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Cadmium, lead, zinc and copper content in herbaceous plants overgrowing furnace waste landfill

Zawartość kadmu, ołowiu, cynku i miedzi w roślinności zielnej porastającej składowisko odpadów paleniskowych

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Słowa kluczowe: roślinność zielna, odpady paleniskowe, metale ciężkie

Abstract
Electricity and heat have been produced in Poland by the combustion of coal. Ash-slag, a by-product of this process, has been mainly deposited in landfills. Until the end of 2011, about 258 million tonnes of ash-slag have been deposited in landfills. Herbaceous plant and soil samples were taken from four directions (east, west, south and north) in three locations on the landfill embankment: base, middle and top. The negative influence of furnace waste landfill on herbaceous plants was confirmed by the phytoaccumulation indexes (WF) of the examined elements: WF Cd – medium to intensive; WF Pb – low to medium; WF Zn – low to intensive and WF Cu – low to medium.

Streszczenie
W procesach wytwarzania energii elektrycznej i cieplnej w wyniku spalania węgli powstają różnorodne odpady energetyczne. Odpady te należą do tzw. odpadów masowych. Przez dziesiątki lat stanowiły poważny problem środowiskowy, bowiem stopień ich gospodarczego wykorzystania był niezadowalający, a znaczna ich część była lokowana na składowiskach. Celem badań była ocena zawartości wybranych pierwiastków śladowych w roślinach zielnych porastających składowisko odpadów Elektrowni Skawina S.A. Pobierano materiał roślinny i glebę na czterech podstawowych kierunkach geograficznych, u podstawy nasypu, w jego środku oraz na koronie składowiska. W tak przygotowanym materiale przeprowadzono mineralizację suchą z roztwarzaniem w odniesieniu do roślin oraz mineralizację na mokro w odniesieniu do gleb. Oznaczenie kadmu, ołowiu, cynku i miedzi wykonano metodą AAS. Negatywny wpływ składowiska odpadów paleniskowych na porastającą go roślinność zielną potwierdzają obliczone wskaźniki fitokumulacji badanych pierwiastków (WF Cd – średni do intensywnego, WF Pb – słaby do średniego, WF Zn – słaby do intensywnego, WF Cu – słaby do średniego). Za najbardziej zanieczyszczoną analizowanymi pierwiastkami należy uznać roślinność zielną porastającą podstawę i środkową część obwałowania po stronie wschodniej składowiska. Jest to widoczne szczególnie w odniesieniu do zawartości kadmu kumulującego się w tych roślinach w stopniu intensywnym. W żadnej z badanych prób roślinnych nie zostały przekroczone dopuszczalne zawartości kadmu i ołowiu zawarte w Rozporządzeniu Ministra Rolnictwa i Rozwoju Wsi, dotyczącego dopuszczalnych zawartości substancji niepożądanych w paszach. Nie stwierdzono jednoznacznej tendencji, co do zawartości badanych pierwiastków w roślinności porastającej charakterystyczne części obwałowania składowiska, tj. podstawę, część środkową i koronę. Wartości te kształtowały się bardzo nieregularnie w zależności od geograficznego kierunku pobrania prób.

1. INTRODUCTION

The combustion of coal to produce electric and thermal energy results in the generation of diverse waste products such as fly ash, slag, ash-slag mixture, microspheres, ashes from fluidised-bed furnaces, gypsum from desulphurisation of flue gas using the hydrated lime method or waste from gas desulphurisation using semi-dry or dry methods. This waste, classified as the so-called mass waste, was a serious environmental problem for dozens of years because the degree of its usage was not satisfactory and most of it was deposited on landfills [Galos and Uliasz-Bochenczyk 2005, Woźniak and Klisik 2007]. In recent years, there has been a significant increase in the degree of usage of this waste in Poland. In the year 2011, 12.8% of the ash-slag mixture from wet waste treatment was used and 95.2% of fly ash from coal. Despite this, about 258 million tonnes of the ash-slag mixture are still deposited in landfills, which has been resulting in many threats concerning environment [GUS 2012]. Landfills of fine-grained (dusty) mineral waste from the coal power industry show high susceptibility to wind and water erosion. Technical methods of its prevention mostly do not eliminate dust nuisance caused by the deposited waste. The creation of a dense plant cover can completely eliminate or successfully minimise dusting and washing out of the landfill. Plant cover can be created as planned or can be created as temporary. The temporary vegetation should be modify to the form and structure planned by environmental engineers [Siuta 2005]. So far, many papers have been devoted to experimental research on the development and chemistry of different types of plants (mainly grasses) in landfills using diverse sowing techniques and fertilisation methods [Kitczak et al. 2003, Siuta 2005, Antonkiewicz and Radkowski 2006, Zieliński 2007, Dyguś and Madej 2012]. The purpose of the study was to evaluate the content of the chosen trace elements in herbaceous plants overgrowing the landfills of the Skawina Power Plant.

