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EFFECTS OF SEWAGE SLUDGE ON THE ACCUMULATION OF HEAVY METALS IN SOIL AND IN MIXTURES OF LAWN GRASSES

The study was conducted on three specially prepared test areas along main streets of Białystok. The experimental plots were sown with two lawn grass mixtures: Eko and Roadside, and three doses of sewage sludge 0.0 (control), 7.5 kg/m^2 and 15.0 kg/m^2 were used. The calculated bioconcentration factor made possible to determine the mobility of the various metals from the soil solution to the aerial parts of plants. The obtained results showed that the plants absorbed Cd, Zn, Cu easier than Pb and Ni suggesting a high mobility, in particular of Cd, Zn, Cu, and reflects the easiness extraction of those metals by plants. Significant correlations between concentrations of metals in soil and grass were observed, nickel in soil was significantly correlated with Cd, Zn and Cu in plants, while the Zn content in soil with the level of Cu in plants.

1. INTRODUCTION

Developing of sewage deposits, increasing in amount from year to year, is a serious problem in the sewage management of the majority of cities. According to data of the Polish Central Statistical Office in 2011, sewage deposits produced in 2000 amounted to 359.8 thousand tons of dry matter (d.m.), and it increased to 526.7 thousand tons d.m. in 2010. Living of human beings is connected with production of sewage [1] and this, in turn, causes the need of their treatment. The sludge formed in the process of wastewater treatment is treated as waste, which can be reasonably used to return to the environment, providing simultaneously valuable nutrients and organic matter.

Sewage sludges are recognised by many authors as perfect fertilizers to be applied to treatment and recultivation of grasslands. The use of such material brings many

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benefits. It allows not only recycling of sewage sludge but also restoration of land degraded by human activity or not used by humans [2]. However, in addition to the favourable impact on plants, sewage sludge can have a negative impact on the environment being secondary pollution due to the content of toxic compounds as PCB, PAHs and heavy metals which remain there causing bioaccumulation. Polish legislation specifies limits for heavy metals in sewage sludge and soil in which they are to be applied, depending on how they are handled [3].

According to Babel and Del Mundo Dacera [4], the total content of heavy metals in sediments typically varies within the limits of 0.5–2.0% d.m. of sludge, and in some cases may increase up to around 4.0% d.m. The main source of heavy metals in sewage sludge is industrial waste water and surface water runoff. Heavy metals in effluents in approximately 80.0–90.0% are collected in sewage sludge and the contents of trace elements in sewage sludge may vary depending on the origin of the waste water.

Road transport is one of the main sources of emissions of air and soil pollutants in cities, strongly intercepting natural environment as a whole and having a negative influence on man. As a result of the combustion of fuels in engines of vehicles, the following pollutants are emitted into the air: carbon monoxide, nitrogen oxides, hydrocarbons, polycyclic aromatic hydrocarbons, particulates and heavy metals. Undoubtedly, a serious threat to the environment is emission from the transport sector. In 2008 in Poland due to car traffic 18.3 Mg of lead (ca. 3.3% of the total annual emission), ca. 5.8 Mg of nickel, ca. 3.4 Mg copper, and ca. 0.4 Mg of cadmium (0.2% of the total annual emission) have been emitted [5].

The aim of this study was to evaluate the accumulation of heavy metals (Cd, Cu, Ni, Pb and Zn) in soil and in the aboveground parts of lawn grasses fertilized with municipal sewage sludge from the Municipal Wastewater Treatment Plant in Sokółka.

2. MATERIALS AND METHODS

The study was conducted on three specially prepared test areas along main streets of Białystok (Hetmańska, Piastowska and Popiełuszki). Each test area was divided into 3 blocks (30 m^2 each) which were treated as replicates. In Fall of 2010, the test areas were fertilized with stabilized municipal sewage sludge from the Municipal Wastewater Treatment Plant in Sokółka (and three doses of sewage sludge (containing 19.3% d.m.): 0.0 (control), 7.5 kg/m^2 and 15.0 kg/m^2 were used. Doses of sewage sludge were established according to Kiryluk [6] who found in several year study that the most effective doses for turfing of municipal waste disposal areas were those above 40 t/ha.

