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Research Article

FRACTIONATION OF SELECTED HEAVY METALS (Zn, Ni, Cu) IN MUNICIPAL SEWAGE SLUDGES FROM PODLASIE PROVINCE

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ABSTRACT

In the samples of fresh dehydrated sewage sludges from municipal treatment plants in Grajewo, Bielsk Podlaski, Sokółka, Dąbrowa Białostocka, Knyszyn, Mońki, Augustów, Suwałki, Sejny and Suchowola the following determinations were made: pH, pseudo-total content of Zn, Ni and Cu, organic matter and dry mass. The contents of the above-mentioned elements in fractions were evaluated using modified BCR method (four fractions: F1-acid soluble and exchangeable, F2-reducible, F3-oxidizable, F4-residual). The zinc content (mean values) in particular fractions can be arranged quantitatively in a sequence: F3 (60.8%) > F2 (20.0%) > F4 (19.9%) > F1 (3.9%), in the case of nickel: F3 (48.6%) > F2 (25.2%) > F4 (25.1%) > F1 (5.6%) and in the case of copper: F4 (66.3%) > F3 (35.3%) > F1 (4.2%) > F2 (3.0%). Cumulative content of zinc in mobile fractions (F1+F2+F3) ranged from 76.0 to 93.3%; for nickel from 56.3 to 89.6% and for copper from 28.8 to 53.3% of pseudo-total content.

Keywords: zinc, nickel, copper, metal fractions, sewage sludge

INTRODUCTION

The municipal sewage sludges, whose amount is constantly increasing, are appreciable source of organic matter and nutrients [Bień, Wystalska 2008], thus they can be useful for improving soil fertility. The presence of organic and inorganic contaminants in sewage sludge may, however, constitutes a danger to the environment. Heavy metal content is one of the major factors limiting the application of sewage sludge to agricultural lands. Their presence at high concentrations may be toxic to both plants and animals, including humans [Bozkurt et al. 2010].

Zinc and copper belong to a group of essential elements for plants, while nickel only for some plant species. Zinc uptake is often excessive (it is usually directly proportional to the concentration in the soil) what causes its toxicity [Łukowski 2006]. The disccused element is relatively little toxic for a human organism. The excess of Zn can cause poisoning, cancer and anemia, due to decrease of digestive tolerance of iron, phosphorus, copper and calcium [Dutkiewicz 1974]. Excessive amounts of copper in plant inhibit growth and impairs important cellular processes (e.g. photosynthetic electron transport) [Łukowski, Wiater 2009]. The excess of Cu in human body causes anemia, liver, capillary bed and kidney damage, diarrhea, pain, intestines shrink, circulatory and reproductive system disturbances, as well as growth disorders. Absorption of copper compounds by respiratory tract causes nasal congestion and gastritis [Michna et al. 1998]. Nickel is toxic for plants at high concentrations. It inhibits a large number of plant enzymes, such as those of nitrogen metabolism and sulphate assimilation [Łukowski, Wiater 2009]. The excess of nickel in human organism is accumulating in lymph nodes. It causes changes in bone marrow, chromosomes and nucleic acids stucture as well as eczema and cancer diseases [Dutkiewicz 1974].

The mobility of heavy metals, their bioavailability and related eco-toxicity to plants, depend strongly on their specific chemical forms or ways of binding. These are the parameters that have to be determined, rather than the total element contents, in order to assess toxic effects and to study geochemical pathways [Fuentes et al. 2004]. Metal fractionation in different chemical forms associated to the matrix sample requires the application of the sequential extraction methods [Pérez-Cid et al. 1999].

EXPERIMENTAL PROCEDURES

The study was based on fresh dehydrated sewage sludges collected in municipal treatment plants from Grajewo, Bielsk Podlaski, Sokółka, Dąbrowa Białostocka, Knyszyn, Mońki, Augustów, Suwałki, Sejny and Suchowola. The sludges were hygenized (mainly with burnt lime), except these from Sokółka, Augustów and Suchowola. Fractionation was made in average samples (three individual samples, dried at room temperature, were mixed and homogenized). In collected samples the following determinations were made: pH potentiometrically, dry mass by drying at 105 ºC, organic matter by heating in oven at 600 ºC until a constant weight was achieved, pseudo-total content of Zn, Ni and Cu was determined (after previous digestion in HNO_3 with 30% H_2O_2) by means of FAAS technique and content of studied metals in fractions by means of GFAAS technique using Varian AA-100 apparatus. The percentage of individual fractions in pseudo-total content of each element was caculated. Recovery was defined as a ratio of metal content in four fractions (F1, F2, F3 i F4) to pseudo-total content.

