

Article

Heavy Metals in Fly Ash as a Factor Limiting Its Use in Fertilizing Composts

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Abstract: Composting enables the recycling of significant amounts of biodegradable waste, while ensuring its disposal. The addition of fly ash increases the concentration of fertilizing substances in the compost. Compost is a source of many nutrients for plants. The addition of fly ash might also cause a slower release of these nutrients into an aqueous solution, meeting the fertilization needs of plants over a longer period of time. Tests were carried out using sewage sludge (WWTP Piaseczno, 163,500 P.E.), straw as a structure-forming material and fly ash (WWTP Krakow, 780,000 P.E.). The compost obtained in the research was analyzed in terms of environmental conditions specified in legal regulations. The most favorable compost mass density (520 kg/L) and the amount of air supplied to the process (5.0 L/(h·kg d.m.)) were determined. The addition of fly ash to the compost mass did not significantly affect the temperature distribution obtained in the process. The increase in fly ash content increased the pH of the compost mass and was associated with higher nitrogen losses. It was found that the factor which may limit the possibility of using fly ash as a compost substrate is the presence of higher concentrations of heavy metals, especially chromium. It was determined that the maximum addition of fly ash to the compost mass was 154 kg d.m._{ash}/(ton d.m._{straw+sludge}).

Keywords: composting; heavy metals; fly ash doses



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1. Introduction

The most common organic fertilizer in Poland is manure. However, since 1990, its production has decreased significantly, so the doses used today often turn out to be insufficient. Therefore, other sources of organic substances that can be used as an alternative fertilizer are sought. One of them is sewage sludge and the compost based on it.

Sewage sludge is formed at various stages of wastewater treatment. The amount is about 1–3% of the volume of treated sewage. Unfortunately, sludge can pose a serious threat to the environment if not managed properly. It contains many substances, e.g., heavy metals and pathogenic organisms [1].

Activities aimed at the treatment of sewage sludge and its appropriate handling are regulated in the relevant framework directive of the European Union [2]. The main purpose of its processing is stabilization and volume reduction.

Sewage sludge management is a significant problem. The increase in the amount of sludge produced is caused by the expansion of the sewage network, and thus, the increase in the amount of wastewater and the use of in-depth removal of biogenic compounds, i.e., nitrogen and phosphorus, for their treatment.

Due to the potential harmful impact of sewage sludge on the environment, further innovation in the combustion methods used for their treatment is forecast, especially in highly urbanized areas [3]. So far, the management of municipal sewage sludge in agglomerations has been carried out in few directions [4,5]. One of them involves the use of thermal methods that lead to the drying and combustion of sewage sludge. Such solutions

are used primarily in larger wastewater treatment plants (WWTPs) serving large urban agglomerations. In smaller objects, technologies are used to prepare the sewage sludge for its agricultural use. The agricultural management of sludge is mainly based on the use of sludge in agriculture and for purposes such as reclamation of landfills, composting, use for growing plants not intended for consumption and production of fodder, as well as its temporary storage at the WWTP. However, in this case, sewage sludge must meet the requirements for the natural use of sewage sludge, which is regulated in Poland by the Regulation of the Minister of the Environment of 6 February 2015 on Municipal Sewage Sludge [6].

The management of sewage sludge in Europe in 2020 included processes such as [7,8]:

- Approximately 30% used in agriculture;
- Approximately 23% processed by incineration;
- Approximately 16% used in landfill;
- Approximately 22% used in compost;
- Approximately 21% used in other ways (forestry, silviculture, melioration, etc.).

The increase in thermal methods of sewage sludge treatment leads to the formation of significant amounts of fly ashes, which also need to be managed. Research is being conducted on the use of fly ashes in concrete technology [9] and the recovery of valuable components from it, e.g., phosphorus [10,11].

From 1 January 2016, regulations limit to landfill use any sewage sludge for which the limit value of TOC < 5% of dry matter, LOI < 8% of dry matter, and the heat of combustion < 6 MJ/kg of dry matter [12].

Sewage sludge can be a source of nutrients in the soil environment. For this reason, it is justified to use it for agricultural purposes. Its use should be correct, taking into consideration the safety of animals, plants, the soil environment and humans. It should not impair the quality of the soils on which it is applied, nor the quality of agricultural products.

Municipal sewage sludge used as fertilizers after composting processes significantly increases soil humus and is a valuable source of nutrients [13,14]. This method of management can be as supplementary fertilization or by replacing other fertilization with natural fertilizers. The presence of sewage sludge in the soil improves its physical and chemical properties and ensures an increase in the fraction of organic matter [15]. The addition of sewage sludge, which is characterized by a low concentration of heavy metals, can contribute to the increase of biomass, and also positively affect the number of microorganisms occurring in the soil [16], which are involved in the mineralization of organic nitrogen.

