Journal of Ecological Engineering
Volume 15, No. 1, Jan. 2014, pp. 74–80
DOI: 10.12911/22998993.1084215

Research Article

CONTENT OF ZINC, LEAD AND CADMIUM IN SELECTED AGRICULTURAL SOILS IN THE AREA OF THE ŚLĄSKIE AND CIĘŻKOWICKIE FOOTHILLS

Agnieszka Józefowska[1], Anna Miechówka[2], Michał Gąsiorek[3], Paweł Zadrożny[4]

[1] Department of Soil Science and Soil Protection, University of Agriculture in Krakow, 21 Mickiewicza Ave., 31-120 Kraków, Poland, e-mail: agnieszka.jozefowska@ur.krakow.pl

[2] Department of Soil Science and Soil Protection, University of Agriculture in Krakow, 21 Mickiewicza Ave., 31-120 Kraków, Poland, e-mail: rrmiecho@cyf-kr.edu.pl

[3] Department of Soil Science and Soil Protection, University of Agriculture in Krakow, 21 Mickiewicza Ave., 31-120 Kraków, Poland, e-mail: rrgasior@cyf-kr.edu.pl

[4] Department of Soil Science and Soil Protection, University of Agriculture in Krakow, 21 Mickiewicza Ave., 31-120 Kraków, Poland, e-mail: rrzadroz@cyf-kr.edu.pl

Received: 2013.11.29
Accepted: 2013.12.17
Published: 2014.01.15

ABSTRACT

The purpose of this study was to evaluate the state of contamination with zinc, lead, and cadmium in selected soils of the Śląskie and Ciężkowickie Foothills and to determine the impact of the type of agricultural use and selected physico-chemical properties of soils on heavy metal content. The test soils were characterized by natural content of zinc, lead, and cadmium in most cases. Only one type of soil located on Śląskie Foothills developed increased levels of Cd ($1.1\ mg \cdot kg^{-1}$). The content of zinc, lead, and cadmium in the surface layer (0–30 cm) was higher in the soils of Śląskie Foothills than in soils of Ciężkowickie Foothills. The bedrocks from which the soils of these two mesoregions are formed differed significantly only in the content of zinc (it was higher in the soils of Śląskie Foothills). The content of Zn, Pb, and Cd in the surface layer of soil depends on its texture and organic carbon and total nitrogen content. There was also a positive correlation between the content of Pb and Cd and hydrolytic acidity and between the content of Zn and Ca and CEC. Different types of land uses did not influence the content of the metals.

Keywords: Śląskie and Ciężkowickie Foothills, heavy metal, land use.

INTRODUCTION

Carpathian soils are relatively well documented for their content of heavy metals [1, 15, 21]. The most well known soils in this regard are those of national parks [19, 22, 31] and the least recognised are the foothill soils used for agricultural purposes [4, 30]. Among the latter ones are the Ciężkowickie and Śląskie foothills soils. Śląskie Foothills is part of West Beskid Foothills [12], in a short distance from Upper Silesian Industrial Region (Polish) and Ostrava-Karwiński Industrial District (Czech Republic). These foothills are characterized by high rainfall, *via* which dust pollution in the atmosphere can get into the soil. Average annual rainfall for the meteorological station in Wisła Centrum (Śląski Beskid) in 2000-2010 was 1286.7 mm (data from the Institute of Meteorology and Water Management). Climatic conditions in this area favour the accumulation of organic matter in soils [16]. Organic matter stops the metals through the creation of specific connections with them [3]. Therefore, in the soils of this mesoregion a higher content of heavy metals can be expected than in the soils of the Ciężkowickie Foothills, located in the Central Beskid foothills at a distance from industrial centres. Soils of the Ciężkowickie Foothills are characterized by lower organic carbon stocks [18] and the occurrence of lower average annual rainfall (801.7 mm for Biecz-Grudna post in 2000–2010) than those of the Śląskie Foothills.