2. MATERIALS AND METHODS

Soil and plant samples were taken in September 2010 from a ash and slag landfill (pools C-2, C-3) of the Skawina Power Plant located in Borek Szlachecki, near Krakow. Samples were taken from three characteristic points, starting from the embankment base, then moving to the middle part and finally to the top, in four basic geographic directions (east, west, north and south). One of the tested sites was an area of about 25 m^2, from which five samples of primal herbaceous plants and soil were taken. After homogenisation, the averaged samples comprised about 1000 g f.m. for plants and about 500 g f.m. for soil. Plants were cut with scissors just over the surface of the soil and soil samples were taken using a soil drill from a layer at a depth of 0–0.2 m. After drying to air-dry matter, plants were ground in a high-speed grinder, whereas soil samples were comminuted in a mortar and then sieved through a 1 mm sieve. For the analysis, 10 g d.m. of homogeneous plant material and 3 g d.m. of homogeneous soil material were taken. Samples were weighed with an accuracy of 0.0001 g. The prepared material was then subjected to dry mineralisation with dissolution for the plant samples and wet mineralisation for the soil samples. The cadmium, lead, zinc and copper contents were determined by flame atomic absorption spectrometry (UNICAM Solaar M6). The phytoaccumulation index (WF) was used as the indicative parameter in the study. It was calculated as the ratio of the metal concentration in a plant to the metal concentration in the soil. WF < 0.01 indicates no phytoaccumulation, WF < 0.1 indicates low phytoaccumulation, WF < 1 indicates medium phytoaccumulation and WF > 1 indicates intensive phytoaccumulation [Łaszewska et al. 2007].

3.RESULTS AND DISCUSSION

The conditions for a plant's growth and life on furnace waste landfills are mostly unfavourable, mainly because of inappropriate air, water and thermal conditions, strong alkaline reaction, difficult access to macronutrients, including an almost complete lack of nitrogen [Siuta 1998, Gilewska and Spychalski 2002, Kitczak et al. 2003]. Moreover, the fact that there is an increased concentration of trace elements, which naturally occur in solid fuel, is also an unfavourable condition for plant growth [Pandey et al. 2009, Jegadeesan et al. 2008]. However, it should be remembered that metals occurring in deposited furnace waste are in many forms and phases that differ in mobility and consequently their toxicity. All these characteristics determine a specific behaviour in environmental conditions and have an influence on living organisms [Świetlik and Trojanowska 2009]. Most often, in order to evaluate the niusance caused by furnace waste, tests are carried out on leachates from a given landfill or on eluates obtained from tested waste by leaching of potential pollutants [Woźniak and Zygadło 2002, Kapuściński and Strzałkowska 2005, Rosik-Dulewska and Karwaczyńska 2008, Gruchot et al. 2012]. The landfill where this research was carried out has been monitored since 2007 for condition and quality of surface water, leachates (supernatant water), ground water and soil. Trend analysis using a temporal system for aggregated data showed that the concentration of sulphur is decreasing over time in water inflows into the landfill and in outflows from the same area [Referat Ochrony Środowiska Elektrowni Skawina 2012].