Composting is a method of biological aerobic treatment of organic waste, including sewage sludge, the primary effect of which is obtaining a stable product. If the raw material used in the composting process is of appropriate quality, the compost obtained, classified as an organic fertilizer according to the applicable regulations, can be used in agriculture [17]. In the composting process, waste is disposed of in sanitary terms, and the main product is compost, i.e., a stable, sanitary material, similar to humus, rich in organic substances and not emitting unpleasant odors, which is obtained in the composting process of selectively collected bio-waste, and meets environmental requirements [18].

Sewage sludge contains important nutrients such as nitrogen, phosphorus and potassium. The improvement of the balance of organic matter in the soil improves the conditions for plant growth. The use of sewage sludge in reclamation processes contributes to the recovery of valuable elements such as nitrogen, phosphorus and other nutrients that are important for plant growth [19]. However, the use of sewage sludge for fertilization and reclamation is associated with certain limitations which are caused by, for example, the presence of hazardous substances, pathogenic microorganisms and an undesirable smell.

Composts containing sewage sludge from municipal sewage may contain excessive amounts of heavy metals, such as Cr, Pb, Cd, Cu, Ni, Hg and Zn, which are toxic in high concentrations. In this case, the use of sewage sludge is not appropriate for agricultural purposes and its use for the reclamation of degraded areas is significantly limited. Under

such conditions, there is a risk that too many toxic elements may be leached into ground and surface waters, creating a threat to the entire ecosystem [20].

Animal-based organic fertilizers and sewage sludge are characterized by a large diversity in the content of basic mineral components, which are important in terms of fertilization usefulness. Comparing sewage sludge to slurry (bovine and pig) or manure (chicken, turkey and cattle) [21–30], the highest contents for N, P and Ca are found in sewage sediments.

Sewage sludge is characterized by a higher content of nutrients (P_2O_5 , K_2O) [31–34].

It can therefore be concluded that sewage sludge is a good fertilizing material and a promising source for composting, which might be used to increase yields. Sewage sludge is rich in nutrients and can be considered as a valuable source of potassium and phosphorus for crops.

It is possible to mix different wastes to obtain the optimal composition of the processed mixture and to obtain compost material of the right quality. Mixing different types of waste together to improve the conditions for conducting biological processes meets the requirements of the Waste Act 2016 [35].

The process of self-composting sewage sludge is ineffective due to excessively high C/N and C/P ratios, high hydration of sewage sludge and its sticky or lumpy consistency preventing or limiting the proper circulation of process gases in the bed. Therefore, before composting, sewage sludge should be mixed with structural materials that are rich in organic matter (allowing for the reduction of the C/N and C/P ratios, lower humidity and improving the structure of the composted material). Wastes from the wood industry (e.g., bark and sawdust) and agricultural production (e.g., straw) are often used as structural material.

Particularly noteworthy are wastes generated after incineration of sewage sludge—fly ashes.

Fly ashes from biomass combustion can have different properties and can be disposed of and used in different ways [36]. They are a valuable source of fertilizing ingredients (mainly nitrogen and phosphorus) that can improve composted sludge, which, in view of the increased content of mineral fertilizing ingredients in composts, makes them an attractive product [37].

On the other hand, the addition of fly ash to compost is a source of heavy metals. It also increases the pH of the compost mass, which results in higher nitrogen losses and immobilization of microelements and a decrease in the number of beneficial microorganisms [38–42].

The presence of significant amounts of sewage sludge and the development of methods for its drying and incineration became a premise for undertaking this research topic related to the joint composting of sludge with fly ashes from sewage sludge combustion.

The aim of the research is to verify the thesis that the composting of sewage sludge with the use of fly ashes from the sewage sludge combustion allows for the obtaining of a high-quality fertilizer product. However, the limiting factor is the content of heavy metals in these ashes.

2. Material and Methods

The aim of the research was to determine the effect of the addition of fly ash to compost from sewage sludge combustion, using wheat straw as a structural material, on the quality parameters of composts produced based on sewage sludge. Additionally, the fertilizing and sanitary properties of the produced composts were determined.

