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Chapter VI

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Relationship Between the Concentration of Trace Elements and the Physical-Chemical Properties of Polluted Soils Near Gas Stations in the Warsaw Agglomeration

INTRODUCTION

The progress of civilization and urbanization and accelerating industrialization has contributed to the continuous increase of the concentration of heavy metals in the environment. This trend, especially with regard to soils, brings in additional hazard for all living creatures.

In combustion gases and dusts the presence of lead as well as cadmium, zinc and other heavy metals can be identified. Cadmium frequently occurs in oil; it is also used as a component in the production cycles of tires; zinc – on the other hand – is part of grease [GORLACH, GAMBUŚ 2000, CURZYDŁO 1998]. Chromium is often emitted into the environment as a by-product of many industrial processes like e.g., paper production, metallurgical technologies, burning of carbon [ALLOWAY, AYRES 1999], combustion of liquid fuels and municipal wastes [DMUCHOWSKI et al. 2001]

With regard to nature protection and food production it is necessary to define the maximum accepted level and the unacceptable thresholds of heavy metals in soils for the vegetation of plants and for the crop quality, regardless the origin of the metals. It is commonly accepted, however, that strong dependence occurs between the natural concentration of heavy metals and soil drainage, particularly the concentration of colloidal particles. The high sorption capacity of colloids decreases the moving ability of heavy metals in the soil solution. Large influence on the solubility of heavy metals and thus their availability for plants has the pH level of the soil. This index should always be considered when assessing the ecological status of soils [SIUTA 1995]. The aim of this paper is to assess the effect of physical-chemical properties of soils on the content of trace elements in soils and the pollution of the soils in the vicinity of gas stations.

MATERIAL AND METHODS

The sub-surface layer of soils (0-20 cm) was studied in the vicinity (at 0, 10, 15, 20 and 100m distance) from 12 randomly selected liquid fuel stations located in the Warsaw agglomeration. The studied stations represented different traffic intensity. The study was carried out between 1999-2000. The following parameters were determined in the samples (collected twice a year):

- near-to-total content of heavy metals (Pb, Cd, Zn, Cu), in 20% HCl – with application of the ASA method (fire atom absorption);
- total organic carbon content (C%) – using the TOC 5000 A Shimadzu Analyzer, presented in the form of humus content following the Tiurin technique (PR%);
- loss in incandescence following the German standard DIN 18128 (11.90) at 550^{o}C, this value has been accepted in the present paper as the equivalent of the organic matter content OM [MYŚLIŃSKA 1998];
- grain size composition according to the Casagrande method in the modification of Prószyński;
- soil pH in 1 mol KCl·dm^{-3} determined potentiometrically.

The statistical analyses were performed with the use of Excel software.

The obtained results were compared to the regulations of the Ministry of the Environment [Dz. U. 02.165.13.59] and IUNG directives [KABATA-PENDIAS et al. 1995].

The complex evaluation of pollution in the studied soils was based on the synthetic coefficient of soil degradation [FORYCIARZ 1996], calculated according to the following formula:

$$S = \sqrt{(\frac{Zn}{70}) + (\frac{Pb}{50}) + (\frac{Cd}{0,5})}$$

where: Zn, Pb and Cd represent concentrations of metals in the sub-surface layer of soil in mg/kg, whereas the numbers in the denominators represent the boundary values of pollution for light soils according to IUNG [KABATA-PENDIAS et al. 1995]. In order to simplify the calculations it was assumed that all studied soils represented soils with neutral or slightly acidic reaction. Three heavy metals were included in the calculations – Zn, Pb and Cd. The following boundary pollution values were assumed: Zn – 70 mg·kg^{-1}; Pb – 50 mg·kg^{-1}; Cd – 0.5 mg·kg^{-1}.

RESULTS AND DISCUSSION

The statistical results of the studied physical-chemical properties of soils are presented in Table 1. Considering their grain size composition, the studied soils generally belong to the group of light soils. The pH in the KCl values range suggests that they are slightly acidic or neutral soils. The contents of humus and organic matter were all at a similar level and typical of most of soils in Poland [KONDRAS, CZĘPIŃSKA-KAMIŃSKA 2004].

The concentration of heavy metals in the 0-20cm layer was variable, depending both on the metal and the distance of the sample from the fuel station. The content of lead reached on the average 42.8 (median), varying from 6.4 to 246 mg·kg^{-1} (Table 2); zinc attained the average level of 95 mg·kg^{-1}, ranging widely from 19.1 to 289 mg·kg^{-1}; the content of cadmium was 0.6 mg/kg on the average, ranging between 0.2-1.5 mg·kg^{-1}; the concentration of copper reached the level of 16 mg·kg^{-1} (median), ranging between 3.2-58.9 mg·kg^{-1}. The geometrical mean of the synthetic coefficient of soil degradation for this layer was 2.1 mg·kg^{-1} and the median was 2.06 mg·kg^{-1}.

