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Research Article

THE CONCENTRATION OF TRACE ELEMENTS IN SEWAGE SLUDGE FROM WASTEWATER TREATMENT PLANT IN GNIEWINO

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ABSTRACT

The sewage sludge originating from wastewater treatment plants (WWTP) serving rural areas is suggested for agricultural or natural usage. However, sewage sludge is beforehand subjected to the several pre-treatments, which involve stabilization, hygienisation and pre-composting. These methods mainly decrease the amount of organic substances and the presence of microorganisms, but hardly affect the concentrations of heavy metals. The advantages of using sludges as fertilizer for improving and sustaining soil fertility and crop production are numerous. The addition of sewage sludge to soils could affect the potential availability of heavy metals. Trace elements are distributed in the soil in various forms: solid phases, free ions in soil solution, soluble organic-mineral complexes, or adsorbed on colloidal particles. The most undesirable heavy metals in sewage sludge that are toxic for the living organisms include: cadmium, chromium, nickel, lead and mercury. In the study, the concentrations of trace elements (Pb, Cd, Cr, Hg, Ni, Zn, Al, As, Se, B, Ba, Br, Ca, Cu, Fe, Mn, Na, Ga, Li, Mo, Sr, Mg, K, Ru, Tl, V, U) were tested in the sewage sludge obtained from a WWTP serving rural areas (PE< 9 000). In each case, the tested sewage sludge was meeting the criteria of stabilization and was used for agriculture and land reclamation purpose. All the samples were collected in 2016 and subjected to microwave mineralization in a closed system in aqua regia. The total amounts of macro and microelements were determined with a spectrophotometer Coupled Plasma emission ICP-OES. It was found that the total concentrations of trace metals in all of sewage sludges are the same as the Polish regulation limit of pollutants for sludge to be used in agriculture. European legislation is less restrictive and permits higher contents of heavy metals in sludge used for agriculture than Asia. The trace elements (cadmium: 1.16 $mg \cdot kg^{-1}$/d.m. in thePolish sewage sludge, are much higher than those in the other countries. Copper and zinc were the most prevalent elements observed (111.28 $mg \cdot kg^{-1}$/d.m. and 282.94 $mg \cdot kg^{-1}$/d.m., respectively). The concentrations of copper in the Polish sewage sludge are much lower (49–130 $mg \cdot kg^{-1}$/d.m.) than european sewage sludge (522–562 $mg \cdot kg^{-1}$/d.m.). The two of the tested heavy metals (beryllium, bismuth) were under the detection limit, while gallium, molybdenum, thallium, vanadium and silver were detected in the concentrations lower than 0.005 $mg \cdot kg^{-1}$/d.m. According to the obtained results, in all the tested samples, the total amount of trace elements, did not exceed the limit values in sewage sludge for their use in agriculture and land reclamation.

Keywords: trace elements, heavy metals, sewage sludge, wastewater treatment plant

INTRODUCTION

Sewage sludge contains heavy metals which occur in the form of different chemical compounds. The presence of heavy metals in the sewage sludge and their high concentrations are caused by the share of industrial sewage in the overall mass of municipal sewage. Heavy metals come from domestic wastewater as well as surface runoffs. Most of the metals found in sewage which are considered to be harmful are retained in the sludge. Such elements include the microelements necessary for living organisms to grow and develop (that is copper and zinc) as well as toxic metals with wide operation range causing numerous diseases, including cadmium, lead, arsenic, mercury [Gawdzik 2012].

The possibility of releasing heavy metals originated from sewage sludge to the environment depends on the form in which they occur [Merrington et al. 2003]. The metals which occur in sewage sludge have different forms and can be absorbed by clay minerals, hydrated iron oxides and organic matter; they also appear in the form of inorganic compounds, including oxides, phosphates, carbonates, sulphates, sulphides [Morita, Tsuboi 2000]. The metals that occur in a soluble and exchangable form are released to the environment most easily (nickel, cadmium), whereas the metals bonded with carbonates, phosphates, sulphides and oxides of manganese, iron, chromium and zinc are less easily released to the environment. Trace elements that occur in sewage sludge are indispensable for plants. However when in excess, they pose a threat to both human health and plants. Depending on the place of origin, the sludge should undergo a qualitative assessment in order to find out a proper manner of its management. The sludge produced in municipal wastewater treatment plants are characterized by high moisture and the sewage sludge may pose a risk to the natural environment. European legislation (European directive 86/278/CEE) is less restrictive and allows higher content of heavy metals in the sludge to be used in agriculture [Babel, del Mundo Dacera 2006].