The Skawina Power Plant's documentation concerning the landfill does not contain specific data about its biological reclamation. Plant material that was taken for the research comprised herbaceous plants overgrowing the entire area of the landfill (apart from working fragment of the landfill). In parts of the embankment on the east and the north, the plant cover also included trees (mainly trembling poplar, birch and locust tree) that are not present at all in the western and southern parts. Such a distinct diversity in flora seems to suggest that some attempts were made in the past to biologically reclaim this area. Trees were found on the leeward part of the landfill, which is a windy region, so it is possible that the trees were planted to protect the houses and fields from dust. At this stage of research, it is difficult to say to what extent biological reclamation is the result of the natural humification processes occurring on the landfill or anthropological actions. Cadmium is one of the metals that migrate within the environment relatively easily with high bioavailability. The concentration of this element in the soil material was in the range from below 0.3 up to 0.53 $mg \cdot kg^{-1}$ d.m. The highest concentrations were determined in samples taken from the base and the top of the embankment on the western side of the landfill (Table 1). Despite the fact that cadmium is not essential for their growth, plants uptake cadmium through roots and leaves alike. Meadow plants can uptake over 50% of this element from the polluted atmosphere. However, metals often remain on the plant but are not within the plant [Jarosz 1994, Filipek-Mazur et al. 2007]. With reference to the present research, it should be noted that the year 2010 was very rainy year. The samples were taken after the rainy period, so dusting, and deposition of metalliferous dust, on the overground parts of the tested plants was slight. The cadmium content in the plant material was in the range 0.10–0.62 $mg \cdot kg^{-1}$ d.m. The highest concentration of this element was observed in the sample taken from the base of the embankment on the east side (Table 1). More than two times lower cadmium concentrations (0.12–0.28 $mg \cdot kg^{-1}$ d.m.) were in grass sword obtained from Warsaw grasslands [Madej et al. 2010]. However, Wowkonowicz et al. [2011] reported that in the grass sword taken from grasslands located around Warsaw, the concentrations were in a much higher range (1.93–3.90 $mg \cdot kg^{-1}$ d.m.). Taking into consideration the fact that the sampled herbaceous plants can be a potential source of food for wild animals, the results can be compared with the permitted values of undesirable substances in fodder as outlined in the Regulation of the Minister of Agriculture and Rural Development [Rozporządzenie 2012]. According to this legal regulation, cadmium content in fodder should not exceed 1 $mg \cdot kg^{-1}$ d.m. In the tested samples, such a concentration was not observed, so it can be concluded that the herbaceous plants covering the landfill fulfil the requirement. To analyse the migration of trace elements from the soil to the plant, phytoaccumulation indexes (WF) are used, which describe the ability of the plant to uptake metals from the soil [Łaszewska et al. 2007]. Intensive cadmium accumulation occurred in the herbaceous plants taken from the base of the embankment (WF = 2.07) and from the middle part (WF = 1.07) on the east side of the landfill (Fig. 1). High indication of phytoaccumulation of this element was also seen in plants overgrowing the middle part on the north side (WF = 1.0) and the base of the embankment on the west side (WF = 1.0). All the other plant samples were characterised by a medium degree of cadmium phytoaccumulation.

In the sampled soil, lead content varied in the range 9.1–38.0 $mg \cdot kg^{-1}$ d.m.; the highest concentration of this metal was in the sample taken from the top of the embankment on the north side of the landfill. Plant samples taken at this point also had the highest content of this element (2.8 $mg \cdot kg^{-1}$ d.m.; Table 1). These values are much lower than reported by Wowkonowicz et al. [2011], who stated that in grasses from the Warsaw area the lead content was in the range 3.9–7.5 $mg \cdot kg^{-1}$ d.m. Madej et al. [2010] also stated that lead content was in the range 1.21–14.1 $mg \cdot kg^{-1}$ d.m. in samples of newly grown grass obtained from the Warsaw area

Table 1. Heavy metal contents in plants and soil samples taken from the Skawina Power Plant landfill

Sample number	Location		Content in plants (mg•kg⁻¹ d.m.)				Content in soil (mg•kg⁻¹ d.m.)			
			Cd	Pb	Zn	Cu	Cd	Pb	Zn	Cu
1	East	Base	0.62	1.3	67.2	4.5	0.30	9.1	45.8	6.6
2		Middle	0.32	2.5	56.1	5.8	0.29	14.1	68.7	22.4
3		Top	0.13	2.4	38.7	8.1	0.31	34.3	161.0	64.2
4	South	Base	0.17	2.2	45.9	5.3	0.28	28.7	135.1	56.0
5		Middle	0.10	0.5	17.7	1.8	0.30	29.7	142.6	62.4
6		Top	0.10	0.8	13.6	2.1	0.31	33.9	134.9	59.6
7	North	Base	0.20	1.9	26.7	5.6	0.28	16.9	74.8	27.5
8		Middle	0.33	1.1	43.0	3.0	0.33	27.1	155.2	74.5
9		Top	0.23	2.8	48.4	7.2	0.40	38.0	143.8	55.8
10	West	Base	0.53	1.0	27.9	2.3	0.53	20.4	70.4	6.9
11		Middle	0.19	1.1	24.4	2.6	0.31	19.2	101.1	61.4
12		Top	0.10	1.3	24.4	3.8	0.37	30.6	130.9	63.5