The concentration of the other three heavy metals studied, chromium, nickel and cobalt, did not vary significantly.

Table 1

The variability coefficient of selected physical-chemical properties of the studied soils

Parameter	pH in KCl	H_h mmol(+)·kg^{-1}	*PR%*	OM%	Fraction <0.02mm	Fraction <0.002mm
			%			
Minimum	4.12	0.3	0.49	0.79	4	0
Maximum	7.22	3.21	4.52	6.12	25	12
Arithmetical mean	6.32	1.23	2.00	2.67	16	5
Geometrical mean	6.30	1.07	1.81	2.43	15	5
Standard deviation	1.11	0.70	0.87	1.15	4	2
Median	6.47	1	1.94	2.58	16	5
Percentile (5-95%)	5.19-7.05	0.6-2.91	0.89-4.01	0.62-5.36	9-20	1-9

Table 2

The concentration of selected heavy metals (mg·kg^{-1}) in the studied soils

Element	Median	Range	Median	Range
	0-20 cm		0-10 cm	
Pb	42.8	6.4-246	57.3	8.9-238.9
Zn	95	19.1-289	133.2	24.2-402.3
Cd	0.6	0.2-1.5	0.6	0.2-1.5
Cu	16	3.2-58.9	26.1	5.5-110.2
Cr	10.9	4.2-21.5	14.2	4.8-22.1
Ni	7.5	3.2-14.6	11.6	6.0-22.0
Co	3.1	2.2-5.2	4.0	2.9-7.0

The study indicated several trends in the content soil heavy metals occurring near the gas stations. The highest concentration was found in the area of the station and within a 5-10 m distance from the station; beyond the concentration of heavy metals became lower, to increase at a distance of 15-20 m. A substantial decrease in the concentration of heavy metals in the soils near gas stations is clearly visible in soils sampled 100 m from the stations (Table 3). This trend is particularly visible for Pb, Cd, Cr, Cu and Ni, with respect to the median, geometrical and arithmetical means. On the other hand, however, neither the maximum nor the minimum concentration values follow this tendency. This may be consequence of the differences in size of particular study objects on the one hand, and the fact that the stations were located along communications routes of variable traffic intensity. In the case of Zn such relationship was not found. The concentration of this element in the studied soils is equally high, even at a distance of 100 m from the studied objects. The pattern in Co was similar; in this case no differences in the concentration depending on the distance from the gas station were found.

Considering the results of the degree of soil pollution with heavy metals as obtained according to IUNG methodology [KABATA-PENDIAS et al. 1995], none of the studied soil samples fell into category III of pollution; besides, the content of nickel and cadmium was in all cases within the range of the natural concentration. The results of this study are comparable with those obtained by the Provincial Inspection of Environment Protection (Wojewódzka Inspekcja Ochrony Środowiska WIOS) in Warsaw during regional monitoring carried out in the sub-surface soil layers (twice a year) along the main traffic road. Only in the case of cadmium, in a few samples the third (III) degree of pollution was determined [WIOŚ - WARSZAWA, 2001]. Similar was also the spatial distribution of pollution in the soil; in general, a noticeable decrease in the concentration of heavy metals occurred along a 100 m wide zone from the highway margin (Table 3), [KONDRAS 2003].

Considering the criteria specified in the Regulation of the Ministry of the Environment from October 4, 2002 [Dz.U.02.165.13.59], the maximum accepted lead content has only been exceeded in 6 samples. The concentration of all other heavy metals fell within the range specified by the ordinance. With regard to lead, polluted soils were found in the vicinity of three studied gas stations. These objects are located in Warsaw downtown, that is in an area with high anthropogenic impact [KONDRAS and CZĘPIŃSKA-KAMIŃSKA 2007]. Summing up, it can be concluded that, following the standards of the cited ordinance, only 4% of soils were polluted with lead.

The soil samples collected from 0-10cm were characterized by higher concentration of heavy metals than samples from 0-20 cm (Tables 3 and 4). The geometrical mean of the synthetic coefficient of soil degradation for this layer was 2.53, and the median was 2.62.