Modified BCR method with the usage of ultrasonic probe Sonics VCX 130 was used to evaluate fractional composition of Zn, Ni and Cu in sludge samples. Extraction included four stages:

1. Acid soluble and exchangeable fraction (F1) – 1g of sludge in 100cm^3 centrifuge tube with 40 cm^3 of 0.11 mol·dm^{-3} acetic acid was sonicated for 7 minutes (power – 20W) at temperature 22±5 ºC. Then, the mixture was centrifuged for 20 minutes at 3000g. Extract was separated for analysis. Residue with 20 cm^3 of deionized water was sonicated for 5 minutes (power – 20W) and centrifuged for 20 minutes at 3000g. Water was discarded.
2. Reducible fraction, bound to Fe/Mn oxides (F2) – to the residue from the first step was added 40 cm^3 of 0,5 mol·dm^{-3} hydroxylamine hydrochloride fresh solution, pH 1.5, and sonicated for 7 minutes (power – 20W) at temperature 22±5 ºC. Then the mixture was centrifuged for 20 minutes at 3000g. The extract was separated for analysis. The residue was rinsed with deionized water, like in the first step.
3. Oxidizable fraction, bound to organic matter (F3) – to the residue from the second step was added 20 cm^3 of 30% hydrogen peroxide and sonicated for 2 minutes (power – 20W) at temperature 22±5 ºC. Then, the volume of H_2O_2 reduced to approx. 1 cm^3 using water bath. To the moist residue was added 50 cm^3 of 1 mol·dm^{-3} ammonium acetate and sonicated for 6 min. (power – 20W) at temperature 22±5 ºC. Then, the mixture was centrifuged for 20 minutes at 3000g. Extract was separated for analysis. The residue was rinsed with deionized water, like in the previous steps.
4. Residual fraction (F4) – the residue from the third step was extracted using concentrated HNO_3 with addition of 30% H_2O_2.

RESULTS AND DISCUSSION

The studied sludges were characterized by alkaline reaction, what is favorable to immobilization of metals (Table 1). However, selected metals (Zn and Cu for example) tend to increase solubility also at alkaline reaction, what is connected with the ability to form anion complexes and bonds of metal ions with ammonium compounds or low molecule organic matter. The solubility of metal compounds in environment depends mostly on the reaction [Wilk, Gworek 2009].

The content of dry mass ranged from 6.6 to 43.5 %. The highest amounts of dry matter have contained mainly the lime-treated sludges.

The content of organic matter in the studied sludges ranged from 15.4 to 73.9 % DM. As stated by Mazur [1995], the content of organic matter in sewage sludge is in the range of 17 to 87% (60% on average). It has the ability to form stabile complexes, which can significantly decrease metal mobility.

The pseudo-total amounts of the studied elements (Table 2) were below values intended for agricultural use of sludge. The content of zinc, nickel and copper in all samples was lower than the average (1350, 30 and 147 mg·kg^{-1} DM, respectively) reported by Maćkowiak [2000], except for sludge from Sejny in the case of Cu.

Table 1. Physico-chemical characteristics of sewage sludges (mean ± SD, n=3) [Łukowski 2017]

Location	pH	DM	Organic matter
		%	
Dąbrowa Białostocka	9.43 ± 0.01	17.9 ± 0.23	29.8 ± 2.1
Grajewo	8.91 ± 0.22	43.5 ± 6.05	16.2 ± 3.6
Sokółka	8.20 ± 0.08	15.4 ± 5.03	62.5 ± 6.5
Bielsk Podlaski	8.14 ± 0.06	17.1 ±1.65	62.8 ± 13.4
Knyszyn	7.79 ± 0.34	6.6 ± 0.76	41.6 ± 18.6
Augustów	8.02 ± 0.05	13.3 ± 1.04	30.2 ± 7.9
Sejny	8.38 ± 0.07	11.1 ± 0.14	15.4 ± 4.0
Suwałki	9.03 ± 0.19	20.6 ± 0.92	36.5 ± 3.0
Mońki	9.42 ± 0.05	13.1 ± 0.63	29.8 ± 13.7
Suchowola	7.78 ± 0.16	10.6 ± 1.07	73.9 ± 5.1

Table 2. Pseudo-total content of Zn, Ni and Cu in sewage sludges (mean ± SD, n=3)

