Environmental Science and Pollution Research (2018) 25:31101–31112
https://doi.org/10.1007/s11356-018-3109-5

RESEARCH ARTICLE

The effect of cellulose production waste and municipal sewage sludge on biomass and heavy metal uptake by a plant mixture

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Received: 16 June 2018 / Accepted: 29 August 2018 / Published online: 5 September 2018

Abstract

Environmental management of cellulose production waste and municipal sewage sludge appears to be substantiated due to various physicochemical properties of these wastes. The aim of the conducted research was to determine the effect of cellulose production waste and sewage sludge on yielding and heavy metal uptake by a plant mixture. The research was conducted under field experiment conditions, determining the fertilizer value of these wastes in the environmental aspect. The research was carried out in the years 2013–2016. Species composition of the plant mixture was adjusted to habitat conditions. It was established that, as compared with the cellulose production waste, the municipal sewage sludge used in the experiment had a higher content of macroelements. The content of heavy metals in the studied waste did not exceed the limits that condition their use in agriculture and reclamation. Applying only the cellulose production waste did not significantly decrease the yield of the plants. Municipal sewage sludge showed the highest yield-forming effect. Mixing the above-mentioned wastes and their application to soil had a significant effect on the increase in the plant mixture yield. The waste applied to soil also increased the content of Cr, Cd, Pb, Cu, and Zn in the plant mix. The level of heavy metal content in the plant mix did not exclude this biomass from being used for fodder or reclamation purposes. The cellulose production waste and municipal sewage sludge increased the heavy metal uptake by the plant mixture. The plant biomass extracted heavy metals from the sewage sludge more intensively than from the cellulose production waste. Among the analyzed heavy metals, the highest phytoremediation was recorded for Ni (30%), followed by Cd (20%), Cr (15%), Pb (10%), and the lowest for Cu (9%) and Zn (8%). Application of the cellulose production waste and sewage sludge to soil also increased the content of the studied heavy metals in soil. However, it did not cause deterioration of soil quality standards. Heterogeneity in the chemical composition of the wastes confirms that each batch intended to be used for environmental management should be subjected to chemical control.

Keywords Cellulose production waste · Municipal sewage sludge · Plant mixture · Heavy metals · Phytoremediation

Introduction

It is increasingly common to undertake innovative studies on different forms of managing waste containing calcium and sodium compounds for elimination of SO_2 from exhaust gases (Tunali and Ertunç 2017). Studies are also being conducted on generating new materials and products from waste containing calcium compounds (Couret et al. 2017; Mymrin et al. 2018). There is little research on environmental management of industrial waste, composed in a way so that calcium compounds and other environmentally useful components could be used (Das and Singh 2004).

Cellulose production waste contains a lot of valuable components, including organic compounds as well as macro- and microelements essential to plants (Das and Singh 2004). The composition of cellulose production waste includes calcium and magnesium, which can occur in carbonate, oxide, and silicate form. Cellulose production waste contains small quantities of calcium and alkaline metal hydroxides as well as aluminosilicates (Kac-Kacas and Drzas 1968; Pérez-López

Responsible editor: Hailong Wang

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et al. 2008). Environmental management of cellulose production waste could improve soil physicochemical properties and create beneficial conditions for plant growth and development. However, on account of specific physicochemical properties, environmental use of cellulose production waste might be reduced. Cellulose production waste is characterized by a substantial content of 'undecomposed' mineral substances and organic compounds that can exclude their environmental management (Harasimowicz-Hermann and Hermann 2007a, 2007b; Das and Singh 2004). Difficulties with managing cellulose production waste result also from a high heavy metal content. The amount of heavy metals and other pollutants in cellulose production waste depends mainly on the type of the used raw material and technological processes (Wan Ngah and Hanafiah 2008). The analyzed cellulose production waste is 'generated' at a factory running a production business focused mainly on waste recycling processes. In the production process, old newspapers and multi-material packagings are used. The final product is 'towel' paper and building board, and the side product is sewage sludge in the form of cellulose production waste, which can be managed as substrate for decorative plants (Decision of MARD 2017).

Another significant waste in terms of fertilizer value is municipal sewage sludge, rich in organic matter and fertilizer components (Deeks et al. 2013). Municipal sewage sludge is also a potential source of heavy metals which, when introduced to the environment, pose a threat and are difficult to remove or detoxify (Corrêa Martins et al. 2016). Phytoextraction is the optimal way to remove these metals from sewage sludge. However, this process is very long and hardly efficient (Antonkiewicz et al. 2018; Nissim et al. 2018). Currently, disposal of organic waste containing substantial amounts of biodegradable substances is being reduced, hence the need for alternative management of this waste in the environment (Waste Act 2012; Directive 99/31/EC 1999). However, in the light of new regulations, there are stricter and stricter requirements determining the natural management of organic and mineral waste (Waste Act 2012; Regulation 2015).

The research undertaken concerns the assessment of the impact of cellulose production waste and municipal sewage sludge, as well as a well-composed mixture of the two, on the soil environment and on plants. The issue of heavy metal content in the above-mentioned wastes, and above all the assessment of the degree of their uptake by plants, is an important part of the assessment of threats that might be expected when using this kind of waste in the environment (Corrêa Martins et al. 2016; Nissim et al. 2018). This study makes an interesting attempt to point to a possibility of reducing the risk within the environment, posed by introduction of waste with different, mutually supplementary physicochemical properties. The following research hypothesis has been formulated: can the heavy metals present in cellulose production waste and municipal sewage sludge be immobilized and then taken up by plants grown on a well-composed waste mixture? Assessment of the possibility of environmental management of these wastes was conducted based on the plant mixture yield, content, and uptake of heavy metals by plants, and on the balance of these metals in the environment.

Material and methods

The research on the effect of cellulose production waste and municipal sewage sludge on the plant mixture was conducted in the years 2013–2016 on an experimental plot of the University of Agriculture in Krakow [50°05′03.7″N 19°51′13.5″E] located in the southern part of Poland.

The soil

The experiment was set up on arable soil with the granulometric composition of sandy loam (Table 1) (Polish Soil Classification 2011; Soil Survey Staff 2014). The soil was classified to the category of light soils, and according to the reaction measured in 1 mol KCl to slightly acid soils (Ostrowska et al. 1991).

The content of heavy metals in the soil on which the experiment was set up was below the permissible content of these elements in soils of agricultural lands (Table 1) (Regulation 2016). According to the Regulation of the Minister of Environment on municipal sewage sludge (Regulation 2015), total heavy metal content in the soil did not exceed acceptable values for using municipal sewage sludge in agriculture or for reclamation. Basing on the six-degree pollution scale developed by IUNG-PIB in Puławy, it was established that the soil had a natural (0°) content of Cr, Ni, Cu, Zn, Cd, and Pb (Kabata-Pendias et al. 1993, 1995).

Cellulose production waste and municipal sewage sludge

Cellulose production waste and municipal sewage sludge ('MSS') were used in the field experiment. They were analyzed for physicochemical properties (Table 1). Cellulose production waste ('CPW') contained approximately 12% calcium, which confirms that this waste has alkaline and deacidifying nature. Total content of N, P, K, and Na in the cellulose production waste was lower compared to the quantities occurring in the municipal sewage sludge (Table 1). The cellulose production waste had a higher content of heavy metals compared to the soil and municipal sewage sludge.