The purpose of this paper was to evaluate the state of contamination with zinc, lead and cadmium in brown soils and lessive soil, which are characteristic for arable land and fresh grasslands occurring in the Śląskie and Ciężkowickie foothills. The next aim was to determine the effect of the different types of land use soils on heavy metal content and to attempt to identify the relationships between the selected physico-chemical properties and concentrations of zinc, lead and cadmium in the soils.

MATERIALS AND METHODS

The study was conducted using soil material collected from 16 profiles located in the Śląskie (in the villages of Ustroń, Górki Wielkie, and Porąbka) and Ciężkowickie (Swoszowa, Joniny, Czermna, and Dobrocin) foothills. In each location, soil pits were made in the neighbouring arable lands (A) and grasslands (G). The exception was the village of Ustroń, where four profiles located in two different positions were examined. The tested soils are representative of the habitat of fresh agricultural use. They are classified into brown soils and lessive soils [27], and most were formed from weathering or landslide material of the Silesian unit of the Carpathian Flysch. The location of these soils and their detailed characteristics are given in previous papers [12, 17].

For the purposes of this study, soil was sampled from layers of 0–30 cm depth (topsoil) and the bedrock horizons. These samples were dried and sieved (2 mm diameter). In the fine earth parts of the soils the following properties were determined:

- texture, using Prószynski's modification of the aerometric Casagrande method [24],
- pH in 1 mol · dm^{-3} KCl solution, using the potentiometric method [5],
- the total exchangeable base (TEB) cation contents, by means of determining individual cations (Ca^{2+}, Mg^{2+}, K^+ and Na^+) after their extraction with 1 mol · dm^3 CH_3COOHN_4 solution [13]; in extracts Ca^{+2}, K^+ and Na^+ were determined by flame photometry, and Mg^{2+} by atomic absorption spectrophotometry,
- hydrolytic acidity (Hh) with the Kappen method using 1 mol · dm^{-3} CH_3COONa solution [13],
- the content of organic carbon (OC) using the Oleksynowa modification of the Tiurin method [23],
- the content of total nitrogen (TN) with the Kjeldahl method [13] using Kjeltec apparatus,
- the total content of zinc, lead, cadmium and calcium after digestion of soil samples in a mixture of nitric and perchloric acids, by atomic emission spectrometry with inductively coupled plasma (ICP-OES).

Cation exchange capacity (CEC) and base saturation (BS) percentage were then calculated as follows:

$$CEC = TEB + Hh;$$

$$BS\ [\%] = \frac{TEB \cdot 100}{CEC}$$

The assessment of heavy metal contamination in soil was carried out by comparing the results obtained with the limit for heavy metals contents specified in the Regulation of the Minister of the Environment of 9 September 2002 regarding the quality standards for soil and ground [25] and based on the limit for the content of heavy metals recommended by the Institute of Soil Science and Plant Cultivation (IUNG) [8]. To assess the degree of heavy metals accumulation in soil, two indicators were used: the accumulation index (AI) [6] and the enrichment factor (EF) [14]. To calculate EF, calcium (Ca) was used as a reference element [14].

The statistical analyses were carried out using the program Statistica 10PL. The two-way analysis of variance (two-way ANOVA) and coefficients of linear correlation were tested and standard deviations and the average values were calculated.

RESULTS AND DISCUSSION

The analysed soils were characterized by diverse physicochemical, physical and chemical properties (Table 1). According to "Particle Size Distribution and Textural Classes of Soils and Mineral Materials" [10], the soils investigated were silty loam, clay loam, loam, and sandy loam.

