovin v 10 m pasu oddaljenosti od ceste, ter v najzgornejšem (0-5 cm globine) delu tal. Zato smo v pričujoči raziskavi vzorčili le zgornjih 5 cm tal v oddaljenosti 1 m, 5 m in 10 m levo in/ali desno od roba ceste (gledano v smeri proti Ljubljani). Podatki o vzorčnih točkah so zbrani v tabeli 2.

Celotno količino (1 kg) vzorca smo posušili na zraku, presejali in zmleli do velikosti delcev, manjše od 0.063 mm. Pred in med sejanjem smo skušali odstraniti čim več drobcev skeleta, vendar to ni bilo mogoče v popolnosti.

Izbranim desetim vzorcem, dvema iz vsakega profila (z oddaljenosti 1 m in 10 m od ceste), smo z rentgensko difrakcijo določili mineralno sestavo (tabela 3). Tridesetim vzorcem (vključno z vzorčnimi in analitskimi ponovitvami) pa je bila, z metodo induktivno vezane plazme (ICP), določena kemijska sestava (tabeli 4 in 5).

Mejne, opozorilne in kritične imisijske vrednosti težkih kovin v tleh (Uradni list RS, št. 68/96) so predstavljene v tabeli 1.

Preiskane vzorce tal (tabela 3) v splošnem sestavljajo: glineni minerali, kremen, karbonati, glinenci ter železovi oksidi in hidroksidi. V skladu s podatki, ki jih poročata KABATA-PENDIAS IN PENDIAS (1986), so se težke kovine v preiskanih vzorcih lahko absorbirale na glinene minerale, Fe-okside in /ali organsko snov. Lahko so se vključile tudi v kristalne rešetke karbonatov.

Lahko potrdimo, da je cestni promet prispeval k onesnaženju tal predvsem s svincem (vsebnosti do 516 ppm), vendar tudi vsebnosti cinka (do 873 ppm), bakra (do 74 ppm) in kadmija (do 1.9 ppm) lahko povežemo s cestnim prometom (tabela 5). Vsebnosti teh kovin upadajo z oddaljenostjo od ceste, vendar le ob cestnih odsekih (profila Dedni dol in Krakovski gozd – le leva stran ceste), ki jih niso zajela razširitvena dela v času po prvem vzorčenju. Profil Dedni dol leži ob avtocestnem odseku, ki je bil zgrajen že leta 1989. Profil Krakovski gozd leži ob cestnem odseku, vzdolž katerega se je na

desni strani, v času lanskega vzorčenja, šele začela gradnja avtocestnega pasa. Ostali trije profili (Ivančna Gorica, Dob in Družinska vas) so leta 1993 ležali ob dvopasovni hitri cesti, leta 2004 pa ob štiripasovni avtocesti. Leta 1993 so vsebnosti Pb, Zn and Cu v teh treh profilih prav tako upadale z oddaljenostjo od ceste (tabela 6) (Zupančič, 1994, 1997). Izgleda, da so razširitvena dela navidezno spremenila trend upadanja vsebnosti z oddaljenostjo od ceste. Visoko kontaminirani površinski del tal, ki je bil vzorčen leta 1993, je bil ob razširitvi ceste odstranjen ali prekrit z novim horizontom tal, ki je vseboval le naravne vrednosti težkih kovin. Nizke koncentracije težkih kovin v novih tleh (profili Družinska vas, Dob in Ivančna Gorica) so povezane s kratkim časom izpostavljenosti tal cestnemu prometu. Nizka vsebnost svinca v teh tleh je povezana tudi s prepovedjo prodaje osvinčenega bencina v Sloveniji v letu 2001.

Neposredna primerjava med vzorci iz leta 1993 (prvo vzorčenje) in 2004 ni možna. Cestni odseki, ob katerih ležijo profili Družinska vas, Dob in Ivančna Gorica, so bili v letih po prvem vzorčenju razširjeni v avtocesto. V profilih Krakovski gozd in Dedni dol pa so bile na oddaljenosti 1 m od ceste vsebnosti težkih kovin razredčene, najverjetneje zaradi nepopolno odstranjenega (predvsem dolomitnega) skeleta pri sejanju. Vzorčne točke, Krakovski gozd 1 m (levo in desno od ceste) ter Dedni dol 1 m in 10 m, so vsebovale okrog 50 % (predvsem dolomitnega) skeleta (tabela 2), ki ga pri sejanju nisem mogel popolnoma odstraniti, zaradi visoke količine majhnih delcev. Na višjo vsebnost karbonatov v omenjenih vzorcih kažejo višje vsebnosti MgO, CaO in C (tabela 4). Tudi rentgenska difrakcija (slika 1) je pokazala, da ti vzorci vsebujejo relativno višjo vsebnost dolomita. Dolomit izvira deloma iz nasutja cestne podlage, deloma iz nasipanja proti zimskemu drsenju vozil, v profilu Dedni dol je vir dolomita tudi matična podlaga.

V profilih Krakovski gozd (z izjemo vzorcev 5 m in 10 m desno) in Dedni dol, kjer so bila pri obeh raziskavah vzorčena ista tla, je nakazano povišanje vsebnosti Pb, Zn in Cu v primerjavi z letom 1993 (tabela 6), vendar dejanskega povišanja, zaradi razredčenja vsebnosti v omenjenih vzorčnih točkah, ni možno oceniti.

Za postavitev sklepa, ali cestninska postaja Dob doprinese k dodatnemu povišanju vsebnosti težkih kovin, in v kakšni meri, je preteklo premalo časa od njenega odprtja v letu 2000. Izmerjene vrednosti so, razen v primeru kadmija, veliko nižje od opozorilnih (Uradni list RS, št. 68/96).

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