Before establishment of experiment both sewage sludge and soil in each combination were analyzed according to the Directive of Evironmental Minister from July 13th, 2010 concerning municipal sewage sludges [3]. Analyzes were done by the Regional Chemical and Agricultural Station in Białystok (Tables 1 and 2). Additionally soil pH in water and in 1 M KCl were determined.

Table 1

Selected physical and chemical properties of soil samples collected on 2010.10.10

Property	Popiełuszki	Hetmańska	Piastowska
pH	7.6	7.9	7.7
Sand contents, %	75.7	75.9	71.9
Silt contents, %	22.3	22.0	25.4
Clay contents, %	2.0	2.1	2.7
Textural class	loamy sand	loamy sand	sandy loam
P_2O_5 contents, mg·(100 g)$^{-1)}$	22.0	7.3	18.4

In the experiment two grass mixtures were used: Eko from Nieznanice Plant Breeding Station which included 30.0% of *Lolium perenne* cv. Niga, 15.0% of *Poa pratensis* cv. Amason, 22.6% of *Festuca rubra* cv. Adio and 32.4% of *Festuca rubra* cv. Nimba. and Roadside from Barenbrug which included 32.0% of *Lolium perenne* cv. Barmedia, 5.0% of *Poa pratensis* cv. Baron, 52.0% of *Festuca rubra rubra* cv. Barustic, 5.0% of *Festuca rubra commutata* cv. Bardiva (BE) and 6.0% of *Festuca rubra commutata* cv. Bardiva (NL).

Table 2

Composition of municipal sewage sludge (MSS) at pH = 6.7

Component	Content
Dry matter, %	19.3
Organic matter, % d. m.	58.4
Total P, % d. m.	2.7
Total N, % d. m.	4.0
Ammonium N, % d. m.	0.1
Ca, % d. m.	5.5
Mg, % d. m.	0.7
Pb, mg·kg d. m.$^{-1}$	23.5
Cd, mg·kg d. m.$^{-1}$	<0.5
Cr, mg·kg d. m.$^{-1}$	58.0
Cu, mg·kg d. m.$^{-1}$	194.0
Ni, mg·kg d. m.$^{-1}$	22.0
Zn, mg·kg d. m.$^{-1}$	1459.0
Hg, mg·kg d. m.$^{-1}$	1.0

In October 2011, samples of above ground grass mixture and soil (0–20 cm) were collected. Concentrations of heavy metals in soils with sewage sludge and in plant material were determined by the Atomic Absorption Spectrometry AAS. Samples were mineralized at 450 °C, the remains were dissolved in 3 M HCl and extracted in aqua regia (3:1 mixture HCl and HNO_3) at 80 °C (PN-ISO 11047:2001) [7].

Bioaccumulation factors for analyzed heavy metals were determined as quotients of average concentration of a given element in plants with respect to its average concentration in soil [8]. Obtained results were analyzed with ANOVA and the coefficients of variation, means and standard deviations were calculated. Correlation between the concentrations of heavy metals in the ground of plants and in soil fertilized with various doses of sewage sludge were calculated using Pearson's correlation factor r for $p \leq 0.05$.

3. RESULTS AND DISCUSSION

Sewage sludge, a by-product of waste water treatment, is becoming an increasingly problematic environmental and economic issue [1]. The addition of soil sludge from the Municipal Sewage Plant in Sokółka positively influenced the growth of the two selected grass mixtures. One of the serious risks of application of sewage sludge may be introducing heavy metals from sludge into soil [9]. Some metals due to their physico-chemical properties may show affinity to other components of sewage sludge and soil [1]. According to Dzierżanowski and Gawroński [10], the factors, which may also indirectly affect the availability of heavy metals are pH, conductivity as well as the presence of other metals, and species of plants themselves.

