INFRASTRUKTURA I EKOLOGIA TERENÓW WIEJSKICH
INFRASTRUCTURE AND ECOLOGY OF RURAL AREAS

Nr IV/1/2016, POLSKA AKADEMIA NAUK, Oddział w Krakowie, s. 1243–1256
Komisja Technicznej Infrastruktury Wsi

DOI: http://dx.medra.org/10.14597/infraeco.2016.4.1.091

ASSESSMENT OF THE CONTENT OF HEAVY METALS IN PLANTS AND SOIL IN THE AREA OF THE TRZEBINIA MUNICIPALITY, POLAND. 1.CADMIUM

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Abstract

The study aimed at finding a relationship between the content of cadmium in the soil material and in the above ground parts of plants taken from the areas of the municipality of Trzebinia. Research areas were designated in areas degraded by mining and processing industries, which are now fallows. The total content of cadmium in soil and plant samples as well as soil factors such as organic matter content, granulometric composition, pH and electrolytic conductivity were marked in laboratory analyzes. The concentration of the element in the soils of the municipality exceeded the national regulations for agricultural land (Journal of Laws 2002). Similarly, the metal content in plants is higher than the acceptable standards for animal feed as stated in Regulation of the Minister of Agriculture and Rural Development (2012a). The results indicate a positive relationship between the content of cadmium in the soil at the level of 0-20 cm and its concentration in the above ground parts of plants of fallow lands of the Trzebinia municipality.

Keywords: heavy metals, cadmium, soil, plant, fallow land, pollution

INTRODUCTION

Among chemical elements cadmium contamination is grave and poses a serious threat to the environment and to human health (Sarkar, 2002, Bąbelewska 2012). This element is easily taken in by plants particularly in the cationic form (Cd^{2+}) and accumulated in all plant tissues. The process of absorption is per-

formed by aboveground parts of plants and roots; however, the micro-organisms of the rhizosphere create a protective barrier, trapping the metal in cells. Acidic soils, contaminated and fertilized with phosphorus fertilizers or with sewage sludge and manure (Kabata-Pendias 2000, Gwóźdź and Kopyra 2003) create the most favourable conditions for the absorption of Cd. Wet and dry precipitation containing cadmium may be the rich source of the element which is supplied to the soil (Bąbelewska 2012). Plants assimilate cadmium occurring naturally in source rocks with difficulty. The element shows a high mobility in soil environment. It is relatively easily leached into groundwater. It reaches the concentration of 300μg/dm^3 in soil solutions. Soils in the areas of mines and zinc and lead metallurgical plants contain the largest amount of the element (Staszewski *et al*., 2008). There is a close and direct causality between the presence of cadmium in soils and in plants. Its natural content in soil is determined by its mechanical composition, type of soil, climate and the content of the element in the bedrock. The amount of cadmium in uncontaminated soils (especially clay and organic ones) fluctuates in the range of 0.01 and 2.5mg/kg, including arable soils of Poland. Terelak *et al*. (2000) denotes the range of 0.01 and 50.0 mg/kg. National regulations (Journal of Laws 2002) set the following limits on the content of the element in (mg/kg) for protected areas: 1, for industrial sites: <15, for agricultural land: <5. In general, the balance of Cd in soil reaches a positive value between 1-3 g/ha/year. Fuel combustion, metallurgy of iron and steel, municipal pollution are the main source of Cd in the air. It is used for the production of plastics, dyes, glass, accumulators and anticorrosive protection (Kabata-Pendias and Mukherjee 2007, Kabata-Pendias 2010).