The uptake of lead from the soil by plants is limited because there is a root barrier that reduces its absorption [Gruca-Królikowska and Wacławek, 2006]. According to Kabata-Pendias and Pendias [1999], for plants the critical concentration of lead in soil is in the range 30–300 mg•kg^{-1} d.m. One of the reasons for the weakening of vegetation under the influence of lead (and also cadmium) is the lowering of the biosynthesis of chlorophyll [Bayçu et al. 2003]. When evaluating the usability of the plant material as fodder in terms of the lead content, it can be stated that all the samples fulfilled the requirements presented in the Regulation of the Minister of Agriculture and Rural Development [Rozporządzenie 2012]. The herbaceous plants were characterised by low (WF 0.016–0.080) and medium (WF 0.11–0.18) degree of phytoaccumulation of lead (Fig. 1). Zinc, one of the micronutrients, is one of the most mobile metals in soil; it accumulates in the top layers, is easily soluble and more available for plants [Węglarzy 2007]. In the soil sample taken for this study, the concentration of zinc was in the range 45.8–161.0 mg•kg^{-1} d.m. The highest concentration of this metal was determined in the sample taken from the top of the embankment on the east side (Table 1). It should be remembered that an excess of this element causes chlorotic and necrotic changes in leaves, and it also reduces sprouting and growth. Toxicity can occur at a concentration of only about 100 mg•kg^{-1} d.m. [Kabata-Pendias and Pendias 1999]. Zinc content in the analysed plant material was about 13.6–67.2 mg•kg^{-1} d.m. The highest concentration of zinc, like in the case of cadmium, was in the sample taken from the base of the landfill embankment on the east side (Table 1). In tests carried out on grasses from the Warsaw area, Wowkonowicz et al. [2011] determined the content of zinc to be in the range 16–48 mg•kg^{-1} d.m. Madej et al. [2010] found that the zinc content in newly grown grass in the Warsaw area was a bit higher, in the range 55.0–96.2 mg•kg^{-1} d.m. In the sample taken from the base of the embankment on the east side, there was an intensive degree of zinc phytoaccumulation (WF = 1.47); the sample taken from the top on the south side was characterised by a low degree of phytoaccumulation (WF = 0.10), and in all the other samples, there was a medium degree of phytoaccumulation of this element (WF 0.12–0.82; Fig. 1). Copper, after zinc, is the second of tested microelements. The uptake of this element from the soil is much harder than the uptake of zinc or cadmium. As Sukreeyapongse et al. [2002] report, it is related to the organic matter content and the soil's reaction; the more acidic the soil, the more weakly the metal is bonded. Power plant waste is mostly characterised by relatively high pH [Kapuściński and Strzałkowska 2005, Rosik-Dulewska and Karwaczyńska 2008]. In the case of the tested soil, the reaction determined in distilled water was in the range 6.5–7.6. The highest concentrations of copper were in soil samples taken from the base of the embankment on the east side (6.6 mg•kg^{-1} d.m.) and the west side (6.9 mg•kg^{-1} d.m.). When a plant uptakes too much copper, similar to lead and cadmium, it lowers the chlorophyll biosynthesis, although copper is an element that is essential for the proper growth of plants. The copper content in the tested plant material varied from 1.8 to 8.1 mg•kg^{-1} d.m. (Table 1). The highest concentration of this element was observed in the sample taken from the top of the embankment on the east side, whereas the lowest concentration, similar to lead, was observed in the middle part of the embankment on the south side. This element was characterised by low (0.03–0.09) and medium phytoaccumulation indexes (WF 0.13–0.68); in both cases, it contained six samples (Fig. 1).

4. CONCLUSIONS

1. The negative influence of furnace waste landfill on overgrowing herbaceous plants is confirmed by the calculated phytoaccumulation indexes of the tested elements (WF Cd – medium to intensive, WF Pb – low to medium, WF Zn – low to intensive and WF Cu – low to medium).
2. The herbaceous plants overgrowing the base and the middle part of the embankment on the east side of the landfill should be regarded as the most polluted with the tested elements. It is particularly noticeable in terms of the cadmium content, which is cumulated in the plants (WF Cd–intensive).
3. In none of the tested plant samples, the values for cadmium and lead content exceeded the permitted level of undesirable substances in fodder as outlined in the Regulation of the Minister of Agriculture and Rural Development.
4. The tested elements in plants overgrowing the characteristic locations of the landfill, that is, the base, the middle and the top, no explicit trend was observed. The concentrations of the elements were very irregular depending on the geographic direction of sampling.