Table 3

Concentrations of heavy metals in above ground parts of grass mixtures fertilized with different doses of sludge

Grass mixture	Dose of sewage sludge [kg/m^2]	Dry matter (d.m.) [g/m^2]	Plant [mg/kg d.m.]				
			Cd	Cu	Ni	Pb	Zn
Eko	0.0	391.6	0.3	7.6	4.7	2.6	46.6
	7.5	627.9	0.5	11.7	7.0	3.0	51.6
	15.0	708.9	0.6	8.8	5.1	1.9	54.4
Roadside	0.0	432.6	0.2	9.5	4.3	3.3	47.3
	7.5	625.5	0.2	9.3	5.6	2.7	49.9
	15.0	773.1	0.5	9.7	5.6	1.8	55.9
Coefficients of variation, %		32.8	80.0	39.1	66.0	60.3	13.5
Standard deviation		188.8	0.4	3.7	3.6	1.6	6.9
Mean		574.8	0.5	9.4	5.4	2.6	51.0

The greatest variability of the content of heavy metals in plants showed cadmium (80.0%), and its concentrations ranged from 0.2 to 0.6 mg/kg d.m. in studied combinations. The highest level of Cd was found in the samples with the highest dose of the sludge in grass mixture Eko, while the lowest in Roadside without sewage sludge. Cadmium content increased in above ground parts of both grass mixtures together with increasing of applied sewage sludge dose. In turn, zinc showed the lowest factor of variation in plants (13.5%), and its contents ranged from 46.6 to 55.9 mg/kg d.m. (Table 3). One observed a decrease in the quantity of Pb in the above-ground parts of plants upon increasing dose of sewage sludge: 2.6 and 3.3 mg/kg d.m. at 0.0 kg/m^2 to 1.8 mg/kg d.m. at 15.0 kg/m^2 for both mixtures, respectively. It may indicate that the applied sludge causes the binding of Pb with various fractions of the soil, thus it is not taken in large quantities by plants and does not affect their development. According to Robinson et al. [25], uptake of most metals by plants depends on the plant variety and species as well as on concentrations of other metals in soil, the type and amount of applied fertilizers and microbial activity of rhizosphere. Each of these factors itself and combined with the others can stimulate as well as impede the uptake of minerals by the plants and thereby affect the composition of cultivated plants. A relatively high correlation between concentrations of Cu and Zn (r = 0.53), Cu and Cd (r = 0.65) and Zn and Cd (r = 0.57), $p \leq 0.05$ was found. Although plants remove little pollutants, it is important that they grow in contaminated soil, thereby reducing the bioavailability of pollutants [25]. In turn, Singh et al. [11] believe that in order to effectively clean the soil, the plants should be of a high resistance to difficult environmental conditions. Moreover, they need to have the ability to accumulate xenobiotics from soil and also to grow rapidly and to produce a large biomass. The grass mixtures used in the experiment show great ability to accumulate dry matter in unfavorable conditions. Together with the increased fertilization dry matter of above ground parts of plants proportionally increased, and at the highest applied sewage sludge dose the dry matter amounted 708.0 and 773.0 g d.m./m^2 for Eko and Roadside, respectively (Table 3).

The binding nature of heavy metals may be due to exchangeable adsorption or to the formation of complex coordination bonds [12]. Such elements as: Cu, Ni, Pb have the highest affinity to organic matter. Organic matter has the ability to create persistent complex bonds that may significantly restrict the mobility of metals. Together with the increased dose of sewage sludge on the experimental fields the concentrations of Pb and Zn in soil not significantly increased, in particular where grass mixture Eko was grown. The greatest variability of heavy metals in the soil showed lead, its contents in the soil ranged from 31.5 to 54.6 mg/kg d.m. and its coefficient of variation was 55.3%. In turn, Ni showed the smallest coefficient of variation in soil (5.6%) and its concentrations in the soil ranged from 43.3 to 47.4 mg/kg d.m. (Table 4). Antonkiewicz and Jasiewicz [13] argued that nickel belongs to the group of quite mobile and easily absorbed elements by plants, however, its movement to above-ground parts of plants is significantly impeded.

The analysis of the concentration of heavy metals in the studied soils showed that the concentrations of Cd, Cu, Ni, Pb and Zn (Table 4) are within the limit values for soils of the urban areas defined in the Regulation of the Ministry of Environment, September 9, 2002 on the standards of soil quality and standards of land quality [14].