Location	Zn	Ni	Cu
	mg·kg^{-1} DM		
Dąbrowa Białostocka	1161 ± 22	17.15 ± 0.27	43.35 ± 1.13
Grajewo	686 ± 147	11.63 ± 0.50	35.41 ± 1.37
Sokółka	1202 ± 9	14.66 ± 0.84	88.92 ± 5.68
Bielsk Podlaski	1254 ± 56	11.45 ± 0.71	84.21 ± 9.10
Knyszyn	1302 ± 7	10.16 ± 0.27	67.81 ± 2.05
Augustów	1255 ± 8	13.68 ± 1.59	130.58 ± 7.92
Sejny	1302 ± 10	11.81 ± 0.55	209.34 ± 10.63
Suwałki	1295 ± 4	18.79 ± 0.84	107.85 ± 6.39
Mońki	1239 ± 5	13.92 ± 0.10	72.95 ± 5.44
Suchowola	1327 ± 5	12.68 ± 0.07	102.98 ± 3.28

Zinc

The highest amount of zinc (60.8% on average) was bound to oxidizable fraction and ranged from 45.9 to 80.1% (Table 3). Different results were noted by Fuentes et al. [Fuentes et al. 2004] investigating unstabilised sewage sludge from treatment plant located in the Region of Murcia (Spain) with BCR method. The authors stated in discussed fraction 38.0% of Zn as compared to the total content, the most among all the fractions. According to Wang et al. [Wang et al. 2008], organic matter of sewage sludge bound only approximately 1% of zinc.

Most soluble and thus bioavailable zinc (fraction F1) constituted only 3.9% of pseudo-total content. Metals corresponding to the exchangeable fraction usually represent a small portion of total metal content in sewage sludges [Filgueiras et al. 2002]. The low content of zinc in water-soluble and exchangeable fraction (l.0% of total content on average) have also stated Jakubus and Czekała [2001], according to Zeien and Brümmer method.

The average zinc content in fraction F2 amounted 20.0% of pseudo-total content. Similar result (21.6%) in the reducible fraction were stated by the above-mentioned Fuentes et al. [2004]. According to Pérez-Cid et al. [1999], this fraction of sewage sludge came from plant in Ourense (Spain) contained 40.8% of Zn. Other authors also reported that Fe/Mn oxides in sludges bind zinc strongly (even up to 60% of total zinc) [Fernández Alborés et al. 2000, Ščančar et al. 2001, Dąbrowska 2004.].

Residual fraction bound 19.9% of pseudo-total Zn content on average. Different result is presented by Gawdzik and Gawdzik [2012]. The authors studied the sludge from treatment plant in Cedzyna using BCR method. They have observed 50% of total zinc in this fraction. Even higher amount of zinc (approx. 80%) in residual fraction stated Wang et al. [2008].

Zinc content in mobile pool (F1+F2+F3) of studied sludges was similar. The most of zinc (93.3% of pseudo-total content) in mobile pool was noted in sludge coming from plant in

Table 3. Content, percentage in fractions and recovery of Zn in the sewage sludges

Location	F1	F2	F3	F4	F1	F2	F3	F4	Recovery
	mg·kg^{-1} DM				%				
Dąbrowa Białostocka	10.84	83.70	930.30	143.90	0.9	7.2	80.1	12.4	101
Grajewo	7.21	21.26	523.83	110.87	1.1	3.1	76.4	16.2	97
Sokółka	96.18	365.78	643.91	125.09	8.0	30.4	53.6	10.4	102
Bielsk Podlaski	62.51	321.10	756.49	156.92	5.0	25.6	60.3	12.5	103
Knyszyn	66.43	350.06	798.41	156.89	5.1	26.9	61.3	12.0	105
Augustów	59.65	236.91	674.87	356.23	4.8	18.9	53.8	28.4	106
Sejny	50.58	341.09	598.01	435.98	3.9	26.2	45.9	33.5	110
Suwałki	79.83	356.90	634.10	270.78	6.2	27.6	49.0	20.9	104
Mońki	4.25	120.85	861.31	359.09	0.3	9.8	69.5	29.0	109
Suchowola	48.74	324.81	768.93	321.00	3.7	24.5	57.9	24.2	110
	Average								
	48.62	252.24	719.02	243.68	3.9	20.0	60.8	19.9	105

Knyszyn. It was caused by high content of organic matter and simultaneously slight amount of minerals, which comprise residual fraction. The same factors decided about the lowest Zn content in mobile pool of sludge from Sejny. It contained the lowest amount of organic matter among all the sludges and the highest Zn percentage (33.5%) in residual fraction at the same time.