The synthetic coefficient of soil degradation by heavy metals may decrease the pollution index in cases when a given sample shows higher concentration of one metal or may increase the index when the values are exceeded for several metals. Coefficient values below 2 characterize areas with low content of heavy metals in soils, whereas areas with the coefficient exceeding 5 reflect significant pollution and should be subject to continuous monitoring [FORYCIARZ 1996, BARTUŚ 1997].

Comparison of the content of heavy metals in both studied soil layers showed that in layer 0-10cm the content of these elements was even several tens of percents higher than in layer 0-20 cm. This may result, as shown by previous reports, from the fact that metals in soils with neutral or alkaline reaction are insignificantly transported to the deeper parts of the soil profile [CZARNOWSKA 1978, CZARNOWSKA, KABATA-PENDIAS 1977, CZARNOWSKA et al. 1983, CZARNOWSKA 1999].

Summing up, it can be assumed that the heavy metal content in the sub-surface layer of soils in most of the studied samples lies within the average values noted in Poland and within the boundary values for the natural concentration of heavy metals.

Comparison of the obtained results with earlier reports [CZARNOWSKA 1999, KOZANECKA et al. 2000, WIOŚ 2001] does not allow recognizing an unequivocal trend in the changes of the heavy metals concentration in soils, although a falling tendency is slightly marked. The WIOS [2001] studies showed high variability in the same sampling sites in particular years, with a falling trend in the heavy metal content, what may result from the recently lower emission of heavy metals as well as washing out and transportation of the elements deeper into the soil profile.

Table 3

Statistical assessment of the results of heavy metals concentration depending on the distance from the gas station (mg·kg^{-1} of soil)

Element	Distance	A*	G*	Median	Standard deviation	Minimum	Maximum
Pb	Station	57.9	41.5	33.1	43.4	9.8	130.3
	5 m	48.1	36.6	47.6	29.6	8.8	94.8
	10 m	44.2	34.4	47.5	25.2	6.4	80.2
	15 m	59.3	39.7	43.6	59.7	9.1	246.0
	20 m	56.6	40.1	52.0	46.1	8.8	189.2
	100 m	40.7	31.8	39.7	23.8	7.9	77.9
Cd	Station	0.7	0.6	0.6	0.3	0.24	1.48
	5 m	0.7	0.6	0.6	0.3	0.36	1.1
	10 m	0.6	0.6	0.6	0.3	0.2	1.2
	15 m	0.6	0.6	0.6	0.2	0.2	1.0
	20 m	0.6	0.5	0.5	0.3	0.2	1.3
	100 m	0.5	0.5	0.4	0.3	0.28	1.3
Zn	Station	108.5	80.3	75.3	85.2	21.2	285.9
	5 m	119.3	91.9	107.5	79.1	19.8	275.4
	10 m	110.1	88.2	98.9	74.3	30.2	266.8
	15 m	118.6	92.3	98.2	82.0	20.1	289.4
	20 m	106.4	83.2	86.5	74.2	23.5	251.9
	100 m	110.0	82.8	95.4	75.5	19.1	257.8
Cr	Station	11.4	10.7	10.1	3.9	5.0	18.0
	5 m	11.9	11.2	11.8	4.2	6.0	21.5
	10 m	11.5	11.0	11.0	3.4	6.0	19.0
	15 m	11.5	10.9	11.6	3.8	4.5	18.5
	20 m	11.7	11.0	11.5	3.7	4.5	19.0
	100 m	10.6	9.6	10.7	4.6	4.2	20.0
Cu	Station	22.8	15.5	17.1	18.4	3.5	58.4
	5 m	20.7	14.9	20.2	14.8	3.4	50.0
	10 m	19.8	14.1	16.2	15.8	3.2	56.4
	15 m	17.5	13.6	15.3	11.9	3.8	40.3
	20 m	21.2	15.3	15.0	15.8	3.3	58.9
	100 m	17.8	12.8	13.2	14.8	3.3	57.8
Ni	Station	7.7	7.3	8.3	2.5	3.2	12.4
	5 m	8.2	7.7	8.5	2.7	3.9	12.4
	10 m	7.8	7.4	7.9	2.6	3.5	13.1
	15 m	7.7	7.4	7.9	1.9	3.9	11.2
	20 m	7.7	7.2	7.1	2.8	3.5	14.6
	100 m	6.7	6.4	6.7	1.9	3.3	10.0
Co	Station	3.3	3.2	3.2	0.7	2.2	5.2
	5 m	3.3	3.2	3.2	0.6	2.4	4.6
	10 m	3.2	3.2	3.2	0.4	2.5	4.1
	15 m	3.3	3.2	3.1	0.5	2.5	4.5
	20 m	3.1	3.1	3.0	0.4	2.5	4.0
	100 m	3.2	3.1	3.1	0.5	2.5	4.4

