FOLIA POMERANAE UNIVERSITATIS TECHNOLOGIAE STETINENSIS
Folia Pomer. Univ. Technol. Stetin., Agric., Aliment., Pisc., Zootech. 2015, 318(34)2, 47–56

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ANTHROPOGENIC INDUSTRISOLS WITHIN EWA PENINSULA IN SZCZECIN PORT PART II. CONTENT OF HEAVY METALS

ANTROPOGENICZNE GLEBY INDUSTRIOZIEMNE W OBRĘBIE PÓŁWYSPU EWA W PORCIE SZCZECIN CZĘŚĆ II. ZAWARTOŚĆ METALI CIĘŻKICH

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Streszczenie. Materiał glebowy zebrany do I części opracowania pt. „Budowa stratygraficzna i właściwości chemiczne, z uwzględnieniem makroskładników gleb industrioziemnych (przykrytych–inicjalnych oraz próchnicznych) w obrębie półwyspu Ewa w Porcie Szczecin" posłużył do ustalenia w wymienionych glebach, uformowanych z warstw nasypowych, zawartości metali ciężkich: Cd, Pb, Zn, Cu, Ni, Co, Mn i Fe. Badania wykazały, że gleby uszczelnione płytami żelbetonowymi w mineralnej warstwie nasypowej na głębokości 0,25–1,50 m, na niewielkiej przestrzeni, zawierają bardzo wysoką zawartość ołowiu wynoszącą 11 755,0 mg · kg^{-1} s.m., któremu towarzyszy zawartość miedzi – 135,6 mg · kg^{-1} s.m. Tak wysokie zanieczyszczenie, w zasadzie punktowe, obejmowało tylko kilka do kilkunastu m^3 nasypowego piaszczystego materiału glebowego. W świetle polskich przepisów prawnych wymieniona zawartość ołowiu przekracza dopuszczalne stężenie tego pierwiastka dla gruntów grupy C (grunty przemysłowe, kopalne i tereny komunikacyjne). Występujące w sąsiedztwie gleb przykrytych tereny pozbawione uszczelnienia gleby industrioziemne próchniczne, stanowiące skwery roślinne, wykazywały także w warstwie 0,0–0,20(0,30) m podwyższoną średnią zawartość metali ciężkich: Cd – 1,36; Pb – 409,4; Zn – 673,6; Cu 89,6 mg · kg^{-1} s.m. Dużą rolę przy nagromadzaniu wymienionych metali prawdopodobnie odegrało rozprzestrzenianie się zanieczyszczeń pyłowych wynikających z przeładunkowej działalności portowej.

Key words: heavy metals, industrisols.
Słowa kluczowe: gleby industrioziemne, metale ciężkie.

INTRODUCTION

Many authors e.g. Czarnowska et al. (1983), Wittig (1995), Burghardt (1996, 2000), Niedźwiecki et al. (2000a, b), Pasieczna (2003), Greinert (2003), Zimny (2005), Czarnowska and Nowakowski (2006), Sun et al. (2010) emphasise that, a unique ecosystem of urban agglomerations, especially industrial-urban, subjected to various anthropogenic pressures, is characterised by intensification of pollution, compared to non-urbanised areas. Pollution

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accumulation is observed both in soils and plants of municipal green areas. According to Wixon and Davies (1994), Górka at al. (1998), Kabata-Pendias and Pendias (1999), and Wei and Yang (2010) among extremely dangerous sources of city environment pollution, posing a threat to the health of inhabitants, animals and plants is dust and gas pollution of city air, containing heavy metals. This serious threat results from the rapid and uncontrollable spreading of emitted dust and gas contamination as well as their immediate harmful impact not only on soils and living organisms but also buildings, water (surface water, in particular) and other elements of urban environment. The measurements of dust fallout, conducted in Szczecin, in the years 1996–2000, by State Sanitary Inspection showed its distinct emission reduction into atmosphere. At the same time, however, there was an increase in lead content in dust particles PM10 (Inspekcja Ochrony Środowiska 2001). Sporadically, mainly due to intensified car traffic, the permissible lead fallout value – 100 mg Pb $\cdot$ m^{-2} $\cdot$ yea^{-1}, was exceeded. Air quality standards for 24-hour concentration of dust particles PM10, in Szczecin agglomeration are still exceeded despite the implementation of air protection programme (Raport… 2013).