FIELD AND LABORATORY METHODS

The study was conducted in the municipality of Trzebinia. Field work was carried out in each case in June and July in the years of 2010-2012. These works included the delineation of research plots, and collection of plant and soil material. Field studies were conducted on the sample plots. Each surface had a shape of square with a side length of 5 m (25 m^2). Geographical coordinates using a GPS (Garmin GPSMAP 62s) and basic parameters of the habitat such as altitude, slope and orientation towards the cardinal directions were identified for each area. The locations of research areas were chosen so as they reflected the diversity of soils and vegetation in the municipality of Trzebinia. The designated areas used to be agricultural lands in the 80s whereas now they are fallow lands. This was done by analysing topographic maps at a scale of 1:10000. Additionally, on the basis of sozological maps, the distribution of deposits and areas of former mining and processing of minerals as well as potentially existing pollution sources were taken into account. In total, 83 areas were selected. The differentiation in number of

research areas selected in each of the rural administrative units resulted from the uneven distribution of both fallow lands and historical as well as current mining sites of mineral deposits and emitters of pollutants. Most areas were designated in Trzebinia (23), the fewest in the rural administrative unit of Dulowa (1). Two samples were collected from each point: one at the depth of 0-20 cm and one at the depth of 20-40 cm. Four samples of plant material were collectively gathered from each research area. A total of 332 plant samples and of 664 soil material samples were collected. The analysis of soil material involved the determination of granulometric composition of the soil with the use of the Casagrande method modified by Prószyński (according to industry standard BN-78/9180-11), soil reaction in 1M KCl and in water with the use of the potentiometric method, electrolytic conductivity of the soil with the use of the conductometric method, content of organic matter in soil by its annealing using the modified Tiurin method, the total Cd content in the soil using the FAA method. The analysis of plant material was based on the determination of the total content of Cd in plants using the atomic absorption spectrometry. The results obtained concerning the content of Cd heavy metals in plant and soil material were verified by reference standards and were statistically analysed. Basic descriptive statistics were calculated i.e. minimum value, maximum value, the arithmetic mean, the standard deviation, the median to determine the quantitative relationship between the contents of respective elements in the soil (separately from the level of 0-20 cm and 20-40 cm) and the content of these elements in the plant, between the value of pH (separately in H_2O and KCl) of soil and the content of these metals in the soil (0-20 and 20-40 cm), between the metal contents in the soil (0-20 cm and 20-40 cm) and the content of organic matter in the soil, between the content of the metals in the soil (0-20 cm) and at the level of 20-40 cm. The content of the element turned out not to have a normal distribution which was examined by Shapiro-Wilk test. For this reason the statistical analysis was performed using Spearman correlation coefficient determining the degree of dependence among variables.

CHARACTERISTICS OF THE RESEARCH AREA

The town and the municipality area of Trzebinia is located on the border of the Olkuska Upland, Pagóry Jaworznickie and the Krzeszowice Trench in the western part of the province of Malopolska, in the Chrzanowski County (Figure 1). In the north, it is adjacent to the communities of Bukowno and Olkusz, in the east it borders the Krzeszowice community, in the south the Alwernia community and in the west it is adjacent to the municipalities of Chrzanów and Jaworzno (Environment Programme 2013).

Basic minerals which have transregional significance such as coal, dolomite and sand backfilling and common minerals such as loam and limestone,

mostly consumed by the local economy were identified in the area of the municipality (Pietraszek 1961, Szuwarzyński 2009). The development of metallurgy of zinc and glass was observed in the first years of the nineteenth century. The first galvanizing plant was launched in Siersza in 1811. A private alum brewhouse operated occasionally there (Pietraszek 1961, Pęckowski 2013).

Figure 1. Location of Trzebinia in the map of Poland

Leached brown soils and pseudo podzolic soils created from pieces of loess of aeolian origin are typical soils of the area of the municipality. They constitute a large area estimated at about 525 hectares in the eastern part of the municipality including the rural administrative units of Psary, Karniowice, Młoszowa and Dulowa. Loess soils of loose sand compactness occur in the area of the rural administrative unit of Karniowice. Eroded leached brown soils with periodic moisture deficit were identified in the loess areas with steeper slopes. These soils are classified as complex III and are characteristic of Karniowice, Psary and Młoszowa. In the loess area brown soils of unquestionably deluvial origin created as a result of slopes being leached by rainwater can be found. They form the bottom structure of the gutter and narrow depressions degraded by water gorges (ravines) (Kiryk 1994, Szuwarzyński and Kryza 1995, Environment Programme 2013, Kot – Niewiadomska 2013, Pęckowski 2013).