In the European community, over 30% of sewage sludge is used as an agricultural fertilizer in agriculture. For example in the UK, [Gove et al. 2002] reported the increasing use of enhanced treated sewage sludge in agriculture. In France, 60% of sewage sludge is used in land application and in Belgium 57% of sludge is utilized for agriculture [Maisonnave et al. 2002].

The sludge should be regularly tested for dynamics of its chemical composition change due to fluctuations in the level of heavy metals [Wilk, Gworek 2009]. The overall content of heavy metals in the sludge ranges from 0.5–2% of sludge.

This study aims at providing characteristics of trace metals occurring in the sewage sludge from a selected municipal wastewater treatment plant as well as determining the level of contamination with heavy metals. The content of trace metals in the sludge was evaluated in terms of the current regulations on the application of sewage sludge and the risk connected with releasing the sludge into the soil.

Acidification of soils increases solubility of heavy metals and their absorption by plants [Lothenbach et al. 1997, Zufiaurre et al. 1998].

MATERIALS AND METHOD

The material used for the tests was sewage sludge which was collected in 2016, in the period from January to September, from a municipal wastewater treatment plant with the capacity 8000 RLM, located in the Pomeranian Voivoidship. The sewage transported to the treatment plant originated from a separate sewer system. The sludge was taken fresh to polyethylene containers directly from a station of mechanical drainage after being pressed and centrifuged according to PN-EN ISO 5667–13:2004 standard. Older sludge was collected from a reed sludge drying bed, at 4 depths, in two different places of the bed, documented in such a way that the age could be reliably defined. The remaining sludge was stored and drained. The trace elements, including Pb, Cd, Cr, Hg, Ni, Zn, Al, As, Se, B, Ba, Br, Ca, Cu, Fe, Mn, Na, Ga, Li, Mo, Sr, Mg, K, Ru, Tl, V, U were marked in the tested samples. The presented results of chemical analyses constitute an average from three repetitions. The pH of the sewage sludge was determined immediately after collection, while the solid contents were determined by oven-drying at 105°C. Next, it was ground and homogenized in an agate mortar and screened through a sieve with mesh size equal to 150 micrometers. Afterwards, it was analyzed for the total concentration of a metal. Organic matter (OM) content of the dry mass (DM) was determined withloss by ignition, exposing the dried sludge for 1 h to a temperature of 550°C. Before the mineralization, 0.5 g of fine dust was collected, aqua regia was flooded (9 ml HCl and 3 ml HNO_3). A calibration curve was made using ICP-IV and ICP-VI ICP-VI multifunctional solutions from MERCK. The final result is given taking into account the uncertainty of the element [PN-EN ISO 11885: 2009]. The concentrations of heavy metals in the analyzed sewage sludge were marked with the use of the atomic emission spectometry with plasma excitation (ICP-OES), (Optima 8300 made by Perkin-Elmer)from the solutions obtained in the course of dissolving sludge samples by aqua regia. All experiments were replicated three times. The statistical results have been presented.

RESULTS AND DISCUSSION

The physicochemical properties of the analyzed sludge samples were determined on the basis of Regulation of the Minister of Environment of 6 February 2015 on municipal sewage sludge (Journal of Laws 2015 item. 257 (Table 1), [Regulation of the Minister of the Environment].

The source: Regulation of the Minister of the Environment of 6 February 2015 on municipal sewage sludge, Journal of Laws 2015 item. 257.

Table 2 presents the physicochemical properties of sludge obtained from WWTPs in Gniewino. Particular physicochemical properties are characterized by slight differences. The values of the trace elements had been shown in (mg·kg^{-1}/dry matter). The results of measurements of the elements content in sediment samples were compared with the reference values showing high compatibility of the determinations.