The municipal sewage sludge ('MSS') that was used in the field experiment was obtained from a mechanical-biological sewage treatment plant. Reaction of the municipal sewage

Table 1 Selected physical and chemical properties of the soil and wastes before experiment establishment

Parameter	Unit	Soil	Cellulose production waste CPW	Municipal sewage sludge MSS
Waste code number			030310[a]	190805[a]
Dry matter (D.M.)	%	Not determined	27.8	23.3
pH_{H2O}		7.02	7.38	7.12
pH_{KCl}		6.52	7.47	6.98
C organic carbon	g kg^{-1} D.M.	20.24	Not determined	269.0
Total nitrogen (N)		0.94	0.15	13.70
Total phosphorus (P)		Not determined	1.13	2.80
Total potassium (K)		Not determined	1.44	1.55
Total sodium (Na)		Not determined	0.59	1.23
Total magnesium (Mg)		Not determined	2.88	2.80
Total calcium (Ca)		Not determined	119.58	273.0
Total chromium (Cr)	mg kg^{-1} D.M.	6.35	47.29	7.59
Total nickel (Ni)		6.46	16.62	2.70
Total copper (Cu)		6.85	132.68	35.5
Total zinc (Zn)		58.83	257.99	426.0
Total cadmium (Cd)		0.68	0.87	0.545
Total lead (Pb)		17.52	33.49	10.2

[a] Waste Catalogue 2014

sludge was neutral, pH_{H2O} value amounted to 7.12, which confirms that the municipal sewage sludge was hygienized with lime. According to the Regulation of the Minister of Environment on municipal sewage sludge (Regulation 2015), content of heavy metals did not exceed permissible values conditioning their use in agriculture or for reclamation. There were also no microbiological contaminants found in the sewage.

Scheme and conditions of the experiment

The one-factor field experiment was set up in 2013 in a randomized block design, in four replications. The plot area was 6 m^2. The distance between the plots was 0.5 m. The experimental design comprised five treatments differing in the dose of the introduced wastes and their mixtures (Table 2). The applied waste doses (50 and 100 Mg ha^{-1} DM) were supposed to be suitable for substrates used in reclamation and revitalization of post-industrial areas (Kicińska et al. 2018). The manner of obtaining the mixture of cellulose production waste and municipal sewage sludge was reserved at the Patent Office of the Republic of Poland (Patent PL411157-A1 et al. 2015). The wastes were applied only once in spring of 2013 on the soil surface, then they were mixed with a 0–20 cm soil layer, 2 weeks before sowing the plant mixture.

The mixture that was sown in the field experiment consisted of seeds of the following grasses and papilionaceous plants: *Dactylis glomerata*—cock's-foot var. 'Amera' (10%); *Festuca pratensis*—meadow fescue var. 'Mewa' (10%); *Lolium pratense*—perennial ryegrass var. 'Solen' (10%) and 'Maradowna' (5%); *Lolium multiflorum*—annual ryegrass var. 'Mowester' (5%); *Poa pratensis*—common meadow grass var. 'Balin' (15%); *Trifolium resupinatum*—white clover var. 'Crasslands Haifa' (20%); *Festuca arundinacea*—tall fescue var. 'Fawn' (5%); *Festuca rubra*—red fescue var. 'Referent' (20%). Species composition of the plant mixture was adjusted to habitat conditions. The plant species chosen have a great capacity to adapt to unfavorable substrate conditions. These species are resistant to water deficit, they form sod well and yield well. Ryegrass germinates quickly; it is characterized by fast development and sod formation. Common meadow grass and fescues are characterized by good growth and development in the later period and in

Table 2 The experimental design and doses of the introduced wastes and their mixtures

No	Treatment	Waste doses [Mg D.M. ha^1]	
		Cellulose production waste (CPW)	Municipal sewage sludge (MSS)
1	Control (C)	0	0
2	Cellulose production waste CPW	50	0
3	Municipal sewage sludge MSS	0	50
4	Mixture I CPW × MSS	25	25
5	Mixture II CPW × MSS	50	50

successive years of vegetation. The mixture of seeds (plants) was sown manually in the third decade of May 2013. The amount of the mixture sown was 30 kg ha^{-1}. The rate of seeding of the mixture was increased due to possible adverse climatic-soil conditions. Initially, on days when air temperature was above 25 °C, the sown mixture was sprinkled with water as the ground was drying up at the depth of 2–4 cm. Emergences were observed 2 weeks after sowing. Mineral fertilization (NPK) was not used in the field experiment, which was explained by saying that the used wastes would be a source of nutrients for sown plant mixture.

Harvesting the plant mixture

In the next years of vegetation (2013–2016), the plant mixture was mowed four times. The first mowing took place at the stage of earing of the dominant grass species, and the other mowings after 6–8 weeks. The date of the plant mixture mowings also depended on weather conditions, including insolation and the amount of precipitation. Each year, on the day of the last harvest of the plant mixture, samples were collected from each plot from topsoil (0–20 cm layer), in order to conduct chemical analyses.

Determination of yield and the heavy metal content in the plants and soil

The plant material

The collected fresh plant material was dried in a forced air circulation dryer at 105 °C until reaching constant weight. The dried plant material was weighed and the amount of the plant mixture yield from each plot was determined. The amount of yield was determined by calculating the obtained yield from a plot per 1 ha. Samples (approx. 20 g DM) of plant material were collected from each plot (each repetition) for chemical analysis. The plant material was incinerated at the temperature of 550 °C for 8 h and the ash was digested in 65% HNO_3 (1:2 *v*/*v*) (Ostrowska et al. 1991).

The soil and waste materials

Basic chemical properties were determined in the topsoil, cellulose production waste, and sewage sludge of the field experiment, using established methods used in chemical-agricultural stations: reaction (pH)—determined in 1 mol dm^{-3} KCl solution, organic carbon—by Tiurin method, total nitrogen—by Kjeldahl method (Ostrowska et al. 1991; Jones and Case 1990). Total content of heavy metals in the soil and wastes was determined after incineration of organic matter and digesting in a mixture concentrated $HClO_4$ (70%) and HNO_3 (65%) acids (3:2 *v*/*v*) (Jones and Case 1990).

The content of heavy metals (Cr, Ni, Cu, Zn, Cd, Pb) in the obtained plant and soil filtrates was determined using a Perkin–Elmer Optima 7300 DV spectrometer (Jones and Case 1990). Spectrally pure reagents and Aldrich standard solutions were used in chemical analyses.

Quality control of analyses

Determinations in each of the analyzed samples were carried out in three replications. Accuracy of the analytical methods was verified based on certified reference materials and standard solutions: CRM IAEA/V–10 Hay (International Atomic Energy Agency), CRM–CD281–Rey Grass (Institute for Reference Materials and Measurements), CRM023–050–Trace Metals–Sandy Loam 7 (RT Corporation).

Computations and statistical analysis of the results

Due to the changeability of conditions in individual years, heavy metal content in the total plant yield is presented as a weighted mean. Heavy metal uptake (P) was calculated as the product of dry matter yield (A) and the nutrient content (X), according to the formula: $p = A \cdot X$. Heavy metal uptake was calculated every year and is presented as the total from the entire research period (2013–2016). Heavy metal balance was calculated from the difference between the amount of metals introduced with the dose of wastes and the amount of metals taken up with the plant yield. Phytoremediation of heavy metals presented in the balance is the percentage share of the uptake of heavy metals in relation to the amounts introduced to the soil along with the wastes.