The most noteworthy energy and industrial emitters of pollutants into the atmosphere of Trzebinia are the following companies: the Trzebinia Refinery Capital Group, „Szczakowa" PCC Rail JSC, the „Siersza" Power Plant in Trzebinia, the metallurgical slag heap of the „Trzebinia" Metallurgical Plant and the stockpile of flotation tailings of the „Trzebionka" Mining plant (Kot 2011; Environment Programme 2013).

FINDINGS AND DISCUSSION

The mean content of cadmium in the plants and in soil was 1.09mg/kg. The range of metal content in plants fell into the brackets of 0.15-10.18 mg/kg. The mean content of cadmium in the soil at the depth of 0-20 cm was equal to 6.2mg/kg. At the depth of 20-40 cm the mean cadmium content was 5.16 mg/kg (Table 1).

Table 1. Basic statistics of the cadmium content in the soil and plants and of selected parameters of soil samples from the municipality of Trzebinia

Parameter		Mean	SD	Min	Median	Max
Organic matter [%]	0-20 cm	3.82	1.44	1.33	3.75	8.1
	20-40 cm	2.7	1.36	0.61	2.35	6.99
Heavy metals in plants [mg/kg]	Cd	1.09	1.31	0.15	0.76	10.18
Heavy metals in soil 0-20 cm [mg/kg]	Cd	6.2	6.44	0.9	3.9	43.5
Heavy metals in soil 20-40 cm [mg/kg]	Cd	5.16	7.55	0.25	3.1	54.37
pH 0-20 cm	in water	6.31	0.48	5.2	6.4	7.4
	in KCl	5.92	0.67	4.4	6	7
pH 20-40 cm	in water	6.7	0.52	5.2	6.8	7.7
	in KCl	6.19	0.68	4.4	6.4	7.4
The electrolytic conductivity 0-20 cm [µS]		175.46	97.89	32	165	464
The electrolytic conductivity 20-40 cm [µS]		155.71	100.25	31	137.7	540
Share of fraction of [%]	sand	68.36	19.41	19	72	95
	particles	22.14	16.63	3	16	74
	silt	9.49	6.45	0	8	37

The mean percentage of organic matter in the soil for the depth of 0-20 cm was 3.82% and the median amounted to 3.75%. The matter content at this depth was in the range of 1.33-8.1%. For the depth of 20-40 cm the mean content of organic matter in the soil was equal to 2.7% and the median was 2.35%. In this layer of soil the matter content ranged from 0.61 to 6.99% (Table 1). The mean electrolytic conductivity of the soil at the depth of 0-20 cm was equal to 175.46 µS and the median amounted to 165 µS. The conductivity in the layer of soil varied from 32.00 to 464 µS. For the depth of 20-40 cm the mean electro-

lytic conductivity was lower and amounted to 155.71 μS, and the median came to 137.7 μS. The range of values of electrolytic conductivity of the soil in the analyzed layer was in the range of 31-540 μS (Table 1). The mean pH of the soil, assuming acidic reaction, determined in water for the 0-20 cm layer added up to 6.31 and the median was 6.4. The pH values ranged from 5.2 to 7.4, that is, from acidic reaction to slightly alkaline one. For the depth of 20-40 cm the mean pH value determined in water came to 6.7 and the median was 6.8 which corresponded to the acidic reaction, similarly to the reaction of the soil in the 0-20 cm layer. The pH values for the depth of 20-40 cm ranged from 5.2 to 7.7 that is from acidic to alkaline reaction similarly to the 0-20 cm layer of the soil. The mean pH of the soil determined in KCl for the 0-20 cm layer corresponded to 5.92 and the median was 6.0 consequently showing acid reaction. The range of values of pH fluctuated between 4.4 and 7.0 oscillating between acidic and neutral reaction. For the depth of 20-40 cm the mean pH value determined in KCl amounted to 6.19, and the median was 6.4. Thus, it was acidic reaction such as in the 0-20 cm layer. At the depth of 20-40 cm the pH range of the soil fluctuated between 4.4 and 7.4 falling into the range from acidic to slightly alkaline reaction. That was in contrast with the 0-20 cm layer wherein the pH ranged from acidic to neutral reaction (Table 1). In terms of granulometric composition, the soils in the municipality of Trzebinia are dominated with clay, sand and silt soils. The biggest share of the samples was defined as sandy loam (15.66%). A significant part of the remaining samples was classified as loamy sand and boyer loamy sand (12.05%), fuquay loamy sand (10.84%). One sample of each type that is ordinary dust, clay dust, dusty sand loam, medium clay and heavy clay (1.20%) was classified. The mean percentage of the sand fraction amounted to 68.36%, of the dust fraction came to 22.14% and of the silt fraction added up to 9.49% (Table 1).