Nickel

Fraction F3 bound the highest amount of nickel (48.6% on average) and its content ranged from 28.0 to 59.8%, as compared to pseudo-total content (Table 4). Kazi et al. [2005] investigating fractional composition of heavy metals in sewage sludge coming from treatment plant in Hyderabad City (Pakistan) have stated similar content (41%) of Ni in discussed fraction using BCR method. The above-cited Gawdzik and Gawdzik [2012] have stated lower content (approx. 38%) of Ni in discussed fraction.

The lowest amount of studied element (5.6% on average) was observed in fraction F1. The higher content of nickel in exchangeable fraction (16.4% of total content) were stated by Fuentes et al. [2004]. Similar result (17%) was obtained by Wang et al. [2008] in acid soluble and exchangeable fraction of sewage sludge from plant in Loujiang (China). According to Wilk and Gworek [2009] nickel belongs to metals, which occur mostly in exchangeable fraction of sewage sludge and its amount is dependent on sorption and desorption processes.

Fraction bound to Fe/Mn oxides comprised 25.2% of psedo-total content on average. Smaller nickel content (approx. 12%) in this fraction was observed by Gawdzik and Gawdzik [2012] as well as by Wang et al. [2008] – approx. 15% of total content. Rosazlin et al. [2007], using modified Tessier method, have stated in reducible fraction of sludge came from treatment plant in Taman Sri Gombak (Malaysia) similar Ni amount (10% of total content).

Residual fraction of sludges bound 25.1% of pseudo-total nickel content on average. The above-cited Fuentes et al. [2004] were noted in disccused fraction 20.4% of total Ni. Shrivastava and Banerjee [2004] reported the highest Ni content (61.4% of total-content) in residual fraction among other fractions. These authors studied the sludge from treatment plant in Delhi (India) using modified Tessier method.

Mobile pool of nickel was the largest (89.6%) in sludge from plant in Grajewo. In the most of sludges the global content in three fractions was similar. The amount of metal in this pool was influenced by content of minerals. It was proved by the Ni percentage in residual fraction, which was the highest in sludges with the lowest mobile pool.

Copper

The highest percentage of copper (66.3% on average) was observed in residual fraction and ranged from 52.1 to 97.2% of pseudo-total content (Table 5). Very similar result (61%) is presented by the above-cited Shrivastava and Banerjee [2004]. According to García-Delgado et al. [2007] in residual fraction of sludge from 5 to 60% of total Cu content can be found.

Table 4. Content, percentage in fractions and recovery of Ni in the sewage sludges

Location	F1	F2	F3	F4	F1	F2	F3	F4	Recovery
	mg·kg^{-1} DM				%				
Dąbrowa Białostocka	1.29	3.57	4.80	8.13	7.5	20.8	28.0	47.4	104
Grajewo	0.25	3.23	6.95	1.45	2.1	27.8	59.7	12.5	102
Sokółka	0.96	3.72	5.30	6.03	6.6	25.4	36.2	41.1	109
Bielsk Podlaski	0.43	2.96	6.85	0.89	3.7	25.8	59.8	7.8	97
Knyszyn	0.35	2.56	5.85	1.81	3.4	25.2	57.5	17.8	104
Augustów	0.39	3.84	5.40	6.02	2.8	28.1	39.5	44.0	114
Sejny	0.87	3.00	5.90	3.29	7.4	25.4	50.0	27.9	111
Suwałki	1.51	4.60	8.45	6.02	8.0	24.5	45.0	32.0	109
Mońki	0.67	3.97	7.52	1.79	4.8	28.5	54.0	12.8	100
Suchowola	1.25	2.64	7.10	0.97	9.9	20.8	56.0	7.6	94
	Average								
	0.8	3.4	6.4	3.6	5.6	25.2	48.6	25.1	105

Table 5. Content, percentage in fractions and recovery of Cu in the sewage sludges