Compost for the research was prepared on the basis of dehydrated sewage sludge and fly ash from an incineration plant resulting from fluidization bed combustion—FBC (efficiency 23,000 Mg d.m./yr.)—of sewage sludge from WWTP Krakow (780,000 P.E., capacity 328,000 m³/d). The structural material was wheat straw from a farm near Warsaw. Sewage sludge used for the tests, which was collected from a municipal wastewater treatment plant near Warsaw (WWTP Piaseczno, 163,500 P.E., capacity 24,000 m³/d), did not contain industrial wastewater with increased concentrations of heavy metals.

The efficiency of the composting process is significantly affected by the proper balance of the components that make up the composted material. Hydration and the content of organic and mineral dry matter in the substrates used for testing are listed in Table 1. Finally, the sewage sludge, after stabilization processes, was selected for the study.

Table 1. Hydration and dry matter in substrates used for research. (Source: authors' research.)

Component	Hydration [%]	Dry Mass [%]	Dry Organic Mass [% d.m.]	Minimum Dry Mass [% d.m.]
	Range (Average)	Range (Average)	Range	Range
Raw sludge *	91.4–92.1 (91.7)	7.9–8.6 (8.3)	76.9–78.8	21.2–23.1
Stabilized sludge **	84.7–88.0 (87)	12.0–15.3 (13)	69.9–70.3	29.6–30.1
Wheat straw	7.6–8.6 (8)	91.4–92.4 (92)	92.4–93.6	6.4–7.1

Note: * Sludge after mechanical thickening; ** Sludge after mechanical thickening, fermentation and dewatering.

After determining the appropriate weight proportions and then mixing, the substrates were placed in rotating bioreactors (Figure 1) with a volume of 150 L. Rotation speed was 5 rpm. The bioreactors were thermally insulated and equipped with an aeration system.



Figure 1. View of the bioreactors used for research.

The aeration installation was powered by a compressor. On the air supply hose to the bioreactors, rotameters were placed to measure the amount of flowing air. During the tests, the temperature in the bed and its humidity were measured.

Fly ash dosage tests were carried out for composts obtained with different mass ratios of substrates, specifically, 1:10:0, 1:10:0.5, 1:10:1, 1:10:1.5, and 1:10:2, in 3 replications. The differences in the doses of fly ash were aimed at determining their effects on the temperature in the obtained compost mass and the content of biogenic and fertilizing substances in water extracts. No tests were conducted with higher ash doses due to the expected increase in pH and large losses of nitrogen.

The tests were carried out at a compost mass density of 520 kg/L and aeration intensity of 5.0 L/(h·kg d.m.). The values were based on preliminary tests optimizing the weight proportions of compost mass substrates and their density and aeration intensity.

Compost samples for testing were collected in accordance with the methodology contained in the PN-R-04006:2000 standard. The test sample was obtained by combining and thoroughly mixing 6 samples taken at the same time from different places of the compost mass. Physical and chemical composition tests were carried out for the sewage sludge and fly ashes and the compost and its water extracts (EN 12457-4:2002 [43]) in terms of: physical properties (i.e., dry matter content [44], dry matter content organic acid [45], TOC [46], and pH [46]), fertilizer indicators (i.e., TKN [47], N-NH₄ [46], N-NO₃ [48], N-NO₂ [48],

total phosphorus [49], P-PO₄ [50], potassium [51], magnesium [51,52], calcium [47,51], chlorides [50], BOD₅ [53], and COD [54]), and heavy metals (chromium, lead, copper, cadmium, nickel, zinc, and mercury) [51,52]. The analyses were outsourced to the accredited laboratory Eurofins OBiKŚ Polska Sp. z o. o. Measurement uncertainty for sensory (e.g., pH and temperature) tests was defined as the geometric mean range. Measurement uncertainty for the rest of the tests was defined as expanded uncertainty, for which the coverage factor was set at the level of $k = 2$, and the probability factor of the measurement result was equal to 95%.

3. Results and Discussion

3.1. Influence of Fly Ash on the Composting Process

In the study, attempts were made to compost sewage sludge and a structure-forming material in the form of wheat straw with the addition of fly ash from sewage sludge combustion. In individual tests, the variable parameter was the amount of added ash (Figure 2).