The concentrations and distribution of heavy metals in soil profiles are determined by quantity of organic matter, soil properties and the soil-forming processes. The natural concentration of trace elements in the soil depends primarily on the type of the maternal rock, which is their original source [8]. The natural contents of metals for Bialystok were: Cd < 0.5 ppm, Cu 20–40 ppm, Ni < 5 ppm, Pb 12.5–100 ppm, Zn 25–400 ppm [15]. Cadmium (0.6–0.8 mg/kg d.m.) and nickel (43.3–47.4 mg/kg d.m.) in the studied soils were above the background geochemical cycle drawn up for Białystok. The concentrations of other studied metals were within the limits of the natural concentrations of soil. In addition, the biologically active rhizosphere plays an exceptionally important role in the geochemical circulation of elements. Roots and associated symbiotic fungi and autochtonic bacteria affect the chemical processes of the rhizosphere, contribute to the activation of ions of heavy metals and biomineralization of organic matter [16]. In our study, concentrations of Zn in the soil were significantly correlated with the concentrations of Pb ($r = 0.42$), Cu ($r = 0.92$) and Ni ($r = 0.42$), while the concentrations of Ni with the level of Cd and Cu ($r = 0.43$ and $r = 0.46$, respectively) at $p \leq 0.05$.

Table 4

Concentrations of heavy metals in soil (0–20 cm)
fertilized with different doses of sludge

Grass mixtures	Dose of sewage sludge [kg/m^2]	Soil [mg/kg d.m.]					pH (KCl)
		Cd	Cu	Ni	Pb	Zn	
Eko	0.0	0.8	14.9	43.3	31.5	68.6	7.5
	7.5	0.7	17.0	44.2	32.6	103.1	7.3
	15.0	0.8	17.0	44.4	54.6	104.6	7.2
Roadside	0.0	0.6	15.0	44.2	36.9	80.8	7.3
	7.5	0.8	16.7	45.1	32.9	80.6	7.3
	15.0	0.6	16.9	47.4	34.4	93.9	7.5
Coefficients of variation (%)		43.0	29.1	5.6	55.3	68.1	2.5
Standard deviation		0.3	4.7	2.5	20.5	60.4	0.2
Mean		0.7	16.2	44.8	37.1	88.6	7.3

In our studies, average values of the indicators for the accumulation of metals in the soil were arranged in the following series: Zn > Ni > Pb > Cu > Cd, what is confirmed by Shirivastava and Benerjee [17], who made similar series: Cr > Ni > Zn > Cu > Pb > Cd. Furthermore, according to Krzywy and Iżewska [18], the concentrations of the six most important heavy metals in sediments can also be arranged somewhat differently, namely by creating a series: Zn > Cr > Pb > Cu > Ni > Cd.

pH of soil is a very important factor deciding directly about the possibility of plant growth, nutrients and heavy metals uptake, direction of biological processes and the development of other forms of acidity [19]. Rosada drew similar conclusions [20] that pH of soil was an important determining factor of solubility of heavy metals and equilibrium of adsorption-desorption processes of hydrogen and metal cations. pH values measured at 1 M KCl for all samples tested ranged from 7.2 to 7.5 (the coefficient of variation = 2.5%) which means that in each studied point, reaction of the soil was slightly alkaline (Table 1), thus heavy metals present in the analyzed soil did not constitute a greater threat to the environment. Slightly alkaline reaction of the soil may be associated with the presence of debris-limestone additives and the deposition of alkali dust. Urban soils are usually formed from construction wastes, so that their structure is dense, concentrations of humic substances are lower, similarly as water capacity and permeability, and biological activity is weaker [25]. An important positive correlation was observed between pH of soil and the metal concentrations in the studied soils. pH was significantly correlated with Zn ($r = 0.65$), Cu ($r = 0.67$), Ni ($r = 0.63$) at $p \leq 0.05$.