The average content of cadmium in the soil for the area in question exceeded (<5 mg / kg dry weight) for agricultural land according to national standards (Journal of Laws 2002). Cadmium content in soils as indicated by the Dutch List (2011) is as follows: for heavily contaminated soils – 20 mg/kg, for moderately contaminated soils – 5 mg/kg, for uncontaminated soils – 1 mg/kg. According to this scale the soil of the municipality should be classified as moderately contaminated. Studies conducted as part of Monitoring of Chemistry of Arable Soils in Poland (2012) revealed a very high content of cadmium in soils of Piekary Śląskie (57.5 mg/kg dry weight), which were under a long-term influence of the steel industry of zinc and lead. In turn, the range of the element in the soils in the vicinity of Metallurgial Plant in Katowice fell into the scope of 4.6-64 mg/kg dry weight which was in line with the results obtained by Abramowska (2006). The heap soil of Bolesławiec near Olkusz contained the extreme concentration of the metal within the range of 170-200 mg/kg (Abramowska 2006). In soils contaminated by mining and other industries cadmium content can be as high as

1780 mg/kg dry weight. In the agricultural soils of the Czernichów county in the vicinity of Krakow the mean cadmium content came to 1.13 mg/kg dry weight (Terelak *et al*. 2000). Kabata-Pendias and Pendias (1999) note that the highest concentration of cadmium in soils appears mainly in the vicinity of metallurgical plants, mines of lead, zinc and copper ores. It is then associated with the production of non-ferrous metals and coal combustion. According to Trzebiatowski (1971) cadmium is mostly present in copper, zinc and lead sulphide ores. The authors' results confirm the relationship between the former and present mining and processing of local raw materials activities and cadmium contamination of the soils of the municipality of Trzebinia and its individual rural administrative units where toxic cadmium content was found in soils. The highest concentration of cadmium in the plant material was found in Psary and Płoki which soils are contaminated with this metal. Vegetation of Myślachowice and Karniowice, where nowadays historical emitters of pollution do not have an impact on cadmium concentration in soils and there are no currently active emitters, contained the lowest content of the element. Kabata-Pendias and Pendias (1999) report that natural cadmium content in plants can fluctuate from <0.1 to 1 mg/kg dry weight, and the critical content in the range of 5-10 mg/kg dry weight. Curzydło (1995) revealed toxic symptoms of cadmium concentration in plants at the content of 5-30 mg/kg dry weight. In the analyzed plant samples of all rural administrative units the mean cadmium concentration equalled 1.09 mg/kg dry weight. Comparing the obtained cadmium contents with the literature data of Kabata-Pendias and Pendias (1999) it can be noted that the test plants contain the element in amount which does not exceed the critical content enabling their growth. According to the Regulation of the Minister of Agriculture and Rural Development (2012a) the maximum permissible concentration of cadmium in animal feed coming from vegetation (hay, silage, fresh grass) is lower or equals to ≤ 1 mg/kg dry weight. That means that vegetation from the research areas cannot be used for animal feed. In the area of the county of Lublin which is considered to be uncontaminated, the mean concentration of cadmium in grasses came to 0.1 mg/kg dry weight (Ward 1997) and it was almost 10 times lower than the concentration of cadmium in vegetation of fallow lands in Trzebinia. The mean cadmium content in grass vegetation in Roztocze, in Łomża and in the surroundings of the metallurgical plant in Dąbrowa Górnicza equalled successively: 0.037 mg/kg dry weight, 0.04 mg/kg dry weight and 0.3 mg/kg dry weight. (Biernacka and Wójcik 1998). In contrast, cadmium content in plants from the border belt of the eastern Poland ranged from 0.05 to 2.62 mg/kg dry weight (Mikos-Bielak and Tujaka 1999). In the area of Jeleniogórskie county cadmium content in plants ranged from 0.1-0.8 mg/kg dry weight, with the mean of 0.2 mg/kg dry weight (Kaszubkiewicz and Kawałko 2009). Although the results of cadmium content for the whole municipality and rural administrative units of Psary, Płoki and the town of Trzebinia indicate contamination of soil with cadmium, the concentra-