As a result, in the case of uncovered areas of Szczecin agglomeration, the studies of Piasecki et al. (1995), Wojcieszczuk and Niedźwiecki (2003), Niedźwiecki et al. (2004, 2009), Chudecka and Krzywy-Gawrońska (2012) indicate a slight accumulation of heavy metals, mostly lead, zinc and copper in the central part of city, first of all, in the surface layer of soils adjacent to streets. Generally, the content of these elements is characteristic of I degree of contamination according to Kabata-Pendias et al. (1995). Apart from that, Piaseczna (2003) draws attention to the occurrence in Szczecin the so-called point sources of pollution connected mainly with former industrial objects which had contributed to high accumulation of heavy metals in soils. The author's papers show that the most contaminated by lead (180–300 mg $\cdot$ kg^{-1}), are surface layers of soils near the former Smelter Plant Szczecin, on Phosphorus Quay, within the former Szczecin Shipyard and in Zdroje near the sewage treatment plant . Maximum lead content (810 mg $\cdot$ kg^{-1}) was found in the surface layer of soil around the former Plant of Artificial Fibres „Wiskord". The above mentioned areas were also strongly contaminated by zinc and copper e.g. in the topsoil of former Szczecin Shipyard maximum content of zinc (1176 mg $\cdot$ kg^{-1}) was detected and in that one near ZWS „Wiskord" – 154 mg $\cdot$ kg^{-1} of copper. Some of these results, were confirmed by Kiepas-Kokot (2014), who recorded an extremely high content of copper – 19 995 mg $\cdot$ kg^{-1} and the highest coefficient of its variability – 195% in the case of the area of Tele-Fonika Kable – Szczecin.

In the light of presented contamination of urban Szczecin soils, there has been the lack of the data on the level of dust particles, their chemical composition and the content of heavy metals in the soils of Szczecin port, where mineral raw materials containing trace elements are reloaded. Existing values concern only the areas adjacent to the port. Great variety of cargoes and operations inevitably has had a harmful impact on natural environment, mostly because of cargo dusting and spilling as well as accumulation of different kind of waste, demanding utilisation. Hence, the accumulation of heavy metals, especially in unsealed soils. The literature on this subject, shows the level of cadmium and lead in the dust fallout, within the warehouses and repair workshops of Grain Elevator „Ewa" (Niedźwiecki et al. 2000a). Another focus of attention is Ostrów Grabowski, whose area is being prepared for future construction works aiming at port extension. This area has been covered with over 5 mln m^3 of bottom sediments dredged from the terminal section of Świnoujście-Szczecin fairway, port basins and canals as well as from deepening water bodies along numerous quays. The sediments dredged from the mentioned places, with over 10% of organic matter, contained considerable amounts of macro and microelements, including heavy metals (Protasowicki and Niedźwiecki 1995; Protasowicki et al. 1999; Niedźwiecki et al. 2001). The cited papers do not deal with negative influence of particular port quays on the environment, including topsoil.

Therefore, the aim of this research was to determine the degree of heavy metal contamination in anthropogenic industrisols within Ewa Peninsula in Szczecin port.

MATERIAL AND METHODS

This paper is the continuation of Part I presenting the stratigraphy and chemical properties, including macroelements, of industrisols (initial and humus) within Ewa Peninsula, in the port of Szczecin. In Part II, the material collected earlier in the form of 72 composite samples was used and the content of Cd, Pb, Zn, Ni, Co, Mn, Fe was determined.

In analysed soil material 57 composite samples (31 mineral and 27 organic) were taken from 12 deep (10–13 m) boreholes within the area, sealed with reinforced concrete slabs, at Slovakian Quay. They represented initial soils (ekranic), in which 3.20 m thick surface layer, consisted of transported (mostly sandy) material to improve a swampy ground before construction works. The remaining material, 15 composite samples (10 mineral and 5 organic) came from the depth of 0–0.20(0.30) m of unsealed surfaces occuring sporadically on the examined peninsula. Sampling from deeper parts of soil profile was impossible because of port infrastructure. This materiał was also transported during area improvement and had similar stratigraphy to sealed soils. These samples represented humus industrisols.

Due to the role of organic matter in shaping sorption capacity and its protective action, consisting in immobilisation of toxic substances (heavy metals, pesticides, petroleum derivatives) penetrating the soil, in this paper, apart from heavy metal content, organic carbon values were determined by the Tiurin method.