A – arithmetical mean, G – geometrical mean

Table 4

Statistical assessment of the results of heavy metals concentration depending on the distance from the gas station (mg·kg^{-1} of soil)

Element	Distance	A*	G*	Median	Standard deviation	Minimum	Maximum
Pb	Station	80.9	62.3	58.7	53.9	16.6	158.3
	5 m	62.8	49.0	64.6	38.6	11.4	139.3
	10 m	59.5	47.3	57.8	35.8	10.3	130.3
	15 m	71.7	48.9	56.7	64.0	10.9	238.9
	20 m	65.1	46.7	57.2	52.3	10.8	200.2
	100 m	46.9	36.2	49.1	28.2	8.9	92.9
Cd	Station	0.77	0.70	0.67	0.33	0.3	1.4
	5 m	0.76	0.70	0.65	0.32	0.4	1.3
	10 m	0.7	0.63	0.63	0.35	0.3	1.4
	15 m	0.6	0.5	0.4	0.4	0,2	1,5
	20 m	0,6	0.5	0.4	0.4	0.2	1.5
	100 m	0,5	0.4	0.4	0.3	0.2	1.3
Zn	Station	155.2	121.9	118.5	107.7	32.5	390.2
	5 m	165.8	128.0	158.2	107.2	28.3	353.4
	10 m	152.8	125.5	142.6	95.5	37.0	324.2
	15 m	153.4	122.5	151.3	95.0	32.1	342.1
	20 m	136.4	102.4	105.1	110.0	29.7	402.3
	100 m	128.7	99.7	128.7	87.5	24.2	329.8
Cr	Station	15.5	14.8	16.0	4.6	6.5	22.1
	5 m	15.2	14.6	15.0	4.4	7.0	20.1
	10 m	15.0	14.6	14.6	3.5	8.8	20.3
	15 m	14.8	14.1	14.5	4.0	5.3	19.3
	20 m	13.9	13.2	14.1	4.0	5.0	19.2
	100 m	12.1	11.1	12.0	4.7	4.8	19.2
Cu	Station	44.3	31.5	42.0	33.9	8.9	110.2
	5 m	34.8	26.9	31.6	24.6	7.5	88.6
	10 m	30.9	23.7	28.3	23.0	6.5	80.0
	15 m	27.6	22.0	26.1	18.4	6.5	71.2
	20 m	32.7	24.2	27.7	23.2	6.0	78.8
	100 m	23.2	17.4	19.3	18.1	5.5	60.7
Ni	Station	14.2	13.6	14.3	4.0	9.5	21.3
	5 m	13.4	12.9	12.4	4.1	8.8	22.0
	10 m	12.4	11.9	12.1	3.5	8.0	18.0
	15 m	12.1	11.8	11.5	3.0	7.9	17.2
	20 m	10.8	10.5	10.9	2.8	6.5	15.4
	100 m	9.1	8.9	9.3	2.2	6.0	12.5
Co	Station	4.6	4.4	4.5	1.4	3.0	6.6
	5 m	4.6	4.5	4.7	1.2	3.2	7.0
	10 m	4.3	4.2	4.2	0.7	3.4	5.2
	15 m	4.2	4.1	4.1	0.9	2.9	5.5
	20 m	4.0	4.0	3.8	0.84	3.1	5.5
	100 m	3.8	3.8	3.6	0.8	3.0	5.4

A* – arithmetical mean, G* – geometrical mean

There are many papers focused on heavy metal pollution of the soil environment in urbanized areas of Poland [CZARNOWSKA 1995, CZARNOWSKA, GWOREK 1991, CZERWIŃSKI, PRACZ 1990, CURZYDŁO 1988, MACIEJEWSKA, SKŁODOWSKI 1995]. On the other hand, relatively few studies are devoted to areas that are subject to the influence of traffic, particularly near gas stations. The available data focus on soil pollution by PAH and mineral oils [KONDRAS 2003; KLUSKA, KROSZCZYŃSKI 2000; BARAN, OLESZCZUK 2001; MIKOŁAJEK et al. 1985; ZERBE et al. 1995], what hampers the evaluation of the obtained results.

Unfortunately, there are no procedures of acting with regard to high pollution of soils in Poland [PALIWA PŁYNNE 2002]. Such procedures have been established in other countries. The actions of the Ministry of the Environment of Netherlands are a good example; since 1999 all gas stations are obliged to:

- test the degree of soil pollution and, when pollution is significant,
- design and fulfill a land reclamation project, as well as modernize the gas station [FARLISZEWSKA 1998].