tion of the element in vegetation of the listed rural administrative units and the whole municipality of Trzebinia does not allow to conclude that it is contaminated with the element. Also, the proposed by Görlach and Gambuś (2000) range of permissible cadmium content in herbaceous plants (1-5 mg/kg dry weight) confirms the assertions of contamination with the element of vegetation in the municipality of Trzebinia. A positive correlation between the content of cadmium in the soil and the content of this metal in plants (Table 2) was revealed. Studies showed that plants in response to the contamination of soil with heavy metals can develop adaptive mechanisms aiming at achieving a particular level of tolerance by adjusting, for example, to excessive content of trace elements in the soil solution, as exemplified by metallophytes and hyperaccumulators (Baranowska-Morek 2003).

Table 2. The relationship between the cadmium content in the soil (0-20 cm) and in the plant

Heavy metal	Spearman correlation coefficient	p-value	Direction of the relationship	Power of the relationship
Cd	0.22	0.046	positive	weak

In his studies Brüggemann (1999) confirms the disproportionate increase in the content of heavy metals in plants in relation to their concentration in the soil solution. The metal content in plants is not only related to their content in the soil, but is determined by many factors. Depending on the place of research, the species composition of the vegetation and the type of heavy metal, some reports found a relationship between metal content in soil and its content in plants whereas some others did not find such a link. The research conducted by Wacławek and Maćko (2001) in the Kłodzka Valley, where contamination of soil with copper (mean content above 19 mg/ kg) was noted, showed no contamination of plant material with the metal. Similarly, in the works of Pratas *et al*. (2013), Szarek-Łukaszewska and Niklińska (2002) and Yoon *et al*. (2006) no contamination of plants with Zn, Pb and Cd despite soil contamination of the aforementioned elements was observed. The lack of correlation between the copper and zinc concentration in the bedrock of the Beskidy Mountains and in the moss (*Polytrichum formosum*) was also noted by Panek (2000). There was no correlation between the cadmium content in plants and in the soil layer of 20-40 cm (Table 3). This can be attributed the high value of organic matter and the soil pH in the top layer of the soil which forming a barrier immobilizing the metal in the soil solution hampered the transport of the elements from the lower layers of the soil to plants.

Table 3. The relationship between the cadmium content in the soil (20-40 cm) and in the plant

Heavy metal	Spearman correlation coefficient	p-value	Direction of the relationship	Power of the relationship
Cd	0.181	0.101	---	---

The revealed correlations with the positive direction of the relationship between the soil pH and the heavy metals content in the soil layers of 0-20 cm (Table 4) and of 20-40 cm (Table 5) suggest that an increase in soil pH results in an increase in the content of sparingly soluble metal ions in the given soil layer. The demonstated relationships reflect findings presented in the literature. Numerous authors (Gorlach, Gambuś 1991, Weber 1993, Chłopecka 1994, Gworek and Borowiak 1994, Wisniewski and Kowalewski 1997) confirmed the increase in the mobility of heavy metals and their bioavailability by plants along with the decrease in the pH of the soil that is with the surge in its acidity.