Total content of above mentioned elements was obtained by soil mineralisation in the mixture of concentrated $HNO_3 + HClO_4$ (ratio 4 : 1), and determinations using atomic absorption spectrophotometer Unicam-Salaar 929.

RESULTS AND DISCUSSION

The quays of Szczecin port, where ships are serviced, are generally covered (sealed) with concrete slabs, bituminous surface or other sealing materials. As it had already been stated in Part I, similar conditions occurred on Slovakian Quay of Ewa Peninsula where deep boreholes (10–13 m below ground level) were made for sampling the material for laboratory studies. Their results are presented in Table 1.

Table 1. Content of heavy metals (mg · kg^{-1} DM) soluble in concentrated HNO_3 + $HClO_4$ in industrisols (initial-ekranic) on Slovakian Quay within Ewa Peninsula in port of Szczecin (mean and extreme values)
Tabela 1. Zawartość metali ciężkich (mg · kg^{-1} s.m.) rozpuszczalnych w mieszaninie stężonych kwasów HNO_3 + $HClO_4$ w industrioziemnych (inicjalnych–przykrytych) glebach na Nabrzeżu Słowackim w obrębie półwyspu Ewa w porcie Szczecin (wartości średnie i ekstremalne)

Kind of soil sample Rodzaj próbek glebowych	Depth Głębokość [m]	C [g · kg^{-1}]	Cd	Pb	Zn	Cu	Ni	Co	Mn	Fe
Mineral Mineralne	0.25–1.50	7.0 2.9–14.1	0.58 0.03–1.71	1114.2* 6.3–11755.0	133.3 43.6–278.8	28.1** 3.8–135.6	9.20 3.32–20.75	1.95 0.04–6.73	247.2 43.3–567.0	12668 4146–50440
Organic Organiczne	1.00–1.50	148.3 132.0–164.6	1.62 0.59–2.65	100.6 71.9–129.4	339.4 296.4–382.0	45.5 44.1–46.9	23.91 20.36–27.46	8.91 7.44–10.39	824.5 473.0–1176.0	29350 22900–35800
Mineral (no organic materiał) Mineralne (materiał organiczny nie występuje)	1.50–3.20	2.2 0.4–4.1	0.72 0.02–1.95	7.4 1.3–19.2	12.6 7.2–19.9	3.1 1.9–6.8	5.15 3.89–8.15	0.50 0.01–1.79	53.6 42.8–93.2	2775 2253–4516
Mineral Mineralne	3.20–4.50	n.o.	1.20 0.46–1.95	6.1 2.5–9.8	12.0 10.4–13.6	2.5 2.2–2.9	6.45 4.75–8.15	0.74 0.51–0.98	52.3 49.3–55.3	2332 2253–2411
Organic Organiczne		174.1 103.6–247.1	0.32 0.01–0.92	26.1 3.7–37.9	79.7 23.2–189.9	25.7 15.2–35.5	22.88 17.40–30.99	6.26 2.04–12.63	692.4 338.5–1450.0	29684 2468–56280
Organic (no mineral materiałl) Mineralne (materiał organiczny nie występuje)	4.50–8.50 (9.00)	183.7 83.1–367.9	0.46 0.01–1.71	9.7 2.9–22.3	51.2 8.9–81.5	17.4 11.5–27.4	22.4 11.47–32.97	7.29 1.07–12.19	817.0 302.9–2103.2	29277 11085–59505
Mineral (no organic material) Mineralne (materiał organiczny nie występuje)	8.50 (9.00)–13.0	n.o.***	0.50 0.01–1.33	2.3 0.5-5.1	17.9 10.1–40.8	2.9 1.6–6.2	5.92 1.69–9.19	1.80 0.42–3.99	189.9 108.2–308.9	10170 3931–18165

* mean content of Pb disregarding maximum value of borehole X (11 755.0 mg Pb · kg^{-1} DM) is 50.1 mg Pb · kg^{-1} DM value range 6.3–141.4 mg · kg^{-1} DM – średnia zawartość Pb bez uwzględnienia wartości maksymalnej występującej w odwiercie XI (11 755,0 mg Pb · kg^{-1} s.m. gleby) wynosi 50,1 mg Pb · kg^{-1} s.m. gleby przy wahaniach 6,3–141,4 mg · kg^{-1} s.m. gleby.
** mean content of Cu disregarding maximum value of borehole XI (135.6 mg Cu · kg^{-1} DM) is 17.3 mg Cu · kg^{-1} DM value range 3.8–41.3 mg · kg^{-1} DM – średnia zawartość Cu bez uwzględnienia wartości maksymalnej występującej w odwiercie XI (135,6 mg Cu · kg^{-1} s.m. gleby) wynosi 17,3 mg Cu · kg^{-1} s.m. gleby przy wahaniach 3,8–41,3 mg · kg^{-1} s.m. gleby.
*** not determined – nie oznaczono.