The statistical analysis of the research results was conducted using a Microsoft Office Excel 2003 spreadsheet and Statistica package version 10 PL. The statistical evaluation of result variability was conducted using single-factor analysis of variance. The significance of differences among mean values was verified using *t* Tukey test at a significance level of $\alpha \leq 0.05$. For selected parameters (ratios), the value of Pearson's linear correlation coefficient (*r*) was computed, at a significance level of $\alpha \leq 0.05$. The maximum 5% dispersion between measurements in chemical analysis was adopted.

Research results

Crop yield

Dry matter yield from the plant mixture obtained in subsequent years of the field experiment is presented as the sum of all cuts collected each year and the mean of the whole research period, of vegetation years 2013–2016 (Table 3). A significant effect of cellulose production waste and sewage sludge and their mixtures on the yield of the plant mixture was observed in the field experiment (Table 3). The yield of the plant mixture dry matter, depending on the treatment and research year, varied within the following range: 3.34–8.15 Mg DM ha^{-1} (Table 3). The highest diversity in yielding was observed in the first research year, where the coefficient of variation amounted to approximately 14%, and the lowest—in 2015 (V = 12%). Observations and statistical analysis show that the diversity in yielding of the plant mixture in individual research years remained at a relatively constant level.

When analyzing the mean yield from the research period (2013–2016), it was established that the highest yield was obtained in treatment 4, wherein the mixture of cellulose production waste and sewage sludge in a single dose was applied. Increase in the plant mixture yield in that treatment was more than 28% in relation to the control. A double dose of the mixture of cellulose production waste and municipal sewage sludge (treatment 5) also increased the plant mixture yield considerably compared to the control. Increase in the plant mixture yield in that treatment was more than 25% compared to the control.

Applying only the municipal sewage sludge (treatment 3) also significantly increased the plant mixture yield by over 23% compared to the control. The increased plant mixture yield can be explained by a substantial load of macroelements that were present in the sludge as well as by the improvement of soil physicochemical properties. Applying only the cellulose production waste (treatment 2) decreased the yield of the plant mixture. However, the decrease in yield was not statistically significant.

The analyzed fertilization treatments can be arranged according to decreasing yield of the plant mixture in the following order: mixture of cellulose production waste and sewage sludge > sewage sludge > control soil > cellulose production waste. Analysis of the plant mixture yield in individual research years showed that the lowest yield was recorded in the first year of vegetation, which resulted (among other things) from a long emergence period and a shorter growing period of the plant mixture. In the successive years of vegetation (2014–2016), the plant mixture yield was much higher. Regardless of treatment, the highest plant mixture yield was obtained in 2016. It was more than 80% higher compared to the first year of the experiment. Analysis of the value of correlation coefficient points to significant relationships between the plant mixture yield obtained in individual years and the amount of Zn introduced with the waste dose ($r = 0.635$–0.723; $p \leq 0.05$). Such a relationship was not observed for the remaining heavy metals.

Heavy metal content in the plant mixture

It was determined that applying only cellulose production waste (treatment 2) significantly increased the content of Cr, Cd, Pb, Cu, and Zn in the plant mixture in comparison to control (Table 4). The increase in content of these metals in the plant mixture amounted to: 39, 29, 28, 8, 32%, respectively, in relation to the control. The increase in content of the metals in the plants was a result of their additional introduction to the soil with cellulose production waste (Table 1).

Applying only municipal sewage sludge (treatment 3) increased the content of Cr, Cu, and Zn in the plant mixture in comparison to control. That increase was 28, 10, 36%, respectively, in relation to the control. Under conditions of the conducted experiment, municipal sewage sludge did not have a significant effect on the increase or decrease in content of Ni, Cd, and Pb in the plant mixture compared to the control. In all probability, this resulted from the relatively low content of these metals in the applied sewage sludge (Table 1). Applying the mixture of cellulose production waste and municipal sewage sludge in single and double dose (treatments 4, 5) significantly increased Zn content in the plant mixture compared to the control. The increase in content of this metal amounted to 17 and 48%, respectively, in relation to the control. In the case of Cr and Ni, no significant effect of the

Table 3 Yield of plant mixture [Mg D.M. ha^{-1}]

No	Treatment[a]	Year of vegetation				Average
		2013	2014	2015	2016	2013–2016
1	C	3.45 ± *0.10*	5.76 ± *0.62*	5.87 ± *0.44*	6.30 ± *0.47*	5.34 ± *0.35*
2	CPW	3.34 ± *0.10*	5.47 ± *0.31*	5.65 ± *0.08*	6.17 ± *0.33*	5.16 ± *0.15*
3	MSS	4.31 ± *0.20*	6.97 ± *0.17*	7.12 ± *0.25*	7.98 ± *0.26*	6.60 ± *0.10*
4	II CPW × MSS	4.51 ± *0.23*	7.42 ± *0.44*	7.32 ± *0.30*	8.15 ± *0.45*	6.85 ± *0.26*
5	II CPW × MSS	4.36 ± *0.13*	7.15 ± *0.09*	7.21 ± *0.30*	8.11 ± *0.17*	6.71 ± *0.06*
V%[b]		13.8	13.4	12.2	13.8	13.2
$LSD_{(\alpha \leq 0.05)}$		0.24	0.57	0.45	0.53	0.32

[a] Explanations as in Table 2

[b] Variation coefficient

Table 4 Heavy metal contents in plant mixture [mg kg^{-1} D.M]

No	Treatment[a]	Cr	Ni	Cd	Pb	Cu	Zn
1	C	1.67 ± *0.12*	1.65 ± *0.03*	0.21 ± *0.01*	2.00 ± *0.02*	5.22 ± *0.18*	39.85 ± *1.89*
2	CPW	2.32 ± *0.33*	1.56 ± *0.08*	0.28 ± *0.01*	2.56 ± *0.09*	5.67 ± *0.08*	52.84 ± *4.04*
3	MSS	2.14 ± *0.07*	1.56 ± *0.13*	0.21 ± *0.01*	1.90 ± *0.09*	5.77 ± *0.20*	54.55 ± *3.90*
4	II CPW × MSS	1.87 ± *0.06*	1.51 ± *0.11*	0.20 ± *0.01*	2.04 ± *0.08*	4.92 ± *0.22*	46.89 ± *1.66*
5	II CPW × MSS	1.79 ± *0.09*	1.64 ± *0.12*	0.24 ± *0.02*	2.16 ± *0.04*	5.72 ± *0.11*	59.11 ± *5.01*
V%[b]		13.7	3.7	14.4	12.1	6.8	14.7
$LSD_{(\alpha \leq 0.05)}$		0.25	0.15	0.02	0.11	0.25	5.34

[a] Explanations as in Table 2

[b] Variation coefficient

mixtures on the increase in content of these metals in the plants was observed (Table 4).

Additionally, the double dose of the mixture of cellulose production waste and municipal sewage sludge (treatment 5) significantly increased Cd and Pb content in the plant mixture (by more than 9 and 8%, respectively) compared to the control. When analyzing Cu content in the plant mixture, it was established that a single dose of the mixture (treatment 4) had a significant effect on the decrease in content of this metal (by 6%), whereas a double dose of this mixture had a significant effect on the increase in the content of this metal (by more than 9%) compared to the control (Table 4). Analysis of linear correlation points to close relationships between Cd and Pb content in the plant mixture and the amount of yield of this mixture ($r = -0.493$–0.604; $p \leq 0.05$). Close relationships between the content of Cd, Pb, and Zn in the plant mixture and the quantity of heavy metals introduced with the sewage sludge dose were also observed ($r = 0.539$–0.823; $p \leq 0.05$).