Table 4. The relationship between the pH value and the content of cadmium in soil (0-20 cm)

pH	Spearman correlation coefficient	p-value	Direction of the relationship	Power of the relationship
in H_2O	0.462	<0.001	positive	average
in KCl	0.489	<0.001	positive	average

The decrease of the soil pH to acidic and strongly acidic results in the increase in the concentration of active metal forms available for plants in the soil solution. The increase in soil pH that is lowering its acidity to a slightly acidic pH as well as neutral and alkaline cause the increase in the concentration of metals in the soil levels (Weber, 1993; Chłopecka 1994).

Table 5. The relationship between the pH value and the content of cadmium in soil (20-40 cm)

pH	Spearman correlation coefficient	p-value	Direction of the relationship	Power of the relationship
in H_2O	0.32	0.003	positive	average
in KCl	0.451	<0.001	positive	average

The demonstrated positive correlations between the content of organic matter and heavy metals in the soil, both in the layer of 0-20 cm (Table 6) and in the layer of 20-40 cm (Table 7) as well as between the organic matter content influenced the vegetation contamination with heavy metals and the higher accumulation of elements in the soil material than in the plant material.

Table 6. The relationship between the content of organic matter and cadmium in soil (0-20 cm)

Heavy metal	Spearman correlation coefficient	p-value	Direction of the relationship	Power of the relationship
Cd	0.503	<0.001	positive	strong

The presented relationships indicate the existence of high impact of organic matter content on the soil pH (Wiater 2008, Kwiatkowska-Malina and Maciejewska 2009, Józefowska 2009), and thereby on the immobilization of heavy metals in the soil layers concerned, on precipitation of sparingly accessible metal ions to plants, on limiting their bioavailability for plants, on obstruction of the transport of metals from the soil environment to the aboveground parts of plants and thereby on reducing the fitoavailability of ions of the elements.

Table 7. The relationship between the content of organic matter and cadmium in soil (20-40 cm)

Heavy metal	Spearman correlation coefficient	p-value	Direction of the relationship	Power of the relationship
Cd	0,585	<0,001	positive	strong

The verification of the naturalness of systems of soil levels in the study area of the municipality of Trzebinia was also the aim of determining the content of trace elements and soil parameters for the two soil layers i.e. 0-20 cm and 20-40 cm. The natural or disturbed order of the layers of the soil profile is one of the important factors affecting the distribution of heavy metals in the soil environment, the transport of elements between soil and plants, and thus their availability for plants (Kabata-Pendias and Szteke 2012, Yan *et al*. 2012, Remon *et al*. 2013). In most research areas, the metal content decreased with the depth of the soil profile which should be associated with the natural distribution of soil levels. The distribution of metals in the soil, therefore, enhanced their bioavailability because plants absorb most of the components from the top layer of soil.

CONCLUSIONS

1. The obtained results of the concentration of cadmium in soil samples are higher than the national standards designated for agricultural lands (Journal of Laws 2002) both at the depth of 0-20 cm and 20-40 cm. It can therefore be concluded that soils of the municipality of Trzebinia are contaminated with cadmium. The causes of the contamination might be associated with intensive, long-term activities of mining and processing conducted in the areas which are currently fallows and industrially active.
2. The research carried out on fallow lands in the municipality of Trzebinia showed a positive direction of the relationship between the concentration of cadmium in the soil layer of 0-20 cm and the above ground parts of plants.
3. Soil factors significantly influenced the migration of the element from the soil environment to plants. The observed concentrations of cadmium in the plant samples should not be regarded as a simple reflection of its content in the soil material.
4. The disclosed content of cadmium in plant material disables its use as feed for breeding purposes. The concentration of the element exceeded the limit values for the feed specified in the Regulation of the Minister of Agriculture and Rural Development (2012a).

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Received: 07.09.2016
Accepted: 13.10.2016