The highest variations in heavy metal content, expressed in mg · kg^{-1} DM, were found in transported mineral layer at the depth 0.25–1.5 m. They concerned, predominantly, the content of: lead (6.3–11755.0), copper (3.8–135.6) and zinc (43.6–278.8). Maximum values for the above mentioned elements at this depth were found in the sandy borehole XI, located in the central part of the study area. Mean content of Pb in this layer, disregarding the maximum value of borehole XI, amounts only to 50.1 mg · kg^{-1} DM, value range – 6.3–141.4 mg · kg^{-1} DM. In the case of Cu, disregarding the maximum value of surface layer of borehole XI, the mean value is 17.3 mg · kg^{-1} DM and the values range from 3.8 to 41.3 mg · kg^{-1} DM. As far as Zn is concerned, the maximum value – 278.8 mg · kg^{-1} DM, obtained in the surface material of borehole XI, was not the only one since similar accumulation of this element, at the depth 0.25–1.50 m took place in borehole XVII. Currently, it is difficult to find the cause of such strong contamination, occuring only on a few or several m^3 of transported sandy material. Contaminants might have been introduced into the examined area during the formation of transported layer because sporadically found here, organic matter, in comparison with indigenous (peat) material occuring at the depth 3.20–8.50 (9.00) m, was more strongly contaminated by cadmium, lead, copper and zinc. However, the contents of: nickel, cobalt, manganese and iron were maintained at the level close to natural (Table 1). The studies of Chudecka (2009) on the soil substrate in anthropogenic layer of the oldest part of Szczecin, close to Ewa Peninsula, support our opinion about the introduction of lead and copper during the area preparation for the construction of port facilities on Ewa Peninsula. Her studies showed that the most strongly contaminated with lead and copper, anthropogenic formations, were located at the depth 5–6 m below ground level within the Old Town, the area outside the defensive walls, the oldest port and fish settlement, where the oldest settlements had appeared. Such soil material might have also been introduced into Ewa Peninsula. The concentration of heavy metals in soils of the oldest urban parts, is more and more frequently mentioned in soil science literature. Lewandowski and Burghardt (1998) states that the soils of similar areas in Poznań contained e.g. high amounts of: copper to 2130 mg · kg^{-1}, lead to 1093 mg · kg^{-1} and zinc to 960 mg · kg^{-1}. It is worth mentioning that lead and its alloys were commonly used in Poland from 14 century (Molenda 1987).

In the case of surfaced areas of Ewa Peninsula, a certain role might have been played by war actions, carpet air-raids by Allies, in particular, during World War II.

Revealed in these soils at the depth 0.25–1.50 m, very high concentration of lead, to 11 755 mg · kg^{-1} DM and above standard (Ordinance of the Minister of Environment, DzU 2002) level of copper to 135.6 mg · kg^{-1} DM points to their immobilisation as its concentrations in the material lying below, were definitely lower (Table 1). The content of analysed heavy metals in the organic indigenous material at the depth below 3.20 m, was within natural values for sediments (Förstner and Müller, 1974) and organic soils (Kabata--Pendias et al. 1995). According to Scalenghe and Marsan (2009) sealing the ground with impervious material is harmful to soil ecological functions but protects the soil from dust and gas pollution produced during unloading mineral raw materials. Uncovered humus industrisols of green areas of Ewa Peninsula are devoid of this protection.

In the topsoil of these humus industrisols, containing 86.6 g · kg^{-1} of organic carbon, the mean content (mg · kg^{-1} DM) of cadmium was 1.36, thus 2.1-fold higher, compared to its amount in sandy soil with 28.6 g · kg^{-1} C (Table 2).