The average content of dry mass in the tested samples was 7.21% of d. m., loss on ignition of the sludge dry mass amouted to 78.78%, the remainder equalled 21.22%, whereas the water content ranged from 87.55 to 98.75%. The sewage sludge produced in a wastewater treatment plant was characterized by high moisture (percentage content of water at the level of 90% and more). High moisture of the sewage sludge is characteristic of small sewage treatment plants with the capacity of 400 m^3 per 24 hours. Ph reaction is the main factor which affects the solubility of metal compounds in the environment. Heavy metals are the natural trace elements found in sewage sludge. Strong acidification of soil can cause the release of heavy metals bonded with oxides of manganese, aluminum and iron as well as other minerals. Cadmium has the highest mobility and is mobilized at pH equal to 6.5. On the other hand the highest solubility is attributed to chromium and phosphorus [Alloway, Jackson 1991]. The pH reaction is a very important parameter describing the quality of the sewage sludge. The tested samples were characterized by the pH of 7.28. The reaction of waste substances is one of the most important physicochemical properties. For the purpose of agricultural utilization of the sludge at the stage of hygienization, it should be enriched by adding calcium. The organic substance of sewage sludge has a large influence on heavy metals bonding and depends on the technological processes and stabilization. The sludge coming from the analyzed wastewater treatment plant was characterized by a similar concentration of organic matter (92.80%). The average content of Kiejdahl nitrogen in the sludge ranged up to (818.02%). The content of calcium in the tested material amounted to 1.68% and was significantly lower than the one given for sewage sludge of other treatment plants. The share of potasium in the overall content was found, ranging from 16.58 to 98.04%, (tab. 2). In the case of magnesium, the average value of the metal content in the sludge was reported to be from 0.33 to 0.39%). Zinc in sewage sludge is strongly bonded by oxides of Mn and Fe (even up to 60%). This was confirmed by tests [Fernandez Albores et al. 2000, Perez-

Table 1. The permissible amount of heavy metals in sewage sludge collected from selective wastewater treatment plant

Metals	The heavy metal content in (mg·kg^{-1}/d.m.) solids of not more than		
	In the application of municipal sewage sludge:		
	in agriculture and land reclamation for agricultural purposes	the reclamation of land for non-agricultural	in adapting land to the specific needs resulting from waste management plans, land use plans or zoning and land, to the cultivation of plants for the production of compost, for the cultivation of crops not intended for human consumption and animal feed
Zinc	2500	3500	5000
Lead	750	1000	1500
Cadmium	20	25	50
Chromium	500	1000	2500
Copper	1000	1200	2000
Nickel	300	400	500

Table 2. The concentration of heavy metals in sewage sludge

No.	Total content/ (mg·kg^{-1}/d.m.)	Average	Standard deviation	Mediana	Minimum	Maximum
1.	Potassium	68.14	37.62	87.31	16.58	98.04
2.	Iron	3.53	0.69	3.46	2.83	4.5
3.	Calcium*	1.68	0.38	1.81	1.14	2.05
4.	Magnesium*	0.37	0.02	0.37	0.33	0.39
5.	Lead	23.92	2.24	24.66	21.15	26.37
6.	Cadmium	1.16	0.56	1.30	0.49	1.66
7.	Nickel	53.85	37.57	45.82	37.27	98.36
8.	Zinc	282.94	75.79	30.68	282.94	353.61
9.	Copper	113.28	43.02	105.77	51.12	173.34
10.	Chromium overall	15.32	3.26	15.72	11.27	19.89
11.	Arsenic	0.14	0.01	0.13	0.13	0.16
12..	Borom	3.33	0.71	3.67	2.24	3.99
13.	Selenium	0.15	0.02	0.15	0.12	0.18
14.	Silver	0.00	0.00	0.00	0.00	0.00
15.	Aluminium	15.64	3.20	16.16	10.27	19.05
16.	Bar	0.13	0.05	0.13	0.08	0.19
17.	Gal	0.01	0.00	0.01	0.01	0.01
18.	Lithium	17.93	1.48	18.56	15.04	18.91
19.	Manganese	211.30	46.09	225.45	153.01	263.87
20.	Molybdenium	0.00	0.00	0.01	0.01	0.01
21.	Sodium	0.16	0.10	0.16	0.00	0.33
22	Rubidium	0,01	0,00	0.01	0.01	0.02
23.	Strontium	30.85	31.84	17.95	11.7	94.18
24.	Thallium	0.00	0.00	0.00	0.00	0.00
25.	Uranium	3.03	7.09	0.16	0.06	17.5
26.	Vanadium	0.01	0.00	0.00	0.00	0.01

*Calcium, magnesium (%).