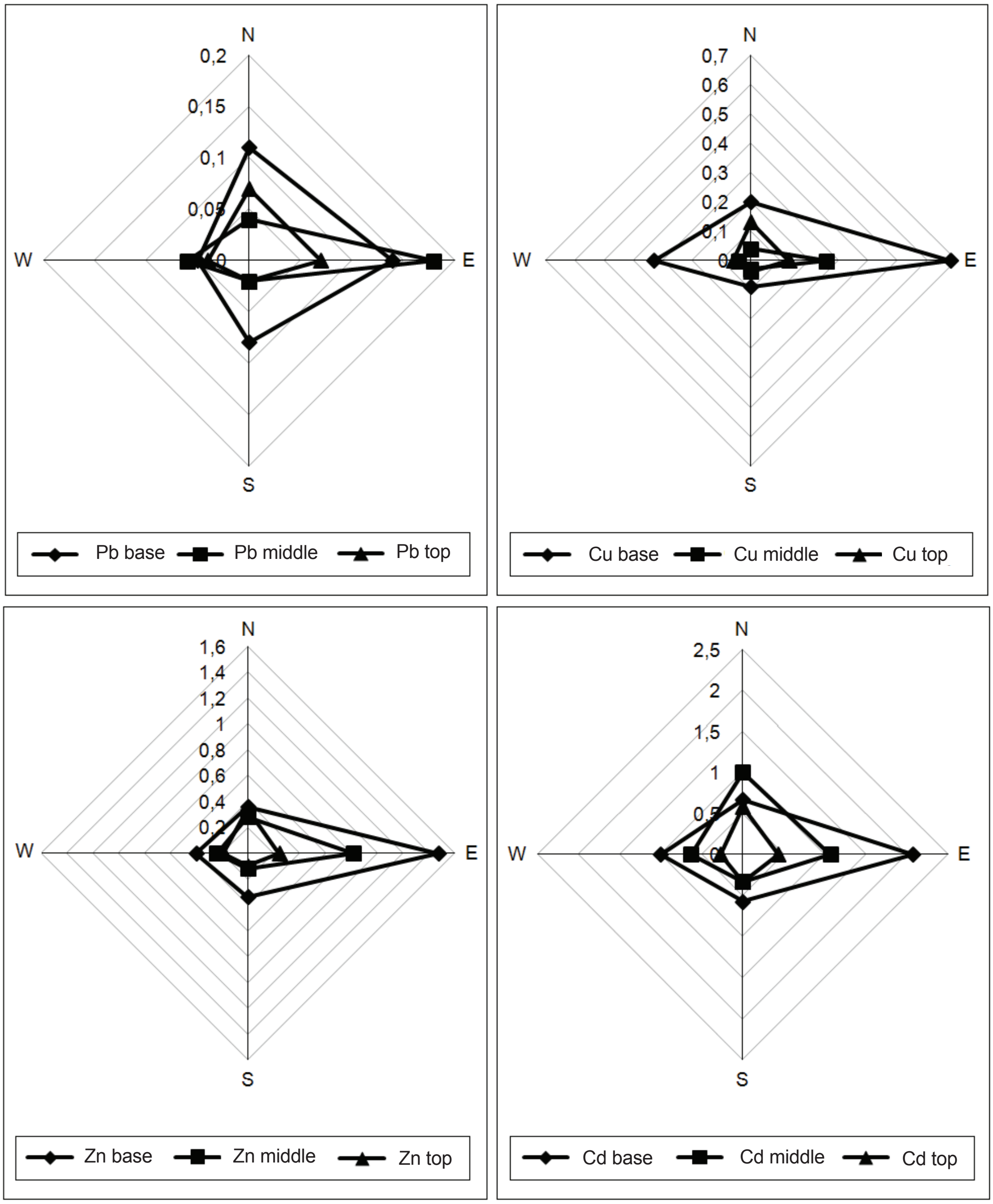

Fig. 1. Lead, copper, zinc and cadmium phytoaccumulation indexes.

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