Table 2. Content of heavy metals (mg · kg^{-1} DM) soluble in concentrated HNO_3 + $HClO_4$ in topsoil of humus industrisols on Slovakian Quay within Ewa Peninsula in port of Szczecin (mean and extreme values)
Tabela 2. Zawartość metali ciężkich (mg · kg^{-1} s.m.) rozpuszczalnych w mieszaninie stężonych kwasów HNO_3 + $HClO_4$ w powierzchniowej warstwie industrialnych próchnicznych gleb na Nabrzeżu Słowackim w obrębie półwyspu Ewa w porcie Szczecin (wartości średnie i ekstremalne)

Kind of Samales Rodzaj próbek glebowych	Depth Głębokość [m]	Number of samples [p.c.s] Ilość analizowanych próbek [szt.]	C [g · kg^{-1}]	Cd	Pb	Zn	Cu	Ni	Co	Mn	Fe
Mineral Mineralne	0.0– –0.20(0.30)	10	28.6	0.64 0.04–3.21	133.6 57.9–562.2	336.7 101.8–1318.8	38.7 22.8–78.6	16.99 10.28–29.47	4.30 2.19–9.87	356 229–1100	11136 8500–17465
Organic Organiczne	0.0– –0.20(0.30)	5	86.6	1.36 0.38–1.91	409.4 88.4–592.2	673.6 402.8–907.4	89.6 57.0–137.4	27.13 18.22–32.51	5.67 4.15–6.42	1777 273–3120	16556 12295–24120

In the case of lead it was 409.4 so 3.1–fold higher than in the sandy soil under study, whereas in the case of zinc – 673,6 exceeding 2,0-fold the content in the sandy soil. The content of copper amounted to 89.6 and 2.3-fold exceeded the mean value for this element in sandy soils. The amount of manganese was 1777 and 5.0-fold surpassed the values found in sandy soil. The differences in the content of nickel, cobalt and iron were considerably smaller (Table 2). The data reported above confirm commonly known high sorption capacity of organic matter but it should be noted that the maximum amounts of cadmium 3.21 and zinc 1318.8 mg · kg^{-1} DM were detected in single samples of mineral soils (Table 2).

Recorded heavy metal accumulation in the topsoil of anthropogenic humus industrisols on Ewa Peninsula, in the light of the limits given by Kabata-Pendias at al. (1995) for the upper layer of arable land, reached II degree in the case of lead and copper, typical of slightly contaminated soils and III degree in the case of zinc, characteristic of moderately contaminated soils. According to current Polish requirements and legal regulations comprised in the Ordinance of the Minister of Environment (Ordinance of the Minister of Environment, DzU 2002) the examined Ewa Peninsula should be included in group C – industrial, mining and traffic areas.

These regulations classify the soil or land as contaminated when the level of, at least one, substance exceeds a permissible value. In the analysed industrisols, this value was only the point high concentration of lead.

CONCLUSIONS

1. In the analysed anthropogenic industrisols – initial (ecranic), sealed with reinforced concrete slabs within Peninsula Ewa in Szczecin Port, on a small area, in transported layer at the depth 0.25–1.50 m, an extremely high concentration of lead was found (11 755 mg · kg^{-1} DM). In the light of current legal regulations for group C (industrial and post-mining areas transport routes) such concentration of lead exceeds the permissible limit. However, lead dislocated to the lower strata of the profile has not been found.
2. Despite the fact that on Ewa Peninsula, no raw mineral materials unloading takes place, the surface layer 0–0.20 (0.30) m of humus industrisols (especially organic) is characterised by accumulation of cadmium, lead, zinc, copper and nickel, originating probably from dust pollution spreading from other port quays.

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Abstract. Soil material collected for Part I, Stratigraphy, chemical properties, including macroelements of industrisols (initial and humus) within Peninsula Ewa in Szczecin Port was used for determining the concentration of heavy metals: Cd, Pb, Zn, Cu, Ni, Co, Mn and Fe in these soils. The studies showed that the soils sealed with reinforced concrete slabs, in their surface layer 0.25–1.50 m, on a small area, contained an extremely high amount of lead 11 755.0 mg · kg^{-1} DM, accompanied by the content of copper amounting to – 135.6 mg · kg^{-1} DM. Such a high, point, contamination occurred only on a few to several m^3 of transported sandy material. In the light of Polish legal regulations, the mentioned level of lead exceeds the permissible concentration for the land of group C (industrial, mining and traffic areas). Adjacent to the sealed soils, humus industrisols of green areas, were characterised by elevated mean content of heavy metals: Cd – 1.36; Pb – 409.4; Zn – 673.6; Cu 89.6 mg · kg^{-1} DM in their surface layer 0.0–0.20(0.30) m. A great role in the accumulation of the above metals might have been played by spreading dust pollution caused by other port operations such as cargo unloading and reloading.