The soils from sites located in the Ciężkowice Foothills were characterized by lighter texture than soil from samples collected in the Śląskie Foothills. The topsoil from these two regions contained significantly different amounts of sand and silt fractions (Table 1). The content of the fraction < 0.02 mm allow, classified the investigated soil as medium-heavy and heavy. It was necessary to evaluate the contamination of soils on the basis

Table 1. Selected properties of the soil surface layer (0–30 cm)

Soil properties	Śląskie Foothills		Ciężkowickie Foothills		Region	The way of land use
	A[a]	G[b]	A[a]	G[b]	F[c] P[d]	F[c] P[d]
pH KCl	4.2-4.9[1] 4.4[2] (0.3)[3]	3.8-4.7 4.2 (0.3)	3.8-5.9 4.4 (0.8)	4.0-7.2 5.6 (1.5)	2.08 0.174	0.86 0.372
Hh [mmol(+) · kg^{-1}]	36.7-70.4 53.1 (11.9)	44.6-106.2 72.7 (21.9)	22.2-65.0 48.5 (15.9)	12.0-56.1 36.3 (16.1)	4.46 0.056	0.15 0.709
TEB [mmol(+) · kg^{-1}]	41.8-98.3 73.3 (20.2)	11.6-111.3 64.2 (36.7)	13.9-102.5 57.5 (38.7)	19.0-172.1 90.4 (54.7)	0.05 0.824	0.27 0.612
CEC [mmol(+) · kg^{-1}]	112.2-135.0 126.4 (8.6)	117.8-155.8 137.0 (16.7)	76.4-144.0 106.0 (29.2)	62.5-184.1 126.7 (45.3)	0.87 0.369	0.90 0.362
BS [%]	38.3-72.8 57.3 (12.3)	9.6-71.8 44.5 (22.6)	17.9-82.3 48.7 (25.0)	26.4-92.8 67.1 (26.1)	0.30 0.596	0.05 0.832
Ø 2-0.05 mm	17.1-27.7 24.2 (4.2)	17.4-26.5 23.9 (3.8)	24.7-58.2 34.4 (13.8)	31.2-75.6 52.6 (18.2)	8.16 0.014*	1.73 0.213
Ø 0,05-0,002 mm	55.7-69.1 61.6 (5.3)	58.7-73.2 63.7 (5.7)	34.3-64.0 50.3 (11.1)	29.0-54.4 45.0 (9.7)	9.16 0.011*	0.05 0.831
Ø <0,002 mm	12.3-18.9 15.2 (2.5)	9.4-15.0 12.3 (2.1)	7.5-25.0 15.3 (6.6)	6.3-19.0 10.7 (5.1)	0.08 0.782	2.09 0.174
Ø <0.02 mm	47.3-52.0 49.7 (1.7)	42.2-50.0 46.7 (3.1)	28.2-55.0 42.1 (9.5)	22.3-49.8 35.6 (9.8)	5.19 0.042*	1.36 0.266
OC [g · kg^{-1}]	9.6-14.6 12.4 (1.8)	16.2-20.0 17.5 (1.5)	3.1-9.5 6.9 (2.4)	10.3-14.2 12.0 (1.6)	26.82 0.000*	22.86 0.000*
TN [g · kg^{-1}]	0.9-1.7 1.4 (0.3)	1.6-2.3 1.9 (0.3)	0.8-1.2 1.0 (0.2)	0.9-1.8 1.4 (0.3)	10.70 0.007*	9.32 0.010*
Ca [mg · kg^{-1}]	1325.4-2107.5 1602.2 (315.1)	806.9-2464.8 1506.1 (603.4)	38.7-544.0 253.7 (191.4)	57.6-2737.3 911.7 (1065.7)	6.92 0.022*	0.58 0.461

Explanation: [1] the range; [2] mean; [3] standard deviation; [a] arable land; [b] grassland; [c] value of two-way ANOVA F-test; [d] probability p; *indicated value significant at p <0.05.

of the limit specified in Regulation [25] and Institute of Soil Science and Plant Cultivation (IUNG) recommendations [8]. The content of organic carbon and total nitrogen in the investigated soils differed both between the regions and depending on the type of use (Table 1). Śląskie Foothills soils were characterized by higher total Ca content than Ciężkowickie Foothills soils. This was mostly conditioned by the content of this element in the bedrock.