Cid et al. 1996, Scancar et al. 2000]. Investigations carried out in China show that nearly 25% of zinc creates a bond with organic matter and carbonates [Qiao 1996]. Zinc concentration was found in China (1754.8-7026.66 mg·kg^{-1}), [Chao Wang et al. 2005, Dai Jia Yin et al. 2006]. Zinc was chosen to be an index element, as its concentration in soil and sewage sludge is usually higher than that of Hg, Cu or Cd. It is believed that the adjustment of zinc amount can not only prevent from harmful influence of Zn on cultivations but also prevent the overload of other heavy metals in the sewage sludge. On average, 1.45% of zinc was marked in the analyzed material which was a significantly higher value than the average content of the element in sewage sludge in Poland [Bojakowska et. al. 2012]. The content of zinc in the Polish sewage sludge ranges from 83 to 5124 mg·kg^{-1}). According to the results of long term research, most of the sludge contain from 1000 to 2000 mg·kg^{-1}of zinc. The content of zinc in the sludge originating from the analyzed wastewater treatment plant was in the range 282.94–484.00 mg·kg^{-1}), staying within the limit equal to 2500 for an element to be allowed in agricultural application. The average zinc content was found (305.68%). No exceeding of zinc content was reported for the analyzed sludge. Sewage sludge from 29 municipal waste water treatment plants situated all over Poland revealed values such as: (1504 mg·kg^{-1}), [Maćkowiak 1999]. Five times less zinc was reported in relation to the permissible amounts included in Regulations (543–655 mg·kg^{-1}), [Rajmund et al., Ignatowicz et. al 2011].

The maximum content of copper in the sludge used for agricultural purposes is 500 mg·kg^{-1}. Copper concentration in the sludge from the studied sewage treatment plant ranged from 107.69 to 160.36 mg·kg^{-1}. These samples of sludge showed a natural content of the element (tab. 2), no exceeding was found.

The sludge samples examined in other European countries were characterized by higher concentration of copper than in Poland (table 3), whereas its highest concentration was found in Great Britain (562 mg·kg^{-1}), followed by that in

Table 3. The content of heavy metals in sludge in different countries (mg·kg^{-1}/d.m.)

No.	Country	Pb	Cu	Zn	Cd	Cr	Ni
1.	Unitet Kingdom	221.5	562	778	3.5	159.5	58.5
2.	Germany	67.7	275	834	1.5	50	23.3
3.	France	119.9	322	837	4.1	69.4	35.5
4.	Sweden	48.2	522	620.5	1.5	38.4	19.3
5.	Poland	211.8	237.5	3641	9.93	144.2	41.1
6	China	69.33	1467.13	7026.66	36.81	210.35	214.29
7.	Slovenia	128	436	2049	2.78	841	622
8.	Japan	53	255	979	2.3	69	40
9.	Hawaii	---	373	819	5.9	---	36.68

[Kumazawa 1997, Li Hua et al 2008, N.V.Hue, Subasinghe A. Ranjith]

Sweden (522 mg·kg^{-1}). However, in China the content of copper was 523.9 mg·kg^{-1}. The content of copper in the sludge from Podkarpackie Voivoidship ranged from (49–149 mg·kg^{-1}), whereas in Siedleckie Voivoidship the concentration was found to be low: (70–80 mg·kg^{-1}). The most toxic heavy metals occurring in the sewage sludge include cadmium, lead, arsenic and mercury. Cadmium is a cancerogenic, toxic and teratogenic element. Contamination with cadmium is significantly higher in Poland than in other European countries, as presented in table 3. The average content of the metal in the Polish sewage sludge is 9.93 mg·kg^{-1}. For the analyzed treatment plant, the occurrence of this element was found to range from 0.49 to 1.66 mg·kg^{-1}, similarly as in Białystok (1.25 mg·kg^{-1}), [Wiater et al. 2014] and Kujawsko-Pomorske Voivoidship (0.77–1.96 mg·kg^{-1}), [Milik et al. 2016]. Although the mobility of lead in soil is small, it is highly toxic for the environment and all the compounds of lead are poisonous. The content of lead in the analyzed samples did not exceed the permissible content of lead for the sludge to be used for agricultural purposes (500 mg·kg^{-1}). In the tested samples, the concentration of lead was found to range from 21.15 to 26.37 mg·kg^{-1}. The content of lead in China was found to equal 28.4 mg·kg^{-1}. As far as arsenic is concerned, its mean value was 0.14 mg·kg^{-1}. No traces of mercury were detected in the examined samples. Heavy metals can prevent iron metabolism, whereas iron can prevent from absorption and transportation of other components, e.g. phosphorus. The carried out tests of the sewage sludge indicate that the content of iron was found at the level of 3.53 mg·kg^{-1}. The content of aluminum was similar for the tested samples (1.02–1.90 mg·kg^{-1}), similarly as in the case of lithium (15.04–18.91 mg·kg^{-1}). The maximum content of chromium for the sludge to be used in agriculture is 500 mg·kg^{-1}. The concentration of total chromium in the sewage sludge studied were generally low (11.27–33.55 mg·kg^{-1}). It was comparable with the results obtained in different facilities in Poland (40.00 mg·kg^{-1}), [Gondek 2006, Fijałkowski et al. 2009]. The share of sodium was similar. The sewage sludge taken from 4 depths contained similar amounts of the element equal to 0.16–0.17%, in the stored sludge (0.33%), whereas in the drained sludge no sodium was found. The biggest amount of nickel was observed in the sewage sludge taken from surface layers (95.82 to 98.36 mg·kg^{-1}), the remaining sludge included relatively lower amounts of the element, from 37.27 to 48.35 mg·kg^{-1}. No presence nickel was found in the drained sludge. The content of nickel in the sludge collected from many different sewage treatment plants in Poland was at the level from 2.2 to 358 mg·kg^{-1}, [Ilba et al. 2014]. Heavy contamination with nickel was observed in the sewage sludge from the southern part of Poland (1171 mg·kg^{-1}), [Siebielec et al. 2008] whereas the content of nickel found in the municipal sewage was higher in the industrial areas.