Heavy metal uptake by the plant mixture

Application of cellulose production waste (treatment 2) caused not only an increase in the content of heavy metals, but also an increase in Cr, Cd, Pb, Cu, and Zn uptake by the plant mixture (Table 5). Increase in uptake of the above-mentioned metals in the treatment with cellulose production waste amounted to 34, 24, 23, 4, 27%, respectively, in relation to the control. Cellulose production waste decreased Ni uptake by the plant mixture, but the decrease was not statistically significant. The decrease in nickel uptake by the plant mixture, grown on cellulose production waste, resulted from lack of effect of this waste on nickel content in the plant mixture yield (Table 5). Compared to the control, application of municipal sewage sludge (treatment 3) increased the uptake of Cr, Ni, Cd, Pb, Cu, and Zn by the plant mixture. The increase in the uptake of these metals amounted to 58, 16, 18, 50, 36, and 69% in relation to the control.

Introduction of the mixture of cellulose production waste and municipal sewage sludge to the soil in single and double dose (treatments 4, 5) also significantly increased the uptake of Cr, Ni, Cd, Pb, Cu, and Zn by the plant mixture as compared with the amounts collected in the control treatment. The increase in the uptake of the above-mentioned metals was, in relation to the control, 43 and 35% for Cr, 17 and 24% for Ni, 16 and 37% for Cd, 30 and 35% for Pb, 21 and 37% for Cu, and 51 and 86% for Zn, respectively. Double dose of the waste mixture (treatment 5) to a higher degree increased the uptake of the mentioned metals to a higher degree compared to single

Table 5 Heavy metal uptake by the plant mixture as the total from the entire research period (2013–2016)

No	Treatment[a]	Cr g ha^{-1}	Ni	Cd	Pb	Cu	Zn
1	C	35.6 ± *3.50*	35.3 ± *2.30*	4.6 ± *0.52*	42.7 ± *2.91*	111.7 ± *9.02*	851.1 ± *61.88*
2	CPW	47.9 ± *6.85*	32.1 ± *1.53*	5.7 ± *0.09*	52.8 ± *2.41*	116.9 ± *3.84*	1088.7 ± *63.34*
3	MSS	56.4 ± *1.18*	41.0 ± *3.08*	5.5 ± *0.23*	50.1 ± *2.19*	152.2 ± *4.78*	1438.8 ± *95.80*
4	II CPW × MSS	51.1 ± *2.77*	41.5 ± *4.03*	5.4 ± *0.35*	55.8 ± *1.51*	135.1 ± *10.65*	1285.9 ± *92.41*
5	II CPW × MSS	48.1 ± *2.69*	43.9 ± *3.06*	6.3 ± *0.37*	57.9 ± *1.39*	153.4 ± *4.09*	1585.6 ± *98.56*
V%[b]		16.0	12.6	11.4	11.4	14.5	23.1
$LSD_{(\alpha \leq 0.05)}$		5.9	4.4	0.5	3.3	10.6	136.7

[a] Explanations as in Table 2

[b] Variation coefficient

dose of this mixture (treatment 4). Analysis of the correlation coefficient showed significant relationships between the uptake of heavy metals (Cr, Ni, Pb, Cu, Zn) by the plant mixture and the mean yield of this plant mixture ($r = 0.605–0.864$; $p \leq 0.05$). The experiment also revealed significant relationships between the content of heavy metals in the plant mixture and the uptake of these heavy metals by the plants ($r = 0.460–0.852$; $p \leq 0.05$).

Simplified balance of heavy metals

The simplified balance of heavy metals is presented in Table 6. The outcome of the balance of heavy metals depended on the amount of metals introduced with the wastes and on the amount of metals taken up with the plant mixture yield. By using cellulose production waste, significantly higher quantities of heavy metals were introduced to the soil than by using municipal sewage sludge.

Zn was an exception, because almost twice as much of this metal (compared to the treatment where only cellulose production waste was applied) was introduced to the soil with the dose of sewage sludge. With cellulose production waste (treatment 2), the following were introduced to the soil: Cr–2.36 kg, Ni–0.83 kg, Cd–0.04 kg, Pb–1.67 kg, Cu–6.63 kg, Zn–12.90 kg ha^{-1}. With municipal sewage sludge (treatment 3): Cr–0.38 kg, Ni–0.14 kg, Cd–0.03 kg, Pb–0.51 kg, Cu–1.78 kg, Zn–21.30 kg ha^{-1} (Table 6). The highest amounts of heavy metals were introduced to the soil after application of a double dose of the mixture of cellulose production waste and municipal sewage sludge (treatment 5). Such a substantial 'inflow' of metals into the soil with applied waste materials caused a significant increase in the amount of these elements in the soil. Uptake of heavy metals by the plant mixture was distinctly lower compared to the amounts introduced. As it was to be expected, the biggest difference in balance was observed in treatment 5, wherein the mixture of cellulose production waste and sewage sludge was introduced in a double dose. Based on phytoextraction of metals, it is possible to evaluate the applicability of the plant mixture for removal of heavy metals from the substrate. The highest removal of Cr, Ni, Cd, Pb, and Cu from the substrate was observed in the treatment where only municipal sewage sludge was applied (treatment 3), and Zn–in the treatment where only cellulose production waste was applied (treatment 2). The lowest effectiveness of phytoextraction of metals was observed in the treatments in which the mixture of cellulose production waste and sewage sludge was applied in a double dose (treatment 5). This was a result of several factors. In this treatment, the highest amount of metals was introduced to the substrate along with the mixture of waste, the plants yielded the least, and because of that the uptake of the studied metals by plants was lower. In the conducted experiment, regardless of dose or type of waste applied to the soil, the highest phytoremediation (recovery) was recorded for Ni–30%, followed by Cd–20%, Cr–15%, Pb–10%, Cu–9%, and the lowest for Zn–8%, in relation to the quantity introduced with wastes. The highest phytoremediation (recovery) of metals from cellulose production waste (treatment 2) was observed for Cd (13%) and the lowest for Cr and Cu (2%). In the case of municipal sewage sludge, the highest phytoremediation of metals was observed for Ni (30%) and the lowest for Zn (7%). Analysis of linear correlation coefficient showed close relationships between Zn