Location	F1	F2	F3	F4	F1	F2	F3	F4	Recovery
	mg·kg^{-1} DM				%				
Dąbrowa Białostocka	2.76	2.72	16.95	27.83	6.4	6.3	39.1	64.2	116
Grajewo	4.48	2.52	10.01	19.02	12.7	7.1	28.3	53.7	102
Sokółka	10.28	6.08	30.23	50.29	11.6	6.8	34.0	56.6	109
Bielsk Podlaski	1.32	0.80	22.11	63.29	1.6	1.0	26.3	75.2	104
Knyszyn	1.48	1.20	21.02	65.92	2.2	1.8	31.0	97.2	132
Augustów	1.32	1.72	39.68	88.19	1.0	1.3	30.4	67.5	100
Sejny	2.12	1.48	83.20	120.46	1.0	0.7	39.7	57.5	99
Suwałki	1.64	1.36	33.52	78.19	1.5	1.3	31.1	72.5	106
Mońki	2.44	2.20	34.21	38.02	3.3	3.0	46.9	52.1	105
Suchowola	1.08	0.80	47.19	68.29	1.0	0.8	45.8	66.3	114
	Average								
	2.9	2.1	33.8	61.9	4.2	3.0	35.3	66.3	109

Fraction F2 gathered the least of Cu (3% on average) among all fractions. Rosazlin et al. [2007] observed in discussed fraction the higher amount of copper – approx. 6%, while Pérez-Cid et al. [1999] only 1.2% of total content.

Fraction F3 comprised 35.3% of pseudo-total content on average. García-Delgado et al. [2007] stated in studied sludges from 5 to 25% of total copper content. Rosazlin et al. [2007] reported that Cu in sewage sludge occurs predominantly in the organic form. They stated 57% of copper in the discussed fraction what is consistent with its known affinity for organic matter ligands. The amount of metal forming complexes of varying stability with organic matter is dependent on its content and quality.

Fraction F1 gathered 4.2% of copper, as compared to pseudo-total content. This result confirms that only small amount of Cu occurs in soluble and exchangeable form [Fuentes et al. 2004, Wang et al. 2008, García-Delgado et al. 2007]. By contrast, Pueyo et al. [2003] investigating heavy metal mobility in sewage sludge by BCR method, obtained 65.5% of Cu in the discussed fraction, as compared to pseudo-total content.

The largest pool of mobile copper (53.2%) was found in sludge from treatment plant in Mońki. It was caused by the low content of minerals, what was confirmed by the amount of Cu (52.1%) in fraction F4 (the lowest among all sewage sludges) and simultaneously appreciable content of high molecular weight organic matter, which bind metals most strongly (fraction F3 bounded the largest Cu amount despite not the highest organic matter content in this sludge). The least of mobile copper (28.9%) was found in sludge from Bielsk Podlaski. It was connected to a high content of minerals (75.2% of Cu in fraction F4) and slight bonding strength of copper by organic matter, what was proved by the lowest Cu content in fraction F3 among studied sludges and one of the highest organic matter content at the same time.

CONCLUSIONS

1. All sludges met the standards related to agricultural use, according to heavy metal content.
2. The zinc content (mean values) in particular fractions can be arranged quantitatively in a sequence: F3 (60.8%) > F2 (20.0%) > F4 (19.9%) > F1 (3.9%), in the case of nickel: F3 (48.6%) > F2 (25.2%) > F4 (25.1%) > F1 (5.6%) and in the case of copper: F4 (66.3%) > F3 (35.3%) > F1 (4.2%) > F2 (3.0%).
3. Assuming the cumulative content of metal in mobile fractions (F1+F2+F3) as the solubility criterion, it can be stated that zinc was most soluble and copper the least soluble.