The highest concentration of nickel was found in Slovenia (622 mg·kg^{-1}) and China (214.29 mg·kg^{-1}), followed by that in United Kingdom (58.5 mg·kg^{-1}), Poland (41.1 mg·kg^{-1}), and Japan (40 mg·kg^{-1}), (table 3), [Kumazawa 1997, Li Hua et al 2008].

The average content of barium was found to equal 0.13 mg·kg^{-1}. The concentration of total strontium in the sewage sludge studies were generally low (11.70–94.18 mg·kg^{-1}). Sewage sludge in the study areas contained between 0.058 and 0.264 mg·kg^{-1} of total uranium, whereas the content of Boron was in the range: 2.24–3.99 mg·kg^{-1}.

The content of selenium ranged from trace amounts to (0.116–0.184 mg·kg^{-1}).

The total Cd, Pb, Zn, Cu, Ni and Cr contents, as well as the control standards for pollutants in sludge for agricultural use in other countries for minixcipal WWTP, are listed in (Table 4).

CONCLUSIONS

The carried out tests of the quality of sewage sludge collected from a selected wastewater treatment plant have revelaed that the content of all heavy metals did not exceed the permissible level specified in the Regulations of the Minister of Agriculture (tab. 1), established for sewage sludge in Poland. There was no seasonal variation in the individual heavy metals. The annual average content of heavy metals during the relevant period was maintained at a similar level, with very small differences between the samples, which is presented in table 2. Trace amounts of heavy metals, including gallium, molybdenum, rubidium, thallium, vanadium and silver were detected. The examined material did not contain beryllium or bismuth. Two of the examined heavy metals (beryllium and bismuth) were near the limit of detectability. The trace amounts of such elements as gallium, molybdenum, thallium, vanadium and silver were detected in theconcentrations lower than 0.005 mg·kg^{-1}. It has been shown that the sludge produced in the study treatment did not exceed the permissible concentrations of heavy metals specified for sewage sludge used for agricultural purposes in any case. Most of the examined sludge was characterized by the natural content of trace elements. The sludge can be utilized thermally, by methods that involve burning the sludge with more than 90% content of dry matter.

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Table 4. Total contents of Cd, Pb, Zn, Cu, Ni and Cr in sewage sludge collected from selective wastewater treatment plants, and control standards for pollutants in sludges for agricultural use in other countries (mg·kg^{-1}/d.m.)

Total content (mg·kg^{-1})	Location						
	Austria	Switzerland	United States	Germany	France	Spain	Holland
Cd	10	30	10–20	30	15	2.78	10
Pb	500	1000	300	1200	300	126	500
Zn	2000	3000	2000–2800	3000	3000	2032	2000
Cu	500	1000	1000–1500	1200	1500	433	500
Ni	200	200	200–420	200	100	621	50
Cr	500	1000	1000–1200	1200	200	856	500

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