The zinc content in the Śląskie Foothills soils was higher than in the soils of the Ciężkowickie Foothills. A similar relation was also observed in the concentration of this element in the bedrock from these two regions (Table 2). In the topsoil at all sites in this mesoregion, zinc content (86.3 mg · kg^{-1}) was higher than the average content of this component in the soils of the Earth, which is 64 mg · kg^{-1} [26] and depends on the natural content of this element in the bedrock (Table 3). Several authors [2, 20, 29] drew attention to the relation between the content of Zn in soils and bedrock. Based on the calculated accumulation index (AI) it is difficult to determine from which region soils have a higher amount of Zn of anthropogenic origin. Average values of WA for soils of the two regions were similar (1.7 for Śląskie Foothills and 1.9 for Ciężkowickie Foothills) and were higher than the value of this ratio given for the soil of the Earth, which is 1.075 [26]. Based on the average values of the zinc enrichment factor (EF_{Zn}), it can be concluded that the accumulated Zn in the Śląskie Foothills soils is of anthropogenic origin (Table 2). Mean values of EF_{Zn} for soils from those regions were higher than 1.5, which, according to Zhang and Liu [32], indicates anthropogenic accumulation of the metal.

Despite the zinc enrichment of the soil surface layers, its content does not exceed the allowable concentrations [25] and the maximum Zn content found in the soils (94.5 mg · kg^{-1}) indicates that they can be classified as soil with a natural content of this element (zero degrees of contamination) [8].

As well as the zinc content, the lead content in the Śląskie Foothills soils was higher than in the Ciężkowickie Foothills soils. However, such relationship has not been reported for the concentration of this element in the bedrock of these two regions (Table 2). In the topsoil of Śląskie Foothills the lead content was higher (31.5 mg · kg^{-1})

Table 2. The content of zinc, lead, and cadmium in the surface layer (0–30 cm) soils and bedrock, accumulation index (AI), and enrichment factor (EF)

Metal	Śląskie Foothills		Ciężkowickie Foothills		Region[a]	The way of land use[a]
	A[a]	G[b]	A[a]	G[b]	F[c] P[d]	F[c] P[d]
0-30 cm [mg · kg^{-1}]						
Zn	75.5-94.5[1] 83.4[2] (7.7)[3]	73.4-113.7 89.1 (15.4)	33-7-67.1 48.0 (12.1)	25.4-57.9 44.8 (12.7)	31.57 0.000*	0.032 0.862
Pb	25.4-32.9 28.3 (2.8)	29.6-48.4 34.7 (7.9)	10.3-20.7 14.5 (3.8)	8.6-20.3 14.6 (4.5)	32.78 0.000*	1.20 0.295
Cd	0.2-1.1 0.5 (0.4)	0.2-0.7 0.4 (0.2)	0.0-0.1 0.0 (0.0)	0.0-0.2 0.1 (0.1)	10.54 0.007*	0.03 0.874
in the bedrock [mg · kg^{-1}]						
Zn	41.9-79.6 60.5 (17.0)	37.7-59.0 48.6 (8.5)	12.2-59.6 43.0 (18.9)	10.3-53.0 25.1 (16.5)	5.07 0.044*	2.69 0.127
Pb	6.6-14.6 10.5 (3.2)	7.7-10.2 8.7 (1.1)	4.0-20.4 11.4 (6.1)	3.1-13.5 6.0 (4.3)	0.14 0.713	2.30 0.155
Cd	0.0-0.3 0.1 (0.1)	0.0-0.2 0.1 (0.1)	0.0-0.0 0.0 (0.0)	0.0-0.0 0.0 (0.0)	1.96 0.187	0.04 0.844
AI						
Zn	1.2-1.8 1.5 (0.3)	1.5-2.1 1.9 (0.3)	0.8-2.8 1.4 (0.8)	1.1-4.0 2.3 (1.1)	0.30 0.593	2.43 0.145
Pb	2.2-3.8 2.9 (0.7)	3.2-4.7 3.9 (0.5)	1.0-3.3 1.6 (0.9)	1.5-4.1 3.0 (1.1)	5.08 0.044*	6.35 0.027*
Cd	3.5-381.0 225.7 (138.6)	2.9-380.0 237.5 (151.1)	1.0-97.0 45.0 (44.4)	11.7-240.8 99.5 (90.9)	5.83 0.033*	0.25 0.625
EF						
Zn	0.8-12.6 4.0 (5.0)	1.1-2.8 1.9 (0.6)	0.4-1.8 1.1 (0.6)	0.1-1.0 0.5 (0.4)	2.13 0.170	0.91 0.358
Pb	10.7-14.9 12.2 (1.7)	9.6-14.9 12.5 (2.1)	4.6-6.8 5.9 (0.9)	6.2-9.4 7.9 (1.3)	36.93 0.000*	1.64 0.224
Cd	3.4-120.8 53.5 (43.3)	2.9-59.4 28.7 (20.7)	0.3-30.0 10.8 (12.2)	2.0-27.6 15.1 (12.5)	3.65 0.080	0.49 0.498