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REFERENCES

1. Bień J., Wystalska K. 2008. The problems of sewage sludge management. Inżynieria i Ochrona Środowiska, 11(1), 5–11.
2. Bozkurt M.A., Yarılgaç T., Yazıcı A. 2010. The use of sewage sludge as an organic matter source in apple trees. Polish Journal of Environmental Studies, 19(2), 267–274.
3. Dąbrowska L. 2004. Speciation of heavy metals in sewage sludge, in: Pathways of pollutants and migration strategies of their impact on the ecosystems. Komitet Inżynierii Środowiska, Lublin, 38–47.
4. Dutkiewicz T. 1974. Chemia toksykologiczna. Państwowy Zakład Wydawnictw Lekarskich, Warszawa.
5. Fernández Alborés A., Pérez Cid B., Fernández Gómez E., Falqué López E. 2000. Comparison between sequential extraction procedures and single extractions for metal partitioning in sewage sludge samples. Analyst, 125, 1353–1357.
6. Filgueiras A. V., Lavilla I., Bendicho C. 2002. Chemical sequential extraction for metal partitioning in environmental solid samples. Journal of Environmental Monitoring, 4(6), 823–857.
7. Fuentes A., Lloréns M., Sáez J., Soler A., Aguilar M.I., Ortuño J.F., Meseguer V.F. 2004. Simple and sequential extractions of heavy metals from different sewage sludges. Chemosphere, 54(8), 1039–1047.
8. García-Delgado M., Rodríguez-Cruz M.S., Lorenzo L.F., Arienzo M, Sánchez-Martín M.J. 2007. Seasonal and time variability of heavy metal content and of its chemical forms in sewage sludges from different wastewater treatment plants. Science of the Total Environment, 382(1), 82–92.
9. Gawdzik J., Gawdzik B. 2012. Mobility of heavy metals in municipal sewage sludge from different throughput sewage treatment plants. Polish Journal of Environmental Studies, 21(6), 1603–1611.
10. Jakubus M., Czekała J. 2001. Heavy metal speciation in sewage sludge. Polish Journal of Environmental Studies, 10(4), 245–250.
11. Kazi T.G., Jamali M.K., Kazi G.H., Arain M. B., Afridi H. I., Siddiqui A. 2005. Evaluating the mobility of toxic metals in untreated industrial wastewater sludge using a BCR sequential extraction procedure and a leaching test. Analytical and Bioanalytical Chemistry, 383(2), 297–304.
12. Łukowski A. 2006. The influence of mineral fertilization on heavy metal fraction contents in soil. Part I. Zinc. Polish Journal of Environmental Studies, 15(2A), 410–414.
13. Łukowski A. 2017. Fractionation of heavy metals (Pb, Cr and Cd) in municipal sewage sludges from Podlasie Province. Journal of Ecological Engineering, 18(1), 132–138.
14. Łukowski A., Wiater J. 2009. The influence of mineral fertilization on heavy metal fraction contents in soil. Part II. copper and nickel. Polish Journal of Environmental Studies, 18(4), 645–650.
15. Maćkowiak Cz. 2000. Skład chemiczny osadów ściekowych i odpadów przemysłu spożywczego o działaniu nawozowym. Nawozy i Nawożenie, 4(5), 131–143.
16. Mazur T. 1995. The condition and perspective of the organic matter balance of the cultivated soils. Zeszyty Problemowe Postępu Nauk Rolniczych, 421a, 267–276.
17. Michna W., Boguszewska M., Bykowski P. J. (Eds.). 1998. Raport z badań monitorowych nad jakością gleb, roślin, produktów rolniczych i spożywczych w 1997 roku. PIOŚ, Warszawa.
18. Pérez-Cid B., Lavilla I., Bendicho C. 1999. Application of microwave extraction for partitioning of heavy metals in sewage sludge. Analytica Chimica Acta, 378(1–3), 201–210.
19. Pueyo M., Sastre J., Hernández E., Vidal M., López-Sánchez J.F., Rauret G. 2003. Prediction of trace

element mobility in contaminated soils by sequential extraction. Journal of Environmental Quality, 32(6), 2054–2066.

20. Rosazlin A., Che Fauziah I., Rosenani A.B., Zauyah S. 2007. Domestic sewage sludge application to an acid tropical soil: Part III. Fractionation study of heavy metals in sewage sludge and soils applied with sewage sludge. Malaysian Journal of Soil Science, 11, 81–95.
21. Ščančar J., Milačič R., Stražar M., Burica O., Bukovec P. 2001. Environmentally safe sewage sludge disposal: the impact of liming on the behaviour of Cd, Cr, Cu, Fe, Mn, Ni, Pb, and Zn. Journal of Environmental Monitoring, 3(2), 226–231.
22. Shrivastava S.K., Banerjee D.K. 2004. Speciation of metals in sewage sludge and sludge-amended soils. Water, Air and Soil Pollution, 152, 219–232.
23. Usman A. R. A., Kuzyakov Y., Stahr K. 2004. Dynamics of organic C mineralization and the mobile fraction of heavy metals in a calcareous soil incubated with organic wastes. Water, Air and Soil Pollution, 158(1), 401–418.
24. Wang P.F., Zhang S.H., Wang C., Hou J., Guo P.C., Lin Z.P. 2008. Study of heavy metal in sewage sludge and in Chinese cabbage grown in soil amended with sewage sludge. African Journal of Biotechnology, 7(9), 1329–1334.
25. Wilk M., Gworek B. 2009. Metale ciężkie w osadach ściekowych. Ochrona Środowiska i Zasobów Naturalnych, 39, 40–59.