Table 6 Simplified balance of heavy metals after 4 years of research

No	Treatment[a]	Introduced g ha^{-1}	Uptake	Balance	Recovery %
Cr					
1	C	0	36	− 36	0
2	CPW	2365	48	2317	2
3	MSS	380	56	323	15
4	II CPW × MSS	1372	51	1321	4
5	II CPW × MSS	2744	48	2696	2
V%[b]		87.3	16.0	90.3	111.7
$LSD_{(\alpha \leq 0.05)}$		–	5.8	–	–
Ni					
1	C	0	35	− 35	0
2	CPW	831	32	799	4
3	MSS	135	41	94	30
4	II CPW × MSS	483	42	441	9
5	II CPW × MSS	966	44	922	5
V%[b]		87.2	12.6	94.6	105.8
$LSD_{(\alpha \leq 0.05)}$		–	4.4	–	–
Cd					
1	C	0	5	− 5	0
2	CPW	44	6	38	13
3	MSS	28	5	22	20
4	II CPW × MSS	36	5	30	15
5	II CPW × MSS	71	6	65	9
V%[b]		72.5	11.4	83.7	31.9
$LSD_{(\alpha \leq 0.05)}$		–	0.5	–	–
Pb					
1	C	0	43	− 43	0
2	CPW	1675	53	1622	3
3	MSS	510	50	460	10
4	II CPW × MSS	1092	56	1036	5
5	II CPW × MSS	2185	58	2127	3
V%[b]		80.1	11.4	83.6	63.1
$LSD_{(\alpha \leq 0.05)}$		–	3.3	–	–
Cu					
1	C	0	112	− 112	0
2	CPW	6634	117	6517	2
3	MSS	1775	152	1623	9
4	II CPW × MSS	4205	135	4069	3
5	II CPW × MSS	8409	153	8256	2
V%[b]		81.7	14.5	84.2	83.9
$LSD_{(\alpha \leq 0.05)}$		–	10.6	–	–
Zn					
1	C	0	851	− 851	0
2	CPW	12,900	1089	11,811	8
3	MSS	21,300	1439	19,861	7
4	II CPW × MSS	17,100	1286	15,814	8
5	II CPW × MSS	34,200	1586	32,614	5
V%[b]		72.8	23.1	76.8	23.7
$LSD_{(\alpha \leq 0.05)}$		–	136.7	–	–

[a] Explanations as in Table 2

[b] Variation coefficient

phytoremediation and yielding of the plant mixture in the studied period ($r = -0.503$–0.583; $p \leq 0.05$). Phytoremediation of Cr, Ni Cd, Pb, Cu was also significantly correlated with Pb content and uptake by the plant mixture ($r = -0.505$–0.754; $p \leq 0.05$).

Discussion

In a sewage treatment process, crude sewage sludge is obtained and then subjected to stabilization and hygienization (Arthurson 2008). Physical, chemical, and biological properties of sewage sludge from industrial plants depend greatly on the used production technology, used raw materials, the generated final product, as well as on the used water (Demirbas et al. 2017). The determined properties of the sewage sludge have a substantial effect on the direction of their management (Grobelak et al. 2017).

Chemical composition of cellulose production waste and municipal sewage sludge

Cellulose production waste and municipal sewage sludge can be a valuable source of macronutrients which can be used environmentally (Arthurson 2008; Grobelak et al. 2017; Kac-Kacas and Drzas 1968). Nitrogen content in the municipal sewage sludge used in the experiment was comparable to other municipal sludge coming from sewage treatment plants located in southern Poland (Grobelak et al. 2017; Kicińska et al. 2018). Cellulose production waste does not contain nitrogen, which results (among other things) from technological processes (Buzzini and Pires 2002). The high calcium content in the municipal sewage sludge and cellulose production waste, resulting from the technology of stabilization of these wastes, is beneficial in the aspect of deacidification of acid soils and improvement of their physicochemical properties (Pérez-López et al. 2008; Kulhánek et al. 2014). In the applied wastes, an average content of phosphorus and magnesium comparable to other sewage sludges was recorded (Kominko et al. 2017). The share of phosphorus and magnesium in the studied wastes was much higher than in natural fertilizers, and this results in a higher fertilizing value of these wastes (Grobelak et al. 2017; Harasimowicz-Hermann and Hermann 2007a, 2007b). Potassium content in the analyzed wastes was low. It is generally stated that the share of potassium and sodium in this kind of waste is lower than in natural and organic fertilizers (Deeks et al. 2013; Kominko et al. 2017; Elouear et al. 2016). Average potassium content in manure amounts to 5.0 g kg^{-1}, whereas the content of this element in the studied wastes did not exceed 1.55 g kg^{-1} DM. Assuming its use in fertilization, it is recommended to take into consideration supplementary fertilization with potassium (Elouear et al. 2016). Heavy metal content in the wastes used in the experiment did not exceed acceptable values determining their use in reclamation (Regulation 2015).

Crop yield

As a result of the carried out field experiment, it was established that the mixtures of cellulose production waste and municipal sewage sludge significantly increased the plant mixture yield. It is possible that the obtained increase in yield of the plants grown on soil with the addition of the tested wastes was a result of improvement of soil physicochemical properties and introduction of a considerable amount of biogenic elements (Arthurson 2008; Kominko et al. 2017). Research conducted by Sharma et al. (2018) showed that waste mixtures containing (for example) ash and sewage sludge increase the yield of cultivated plants. Other research also showed that sludge-ash mixtures, apart from improving soil physicochemical properties, constitute an alternative form of recycling of nutrients and, at the same time, can have a beneficial effect on yielding of cultivated and energetic plants (Baran et al. 2015; Deeks et al. 2013; Kominko et al. 2017).

In the authors' own research, cellulose production waste reduced yielding of the plant mixture, but the difference was not statistically significant. Mineral waste often contains many alkaline compounds which can quickly react in the soil sorption complex, thus contributing to inhibition of germination, a reduction in plant emergence, growth and development, and in consequence to a reduction in plant yield (Baran and Antonkiewicz 2017; Das and Singh 2004). In the research conducted by Kac-Kacas and Drzas (1968), it was shown that waste limes, on account of their specific chemical properties, can have an adverse impact on the yield of cultivated plants, for example brewer's barley or brown mustard. Cellulose production sludge with a higher concentration of free alkalis resulted in a higher decrease in yield compared to reactive quicklime (Kac-Kacas and Drzas 1968). Other studies indicate that carbonated lime, similarly to cellulose production waste, in its composition contains calcium in carbonate form ($CaCO_3$), which 'acts' more slowly in light soils compared to quicklime (Holland et al. 2018). That is why cellulose production waste, after being applied to soil, should become a slow-release fertilizer and should not cause a rapid response in soil or other substrate (Pérez-López et al. 2008).

In the research on waste composites containing cellulose production waste and municipal sewage sludge, it was shown that these mixtures improve yielding of crops, e.g., mustard (Harasimowicz-Hermann and Hermann 2007b). By contrast, research conducted by Lahori et al. (2017) showed that lime with the addition of waste materials decreases the biomass of crucifers. Lahori et al. (2017) believe that the above-mentioned materials reduce the toxicity of soil contaminated with heavy metals. The authors' own research has shown that municipal sewage sludge increased the yields of the plant mixture. The increase in plant yield can be explained by a substantial content of organic matter, macroelements, including yield-enhancing nitrogen, occurring mainly in organic form (Kominko et al. 2017). However, research results are inconclusive also in this matter. Corrêa Martins et al. (2016) determined that with a substantial content of organic compounds in sewage sludge, these wastes can have a toxic effect on plants, limiting their growth, development, and yielding.