Explanation: see Table 1.

than the average content in the soil of the Earth, which is 22.6 mg · kg^{-1} [26].

The content of this component in the upper layer of soil was not related to the amount of this element in the bedrock (Table 3), which could indicate an anthropogenic origin. The calculated value of the accumulation index (AI) also confirms the anthropogenic origin of this element. Mean values of AI_{Pb} for both regions are higher (in soil of Śląskie Foothills 3.4 and Ciężkowickie Foothills 2.3) than the value of this ratio specified for soil of the Earth, which is 1.364 [26]. It is also worth noting that the accumulation index of lead determined for grassland soils was higher than for arable land soils, regardless of the region (Table 2).

The unnatural lead enrichment in foothills soil is also reflected by the high values of the enrichment factor (EF_{Pb}), which are usually in the range from 5 to 20. Despite the significant lead enrichment of the soil surface layers, the content of these elements, like zinc, does not exceed the permissible levels of this metal specified in the Regulation of the Minister of the Environment on the quality standards for soil and ground [25]. Even the highest content of Pb, 48.4 mg · kg^{-1}, determined in soil collected from grassland in the village of Porąbka, does not exceed the maximum allowable level for the uncontaminated Pb soils [8]. At the same time, all tested soils contain more than 25 mg · kg^{-1} of lead, which is the amount that suggests a human impact on soil contamination by this element according to Kabata-Pendias and Pendias [7].

In both the topsoil and the bedrock, the cadmium concentration in Śląskie Foothills soils was higher than in Ciężkowickie Foothills soils (Table 2). The average content of Cd in the topsoil from this mesoregion was higher (0.5 mg · kg^{-1}) than the average content in the surface layer of soil on Earth, which was 0.145 mg · kg^{-1} [26], whereas in an analogous layer of soil from the Ciężkowickie Foothills the Cd content was significantly lower (0.1 mg · kg^{-1}). The high accumulation index of

Table 3. Linear correlation coefficients defining the relationship between the heavy metals in the topsoil contents and their contents in the bedrock

Layer		in the bedrock		
		Zn	Pb	Cd
0-30 cm	Zn	0.699*	0.558*	0.216
	Pb	0.414	0.319	-0.088
	Cd	0.145	0.166	0.882**

Explanation: Coefficient significant at * $p < 0.05$; ** $p < 0.01$.

cadmium (AI_{Cd}) calculated for the investigated soils significantly exceeded the value specified for soils of the Earth, which is 1.477 [26]. High values of EF_{Cd} could indicate the anthropogenic origin of this metal (Table 2). The cadmium content in the surface layer of the investigated soils in both foothills does not exceed the limit values specified in the Regulation of the Minister of the Environment [25]. Taking into account the limit values given by Kabata-Pendias et al. [8], most of the investigated soils were characterized by a natural content of cadmium (0º of soil pollution). An increased content of this element (greater than 1 $mg \cdot kg^{-1}$) occurred only in the soil of arable land in the village of Porąbka (Iº of soil pollution).