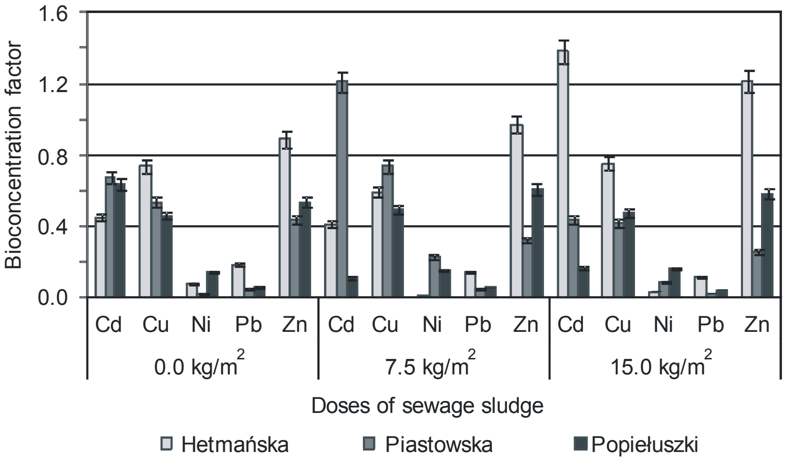

Fig. 1. Bioconcentration factor of heavy metals (Cd, Cu, Ni, Pb and Zn) in lawn grasses

The calculated bioconcentration factor (BCF) allowed determining phytotoxicity of individual metals. To a great extent the toxicity of heavy metals depends on the function performed in metabolic processes in living organisms and their possibility of bioaccumulation. The obtained BCF value allowed estimation of the capacity of the studied plants to absorb heavy metals from the soil. It also reflects the quantity and speed of movement of heavy metals from soil fertilized with various doses of sludge to the above-ground parts of plants (Fig. 1). Based on the obtained results, it was concluded that it was easier for the plants to retrieve Cd, Zn, Cu than Ni, Pb, which might indicate a high mobility of Cd, Zn, Cu in comparison with other metals and reflects the easy retrieval of metals by plants. As Bielińska states [21], Cd, Zn and Cu are metals very easily gathered in plant tissues, including themselves into the trophic chain and they may cause physiological and anatomical changes in plants.

Drążkiewicz et al. [22] argue that the bioaccumulation rate of various heavy metals, especially cadmium, is differentiated among plant species and among varieties. Although cadmium is not needed for the development of plants, it is absorbed extremely easily, both by root system and leaves, usually proportionally to its concentration in the environment [23].

A significant correlation exists between the metal concentrations in soils and the metal concentrations in above-ground parts of plants. Nickel in soil was correlated with Cd ($r = 0.48$), Zn ($r = 0.63$) and Cu ($r = 0.49$) in plants, while the concentrations of Zn in soil was significantly correlated with the level of Cu ($r = 0.66$) in plants. The elevated concentrations of Cd and Zn in plants, BCF 1.4 and 1.2, respectively, were recorded at Hetmańska Str. when the highest sewage sludge dose was used and Cd (BCF = 1.2) in Piastowska Str. with 7.5 kg/m^2 of sewage sludge applied. The accumulation of Pb and Ni in plants was the same independently on the applied fertilization sludge sewage (Fig. 1). It is important to understand the (bio)chemical processes in the vicinity of roots because they affect the transfer of metal ions to plants and other living organisms [25]. Heavy metals present in the sludge are strongly absorbed by the matrix biowaste, thus added to the soils are less available than added in the form of inorganic salts. In fact, sewage sludge added to soil can indeed serve as an absorber of metals from soil solution [9]. Such situation was probably in our study (Table 4).

Based on studies carried out by Kandziora et al. [24], it seems that one way of plant defense against heavy metals is synthesis of phytochelatins. Some heavy metals can influence the formation of phytochelatins, thereby increasing the efficiency of detoxification. Sulphydryl groups (–SH) in phytochelatins bind heavy metals, thereby reducing their negative impact on the plants. According to Robinson et al. [25], plants have developed defense mechanisms, which allow them to grow and develop in the polluted environment, e.g. restriction of transport across plasmalemma into the cell, production of heat-shock proteins that repair cellular damage or active effluex into the apoplast, etc.

4. CONCLUSIONS

1. After using of sewage sludge (7.5 and 15.0 kg/m^2) concentrations of Cd and Ni in the humic levels of soils were higher in comparison with their concentrations in maternal rock but the levels of these elements did not exceed the limit values, as referred to in the regulation of the Ministry of the Environment (2002).

2. The concentrations of all studied heavy metals (Cd, Cu, Ni, Pb and Zn) in the surface layer of the soil were correlated with their concentrations in grass biomass.

3. The application of 7.5 and 15.0 kg/m^2 of sewage sludge increased the biomass of above ground plant parts of both investigated grass mixtures grown along the roads.

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