Heavy metal content in the plant mixture

In the conducted experiment, a significant effect of cellulose production waste on the increase in the content of Cr, Cd, Pb, Cu, and Zn in the plant mixture was determined. The source of these metals in the plant mixture was waste raw material in the form of waste paper (Demirbas 2008). The Paper and Cardboard Factory produces, among other things, building board and paper towels (made of waste paper from color press, denim jeans) that contain dyes which in turn contain heavy metals (Das and Singh 2004; Tucker et al. 2000). In the course of processing waste paper into paper products, heavy metals (which can pass into the cellulose production waste) are released (Ngah and Hanafiach 2008). In addition, cellulose production waste contains many other nutrients, including micro- and macroelements which can be used under non-consumable plants (industrial, decorative, flowerbed plants) (Harasimowicz-Hermann and Hermann 2007a, 2007b; Kac-Kacas and Drzas 1968). Applying municipal sewage sludge to soil may contribute to an increase in heavy metal content in plants (Antonkiewicz et al. 2018; Sharma et al. 2018).

Municipal sewage sludge applied to soil in the authors' own research increased the content of Cr, Cu, and Zn in the plant mixture. The increase in content of these metals can be explained by rapid mineralization of organic matter from sewage sludge and by the release of these nutrients to plants (Wójcikowska-Kapusta et al. 2017). The mixture of cellulose production waste and sewage sludge increased the content of Cd, Pb, Cu, and Zn in the plant mixture, but it did not increase the content of Cr and Ni in the plant mixture. Numerous studies confirm that certain metals, despite alkalizing with cellulose production waste, are (in these conditions) mobile in the environment, quickly migrate from the substrate to the aboveground parts of plants (Kabata-Pendias and Mukherjee 2007). The mixture of cellulose production waste and municipal sewage sludge applied (in a double dose) in the authors' own research increased the content of Cd, Cu, and Zn in the plant mixture. Scientific literature indicates that mixtures containing sewage sludge and waste rich in calcium compounds (e.g., ash, waste carbonate lime) can inhibit the uptake of heavy metals by plants grown on these wastes (Holland et al. 2018; Sharma et al. 2018). In addition, calcium compounds occurring in waste can be taken up by plants and then influence heavy metal retention in these plants (Zhu et al. 2016; Lahori et al. 2017). Other studies show that waste containing cellulose participates in the biosorption, which includes chemisorption, complexing, as well as adsorption on the surface and pores, ion exchange, microprecipitation, heavy hydroxide condensation on biolayers, and surface adsorption (Demirbas 2008). The above-mentioned biosorption processes can also take place in soil solutions.

Assessment of heavy metal content in the plant mixture

Biomass of the plant mixture obtained in the field experiment meets requirements in terms of the content of Cr, Ni, Cd, Pb, Cu, and Zn set for good quality feeds (Kabata-Pendias et al. 1993). The applied cellulose production waste and municipal sewage sludge, as well as their mixtures, did not result in the content of the above-mentioned metals in the plant biomass intended for feed purposes being exceeded. Low content of heavy metals in the plant biomass results from low solubility of these metals from cellulose production waste (Demirbas 2008; Harasimowicz-Hermann and Hermann 2007a). Other studies confirm that alkaline waste (containing calcium compounds) contains heavy metals in forms hardly available for plants (Holland et al. 2018; Baran and Antonkiewicz 2017).

The biomass obtained in the conducted experiment, grown on the tested waste, should not, however, be intended for feed purposes. This biomass should be intended for compost for reclamation, or for energy purposes, or used for other non-feed-related purposes (Dinel et al. 2004). The applied cellulose production waste and municipal sewage sludge, as well as their mixtures, can be used as a substrate, a medium for growing plants cultivated for non-consumption and non-feed-related purposes (Antonkiewicz et al. 2018). It can be mainly intended for energy purposes or for production of composts intended to be used in floriculture, cultivation of decorative plants etc. (Decision of MARD 2017).

Heavy metal uptake by the plant mixture

The amount of uptake of elements depends on the amount of plant yield and on the content of these nutrients in biomass (Antonkiewicz et al. 2018; Sarma 2011). The field experiment showed a significant effect of cellulose production waste on heavy metal uptake by the plant mixture. The increase in heavy metal uptake by the plant mixture can be explained by a higher content of these elements in the used waste. Cellulose production waste was rich in trace elements and that is why the plant mixture took them up in higher amounts than the control. Cellulose production waste, despite alkaline character, can make metals available to plants, which finds confirmation in the research by Harasimowicz-Hermann and Hermann (2007b). In the research conducted by Otero et al. (2005), it was shown that fertilizers containing calcium can be a source of heavy metals potentially available to plants. Another research (Nurmesniemi et al. 2010) showed that industrial waste from paper industry, containing heavy metals in its composition, can cause a higher uptake of heavy metals by plants. Heavy metal uptake by plants can be reduced by adding sewage sludge to this alkaline waste, which was confirmed in previous research (Baran and Antonkiewicz 2017). In the authors' own research, it was also established that mixing cellulose production waste with sewage sludge reduced the intensity of heavy metal uptake by the plant mixture. Scientific literature reports that waste limes can be a source of metals that are taken up by plants, even under neutral or alkaline reaction of the substrate (Lahori et al. 2017; Zhu et al. 2016).

Simplified balance of heavy metals

By using cellulose production waste and municipal sewage sludge, as well as their mixtures, considerable quantities of heavy metals (whose certain amounts were taken up by the cultivated plants) were introduced to the soil. Understanding the circulation of heavy metals in the soil-waste-plant system, through simplified balance, will allow to assess the effectiveness of phytoextraction of these metals by applying the plant mixture (Antonkiewicz et al. 2018; Nissim et al. 2018; Sarma 2011). Negative value of the balance in control treatments resulted from the simplified balance in which the supply of metals from precipitation was not taken into account. Phytoremediation can be used to assess the effectiveness of removing metals from the substrate (Antonkiewicz et al. 2018; Nissim et al. 2018). The research indicates that the plant mixture removed Ni from the substrate most effectively (30%), followed by Cd (20%), Cr (15%), Pb (10%), and Cu and Zn to the smallest extent (9 and 8%, respectively). Research by Li et al. (2011) showed that grasses are effective in phytoextraction of nickel from waste-contaminated substrate. Research by Murray-Gulde et al. (2005) confirms that Cu is an element which is difficult to extract from the substrate. That is why specific plant species capable of absorbing large amounts of this element are sought. Other research confirms that grasses and mixtures of grasses are effective in purification of heavy metal-contaminated soils (Wójcikowska-Kapusta et al. 2017). Research by Garcia-Rodríguez et al. (2014) confirms our own results that individual grass species are effective in extracting heavy metals from sewage sludge.

Conclusions

1. A significant effect of the mixtures of cellulose production waste and municipal sewage sludge on the increase in the plant mixture yield was observed. Applying only the sewage sludge also increased the yield of the plants. Applying only the cellulose production waste did not significantly decrease the yield of the plants.
2. The cellulose production waste significantly increased the content of Cr, Cd, Pb, Cu, and Zn in the plant mixture. The municipal sewage sludge increased the content of Cr, Cu, and Zn in the plant mixture. The mixture of cellulose production waste and municipal sewage sludge applied (in a double dose) increased the content of Cd, Pb, Cu, and Zn in the plant mixture.
3. The cellulose production waste significantly increased the uptake of Cr, Cd, Pb, and Zn by the plant mixture, whereas the sewage sludge significantly increased the uptake of Cr, Ni, Cd, Pb, Cu, and Zn by the plant mixture. Introduction of the mixture of cellulose production waste and sewage sludge (in single and double dose) to the soil also increased the uptake of these metals in relation to the control treatment.
4. The plant mixture extracted more heavy metals from the sewage sludge than from the cellulose production waste. Among the analyzed heavy metals, the highest phytoremediation (recovery) was observed for Ni (30%), followed by Cd (20%), Cr (15%), Pb (10%), and the lowest for Cu (9%) and Zn (8%).
5. Due to the chemical composition of the wastes, it can be stated that each batch intended to be used in environmental management should be bioassayed and undergo chemical tests.