In the literature it is often emphasized that the grain size composition is very influential with regard to the content of heavy metals and their forms available to plants in soils [DZIADEK, WACŁAWEK 2005, GĘBSKI 1998, KABATA-PENDIAS, PENDIAS 1999, LIPIŃSKI 2000, ŁABĘTOWICZ, RUTKOWSKA 2001].

In this study almost all heavy metals with the exception of nickel and cobalt showed a significantly positive correlation (α=0.01) with the content of organic matter (PR% and MO%). Considering soil pH values, significant correlation occurred with Pb, Zn, Cu and Ni, while in the case of Co the correlation was not significant (Table 5).

Table 5

Linear correlation coefficients between the concentration of trace elements and seasonal soil properties (during winter and summer)

	Winter					Summer				
	<0.02	<0.002	PH	PR%	OM%	<0.02	<0.002	pH	PR%	OM%
Pb	-0.14	-0.03	0.36^{**}	0.31^{**}	0.31^{**}	-0.10	-0.03	0.44^{**}	0.33^{**}	0.33^{**}
Cd	-0.12	-0.24^{*}	0.19	0.31^{**}	0.36^{**}	-0.12	-0.15	0.32^{**}	0.27^{*}	0.31^{**}
Zn	-0.23	-0.36	0.37^{**}	0.48^{**}	0.49^{**}	-0.18	-0.31	0.49^{**}	0.44^{**}	0.41^{**}
Cr	0.01	0.07	0.27^{*}	0.43^{**}	0.46^{**}	0.05	0.07	0.38^{**}	0.44^{**}	0.43^{**}
Cu	-0.16	-0.25	0.37^{**}	0.29^{*}	0.35^{**}	-0.13	-0.22	0.38^{**}	0.30^{**}	0.31^{**}
Ni	-0.06	0.07	0.40^{**}	0.28^{*}	0.27^{*}	-0.07	0.001	0.49^{**}	0.27^{*}	0.28^{*}
Co	0.19	0.37^{**}	0.16	0.27^{*}	0.31^{**}	0.29^{*}	0.41^{**}	0.18	0.20	0.20

Alfa	0.05	0.01
Critical value r for n=69	0.23	0.29
Signifcance level for compartments of the trust 10		

**= significant at level 0.01; * = significant at level 0.05; 0 = not significant

No positively significant correlations were noted between the concentration of heavy metals and the washable and colloidal particles; in some cases the correlation was even negative.

Similar results have been reported in many papers. PIOTROWSKA and TERELAK [1997], STRĄCZYŃSKA and STRĄCZYŃSKI [2000], as well as WŁAŚNIEWSKI [2000] have not confirmed the dependence between the concentration of cadmium and the grain size composition of soils, whereas TERELAK and PIETRUCH [2000] have noted only weak relationship between the content of total cadmium in soils and the concentration of the washable fractions. KALEMBASA et al. [2005] found a significantly positive correlation between the concentration of Cr, Ni and Cu on the one hand, and the colloidal clay in the humus layer, on the other. The sometimes mutually incoherent results as obtained by different researchers concerning the relationship between the washable fraction content and the amount of heavy metals could have been the consequence of the application of different research techniques for the extraction of elements from the soil. It seems that the washable fraction in soil may be more influential for the output amount of plant-available forms of heavy metals than concerning the total content of metals in soils and thus it may modify their uptake and accumulation in the plants [SADY, SMOLEŃ 2004]. The studied soils are characterized by rather even structure of grain size composition; most of them were classified as light soils. This, in turn, modifies the obtained results of the heavy metal concentration in the soils as well as the conclusions of statistical analysis.

It has to be emphasized, however, that the concentration of heavy metals in the studied soils is under the influence of a number of other factors like, e.g. distance from the gas station, the size of the station and the intensity of traffic in the region of study.

CONCLUSIONS

1. The degree of soil pollution with heavy metals in the vicinity of the studied gas stations is variable, even though in most of the cases the concentrations of heavy metals fell within the limits determined for natural soils. Close to stations with the heaviest traffic, the concentration of lead exceeded the standard (accepted highest) level.

2. The total content of heavy metals shows a weak decreasing tendency with distance from the gas station.

3. Heavy metals, which are poorly mobile in neutral and alkaline soils, have concentrated in a thin sub-surface layer of the soil.

4. The concentration of heavy metals in soils adjacent to gas stations is influenced, to a certain degree, by the physical-chemical properties of the soil

and, moreover, by the actual concentration of organic matter and the pH value. No significant dependence was found between the concentration of heavy metals in the soil and the amount of washable particles.

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