The results confirmed the relationship between the concentration of heavy metals and texture, organic carbon content, and sorption properties of the soil already described by several authors [6, 11, 20]. The content of Zn and Pb in soils depended on their texture. This is confirmed by a statistically significant correlation coefficient between Zn and Pb content and the amount of the fraction lower than 0.02 mm (Table 4). Similar results were obtained by Kołodziej et al. [11], who found that the heavy metal content in clayey flysch soils is higher than in loamy flysch soils.

The relation between the content in the soil fraction below 0.02 mm and the concentration of zinc is not confirmed by studies by Kaniuczak et al. [9]. The Zn and Pb content in soils was significantly dependent on the content of organic carbon and total nitrogen. The accumulation of lead in the topsoil is connected with this metal's particularly strong capability to form interconnections with organic matter [7, 22, 28]. These dependencies are not shown in the studies of Kaniuczak et al. [9], who found no relation between the humus content and concentration of heavy metals in agricultural soils derived from the Carpathian flysch. Moreover, the total content of lead and cadmium is positively correlated with hydrolytic acidity, and the zinc content is strictly positively correlated with the content of calcium and the total volume capacity of the sorption complex (Table 4).

The soils from the Śląskie Foothills were characterized by a higher content of heavy metals than the soils from the Ciężkowickie Foothills. On the basis of these studies and available literature, it can be assumed that the Śląskie Foothills soils are more exposed to the possibility of contamination from the air. In the bedrock of the Śląskie Foothills there is a higher concentration

Table 4. The correlation coefficients between the heavy metals content in the topsoil and selected soil properties at $p < 0.05$

Soil properties	Zn	Pb	Cd
pH KCl	-0.227	-0.358	-0.230
Hh [$mmol(+) \cdot kg^{-1}$]	0.346	0.523*	0.510*
TEB [$mmol(+) \cdot kg^{-1}$]	0.192	0.037	-0.133
CEC [$mmol(+) \cdot kg^{-1}$]	0.500*	0.415	0.176
BS [%]	0.136	-0.065	-0.261
Ø <0.002 mm	0.422	0.230	-0.014
Ø <0.02 mm	0.767***	0.653**	0.277
OC [$g \cdot kg^{-1}$]	0.700**	0.809***	0.314
TN [$g \cdot kg^{-1}$]	0.756***	0.780***	0.207
Ca [$mg \cdot kg^{-1}$]	0.546*	0.437	0.405

Explanation: Coefficient significant at * $p < 0.05$; ** $p < 0.01$; *** $p< 0.001$.

of metal, and the properties such as the soil parts below 0.02 mm and humus content are conducive to the accumulation of zinc, lead, and cadmium.

CONCLUSION

1. The investigated soil of the Śląskie and Ciężkowickie Foothills, in most cases, is characterized by natural content of zinc and lead as well as cadmium.
2. The Śląskie Foothills soils had significantly higher accumulation index of Pb and Cd and enrichment factor of Pb than the Ciężkowickie Foothills soils.
3. In the analysed soils of the Śląskie Foothills, zinc, lead, and cadmium content in the 0–30 cm layer was higher than in the soils of the Ciężkowickie Foothills.
4. Different types of land use did not affect the content of heavy metals.
5. Only the accumulation index of lead was higher in grassland soils than in arable land soils.

Acknowledgements

The studies were financed by the Ministry of Science and Higher Education, Poland, under the grant No. N N310 312434.

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