Funding The research was financed by the Ministry of Science and Higher Education of the Republic of Poland, subject No. 3101.

References

Antonkiewicz J, Kołodziej B, Bielińska E, Witkowicz R, Tabor S (2018) Using Jerusalem artichoke to extract heavy metals from municipal sewage sludge amended soil. Pol J Environ Stud 27(2):513–527. https://doi.org/10.15244/pjoes/75200

Arthurson V (2008) Proper sanitization of sewage sludge: a critical issue for a sustainable society. Appl Environ Microbiol 74(17):5267–5275. https://doi.org/10.1128/AEM.00438-08

Baran A, Antonkiewicz J (2017) Phytotoxicity and extractability of heavy metals from industrial wastes. Environ Prot Eng 43(2):143–155. https://doi.org/10.5277/epe170212

Baran S, Wójcikowska-Kapusta A, Żukowska G, Bik-Małodzińska M, Wesołowska-Dobruk S (2015) Influence of sludge-ash composts on some properties of reclaimed land. Arch Environ Protect 41(2):82–88. https://doi.org/10.1515/aep-2015-0022

Buzzini AP, Pires EC (2002) Cellulose pulp mill effluent treatment in an upflow anaerobic sludge blanket reaktor. Process Biochem 38(5):707–713. https://doi.org/10.1016/S0032-9592(02)00190-5

Corrêa Martins MN, de Souza VV, Souza TS (2016) Genotoxic and mutagenic effects of sewage sludge on higher plants. Ecotoxicol Environ Saf 124:489–496. https://doi.org/10.1016/j.ecoenv.2015.11.031

Couret L, Irle M, Belloncle C, Cathala B (2017) Extraction and characterization of cellulose nanocrystals from post-consumer wood fiberboard waste. Cellulose 24(5):2125–2137. https://doi.org/10.1007/s10570-017-1252-7

Das H, Singh SK (2004) Useful byproducts from cellulosic wastes of agriculture and food industry — a critical appraisal. Crit Rev Food Sci Nutr 44(2):77–89. https://doi.org/10.1080/10408690490424630

Decision of MARD (2017) Decision of the Ministry of Agriculture and Rural Development No G-649/17 dated 16 August 2017 on the applicant's permission to release an organic soil conditioner to the market

Deeks LK, Chaney K, Murray C, Sakrabani R, Gedara S, Le MS, Tyrrel S, Pawlett M, Read R, Smith GH (2013) A new sludge-derived organo-mineral fertilizer gives similar crop yields as conventional fertilizers. Agron Sustain Dev 33(3):539–549. https://doi.org/10.1007/s13593-013-0135-z

Demirbas A (2008) Heavy metal adsorption onto agro-based waste materials: a review. J Hazard Mater 157(2–3):220–229. https://doi.org/10.1016/j.jhazmat.2008.01.024

Demirbas A, Edris G, Alalayah WM (2017) Sludge production from municipal wastewater treatment in sewage treatment plant. Energy Sources Part A 39(10):999–1006. https://doi.org/10.1080/15567036.2017.1283551

Dinel H, Marche T, Schnitzer M, Paré T, Champagne P (2004) Co-composting of paper mill sludge and hardwood sawdust under two types of in-vessel processes. J Environ Sci Health B 39(1):139–151

Directive 99/31/EC (1999) Council Directive 1999/31/EC of 26 April 1999 on the landfill of waste. Off J Eur Communities 182(16):1–19

Elouear Z, Bouhamed F, Boujelben N, Bouzid J (2016) Application of sheep manure and potassium fertilizer to contaminated soil and its effect on zinc, cadmium and lead accumulation by alfalfa plants. Sustain Environ Res 26(3):131–135. https://doi.org/10.1016/j.serj.2016.04.004

Garcia-Rodríguez A, Matamoros V, Fontaź C, Salvadó V (2014) The ability of biologically based wastewater treatment systems to remove emerging organic contaminants - a review. Environ Sci Pollut Res 21:11708–11728. https://doi.org/10.1007/s11356-013-2448-5

Grobelak A, Placek A, Grosser A, Singh BR, Almås ÅR, Napora A, Kacprzak M (2017) Effects of single sewage sludge application on soil phytoremediation. J Clean Prod 155(Part 1):189–197. https://doi.org/10.1016/j.jclepro.2016.10.005

Harasimowicz-Hermann G, Hermann J (2007a) Organic and mineral waste mixtures intended for using in agriculture. Part I Composition of wastes and their mixtures. Adv Agric Sci Probl Issues 520:295–301 [In polish]

Harasimowicz-Hermann G, Hermann J (2007b) Organic and mineral waste mixtures intended for using in agriculture. Part II. Use of waste mixtures for white mustard (Sinapsis alba L.) fertilization. Adv Agric Sci Probl Issues 520:303–312 [In polish]

Holland JE, Bennett AE, Newton AC, White PJ, McKenzie BM, George TS, Pakeman RJ, Bailey JS, Fornara DA, Hayes RC (2018) Liming impacts on soils, crops and biodiversity in the UK: a review. Sci Total Environ 610-611:316–332. https://doi.org/10.1016/j.scitotenv.2017.08.020

Jones JB Jr, Case VV (1990) Sampling, handling, and analyzing plant tissue samples. In: Westerman RL (ed) Soil testing and plant analysis, SSSA Book Series, vol 3, 2nd edn. Soil Science Society of America, Madison, pp 389–427 ISBN:0-89118-793-6

Kabata-Pendias A, Motowicka-Terelak T, Piotrowska M, Terelak H, Witek T (1993) Assessment the degree of soil and plant pollution with heavy metals and sulphur. Framework guidelines for agriculture. Puławy, series P. (53), IUNG, pp. 20. [in Polish]

Kabata-Pendias A, Piotrowska M, Motowicka-Terelak T, Maliszewska-Kordybach T, Filipiak K, Krakowiak A, Pietruch C (1995) Basis for the assessment of chemical contamination of soil - heavy metals, sulfur and PAHs. State Inspectorate for Environmental Protection. Library of Environmental Monitoring, Warsaw, p 41

Kabata-Pendias A, Mukherjee AB (2007) Trace elements from soil to human. Springer-Verlag, Berlin, pp 87–93. https://doi.org/10.1007/978-3-540-32714-1

Kac-Kacas M, Drzas K (1968) Evaluation of some by-products of the industry as lime fertilizers. Part I. Lime by-products from soda production, paper works carbonate, lime waste product after acetylene production, lime by-product from the Sulphur industry, lime by-product from the power station (ash from brown coal). Pamiętnik Puławski - Prace IUNG 32:67–87 [in Polish]

Kicińska A, Kosa-Burda B, Kozub P (2018) Utilization of a sewage sludge for rehabilitating the soils degraded by the metallurgical industry and a possible environmental risk involved. Hum Ecol Risk Assess 24(7):1990–2010. https://doi.org/10.1080/10807039.2018.1435256

Kominko H, Gorazda K, Wzorek Z (2017) The possibility of organo-mineral fertilizer production from sewage sludge. Waste Biomass Valoriz 8(5):1781–1791. https://doi.org/10.1007/s12649-016-9805-9

Kulhánek M, Balík J, Černý J, Vašák F, Shejbalová Š (2014) Influence of long-term fertilizer application on changes of the content of Mehlich-3 estimated soil macronutrients. Plant Soil Environ 60(4):151–157

Lahori AH, Zhang Z, Guo Z, Mahar A, Li R, Awasthi MK, Sial TA, Kumbhar F, Wang P, Shen F, Zhao J, Huang H (2017) Potential use of lime combined with additives on (im)mobilization and phytoavailability of heavy metals from Pb/Zn smelter contaminated soils. Ecotoxicol Environ Saf 145:313–323. https://doi.org/10.1016/j.ecoenv.2017.07.049

Li GY, Hu N, Ding DX, Zheng JF, Liu YL, Wang YD, Nie XQ (2011) Screening of plant species for phytoremediation of uranium, thorium, barium, nickel, strontium and lead contaminated soils from a uranium mill tailings repository in South China. Bull Environ Contam Toxicol 86(6):646–652. https://doi.org/10.1007/s00128-011-0291-2

Mymrin V, Pedroso DE, Pedroso C, Alekseev K, Avanci MA, Winter E Jr, Cechin L, Rolim PHB, Iarozinski A, Catai RE (2018) Environmentally clean composites with hazardous aluminum anodizing sludge, concrete waste, and lime production waste. J Clean Prod 174:380–388. https://doi.org/10.1016/j.jclepro.2017.10.299

Murray-Gulde CL, Huddleston GM, Garber KV, Rodgers JH Jr (2005) Contributions of *Schoenoplectus californicus* in a constructed wetland system receiving copper contaminated wastewater. Water Air Soil Pollut 163(1–4):355–378. https://doi.org/10.1007/s11270-005-1297-3

Ngah WSW, Hanafiach MAKM (2008) Removal of heavy metal ions from wastewater by chemically modified plant wastes as adsorbents: a review. Bioresour Technol 99(10):3935–3948. https://doi.org/10.1016/j.biortech.2007.06.011

Nissim WG, Cincinelli A, Martellini T, Alvisi L, Palm E, Mancuso S, Azzarello E (2018) Phytoremediation of sewage sludge contaminated by trace elements and organic compounds. Environ Res 164:356–366. https://doi.org/10.1016/j.envres.2018.03.009

Nurmesniemi H, Pöykiö R, Watkins G, Dahl O (2010) Total and extractable heavy metal, phosphorous and sulfur concentrations in slaker grits from the causticizing process of a pulp mill for use as a soil amendment. Chem Speciat Bioavailab 22(2):87–97. https://doi.org/10.3184/095422910X12692615383543

Ostrowska A, Gawliński S, Szczubiałka Z (1991) Methods of analysis and assessment of soil and plant properties. A Catalgoue. Institute of Environmental Protection – National Research Institute, Warsaw, p 334

Otero N, Vitòria L, Soler A, Canals A (2005) Fertiliser characterisation: major, trace and rare earth elements. Appl Geochem 20(8):1473–1488. https://doi.org/10.1016/j.apgeochem.2005.04.002

Patent PL411157-A1, Antonkiewicz J, Michalak P, Targosz B (2015) Producing fertilizer from sewage sludge for improving soil structure in agricultural applications, comprises mixing thoroughly sewage sludge and binder in the form of flour and adding quick lime, and dried bentonite, granulating and drying. Uniwersytet Rolniczy im. Hugona Kołłątaja w Krakowie z siedzibą w Krakowie, HELIOCHEM z siedzibą w Ciechocinku, Fabryka Papieru i Tektury „Beskidy" Spółka Akcyjna z siedzibą w Wadowicach. http://apps.webofknowledge.com/full_record.do?colName=DIIDW&recordID=201654961Y&log_event=no&search_mode=GeneralSearch&qid=1&log_event=yes&product=UA&SID=C5hjaxUVDr1nhrUYGSq&viewType=fullRecord&doc=10&page=1

Pérez-López R, Montes-Hernandez G, Nieto JM, Renard F, Charlet L (2008) Carbonation of alkaline paper mill waste to reduce CO_2 greenhouse gas emissions into the atmosphere. Appl Geochem 23(8):2292–2230. https://doi.org/10.1016/j.apgeochem.2008.04.016

Polish Soil Classification (2011) Soil Sci Ann 62(3):1–193 http://www.ptg.sggw.pl

Regulation (2015) Regulation of the Minister of the Environment on municipal sewage sludge dated 6 February 2015. J Law Poland, item 257. http://isap.sejm.gov.pl/DetailsServlet?id=WDU20150000257

Regulation (2016) Regulation of the Minister of the Environment on how to conduct land surface pollution assessment dated 1 September 2016. J Law Poland, item 1395. http://isap.sejm.gov.pl/DetailsServlet?id=WDU20160001395

Sarma H (2011) Metal hyperaccumulation in plants. A review focusing on phyoremediation technology. J Environ Sci Technol 4(2):118–138. https://doi.org/10.3923/jest.2011.118.138

Sharma B, Kothari R, Singh RP (2018) Growth performance, metal accumulation and biochemical responses of Palak (*Beta vulgaris* L. var. Allgreen H-1) grown on soil amended with sewage sludge-fly ash mixtures. Environ Sci Pollut Res 25(13):12619–12640. https://doi.org/10.1007/s11356-018-1475-7

Soil Survey Staff (2014) Keys to soil taxonomy, 12th edn. USDA-Natural Resources Conservation Service, Washington, DC

Tucker P, Douglas P, Durrant A, Hursthouse AS (2000) Heavy metal content of newspaper: longitudinal trends. Environ Manag Health 11(1):47–66. https://doi.org/10.1108/09566160010314189

Tunali Y, Ertunç S (2017) Use of natural soda ash production process waste for SO_2 removal. Environ Prot Eng 43(2):69–80. https://doi.org/10.5277/epe170206

Wan Ngah WS, Hanafiah MAKM (2008) Removal of heavy metal ions from wastewater by chemically modified plant wastes as adsorbents: a review. Bioresour Technol 99(10):3935–3948. https://doi.org/10.1016/j.biortech.2007.06.011

Waste Act (2012) Waste Act dated 14 December 2012. J Law Poland. No O, Item 21. http://isap.sejm.gov.pl/DetailsServlet?id=WDU20130000021

Waste Catalogue (2014) Regulation of the Minister of the Environment on catalog of wastes dated 9 December 2014. J Law Poland, Item 1923. http://isap.sejm.gov.pl/DetailsServlet?id=WDU20140001923

Wójcikowska-Kapusta A, Urban D, Baran S, Bik-Małodzińska M, Żukowska G, Pawłowski A, Czechowska-Kosacka A (2017) Evaluation of the influence of composts made of sewage sludge, ash from power plant, and sawdust on floristic composition of plant communities in the plot experiment. Environ Prot Eng 43(2):129–141. https://doi.org/10.5277/epe170211

Zhu H, Chen C, Xu C, Zhu Q, Huang D (2016) Effects of soil acidification and liming on the phytoavailability of cadmium in paddy soils of central subtropical China. Environ Pollut 219:99–106. https://doi.org/10.1016/j.envpol.2016.10.043