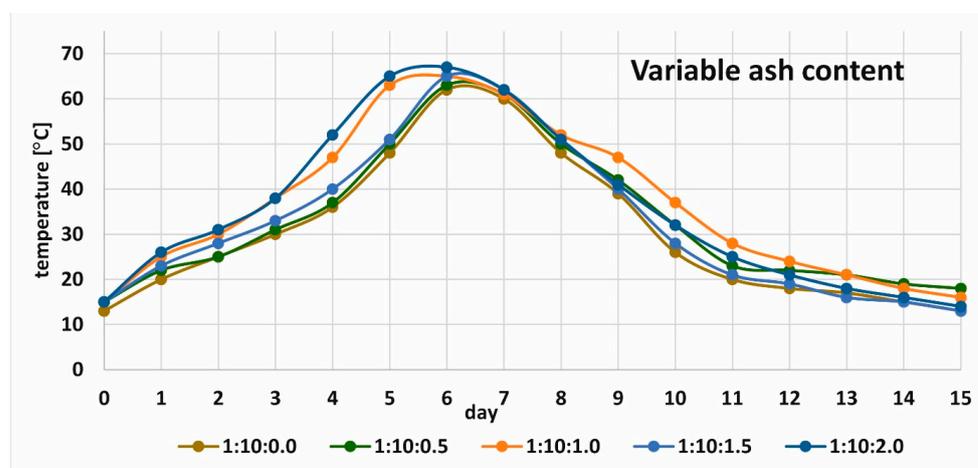


Figure 2. Composting of stabilized sludge with straw and a variable amount of ash from sewage sludge combustion at constant density (520 kg/m³) and amount of air supplied (5.0 L/(h·kg d.m.))—average of three replications.

The temperature distribution presented in Figure 2 indicates that the addition of ash does not significantly affect the obtained temperatures in the bed. The composting process proceeds properly, reaching the maximum temperature of the compost mass around the 6th day of the process. Then the temperature of the mass decreases, passing, at around the 12th day of the process, into the phase of compost maturation.

The pH for individual samples was 6.4–7.6, 6.8–7.9, 7.2–8.4, 7.5–8.9 and 7.9–9.4 for ratios of 1:10:0, 1:10:0.5, 1:10:1, 1:10:1.5 and 1:10:2, respectively. The measured pH values indicate that the higher the content of ash added to the compost mass in the analyzed samples, the higher the pH value, which is related to the addition of increasing amounts of alkaline ions (Ca²⁺, Mg²⁺, K⁺, Na⁺) to the compost. At the same time, an increase in the ash content in the composted material results in a decrease in the humidity of the composted bed, due to the much lower hydration of the ash compared to the other components of the compost mass.

Figure 3 shows the view of the compost obtained after 15 days of the process for the substrate ratio of 1:10:1.



Figure 3. View of the compost after 15 days of composting (straw:sewage sludge:fly ash—1:10:1).

3.2. Organic, Biogenic and Fertilizing Substances

The main objective of the conducted research was to determine the effect of the fly ash additive from the incineration of sewage sludge on the quality parameters of composts produced on the basis of sewage sludge, with the use of wheat straw as a structural material.

The test results listed in Table 2 indicate a high content of organic substances in the obtained composts. In all compost samples, the content of organic substances exceeds the minimum value of 30% specified in the Regulation of the Minister of Agriculture and Rural Development on the Implementation of Certain Provisions of the Act on Fertilizers and Fertilization [55]. There is also a clear dependence, indicating that the share of organic substances decreases with the increase in fly ash content in the compost.

Table 2. The content of organic substances in substrates and composts using straw as a structural material.

Indicator	Unit	Sewage Sludge	Fly Ash	Compost—Weight Proportions: Straw:Sewage Sludge:Fly Ash					[55]
				1:10:0	1:10:0.5	1:10:1	1:10:1.5	1:10:2	
TOC	mg/kg d.m.	342,600 ±68,520	<5000 -	306,400 ±61,280	252,900 ±50,580	267,800 ±53,560	151,400 ±30,280	246,000 ±49,200	-
Dry mass loss	% d.m.	80.3 ±8.03	<0.5 -	74.1 ±7.4	54.8 ±5.5	47.3 ±4.7	35.2 ±3.5	36.0 ±3.6	above 30% d.m.

Important parameters characterizing the possibility of agricultural (natural) use of compost include the content levels of fertilizing substances. Among them, the following parameters are distinguished: TKN, N-NH₄, total phosphorus, potassium, calcium, magnesium and chlorides. The list of fertilizing substances in composts with different ash content, as compared to the content in sewage sludge and fly ash, is shown in Figure 3. A significantly higher concentration of fertilizing substances in the ash compared to sewage sludge results in an increase of the concentrations in the obtained compost. It can be concluded that the addition of fly ashes from sewage sludge incineration to compost is a source of many nutrients for plants. At the same time, the release of fertilizing substances into the solution in the case of fly ashes is comparable or even slower than in the case of sewage sludge. This is a beneficial phenomenon, as it does not increase the level of eutrophication of the environment. It affects the balanced satisfaction of the fertilization needs of plants in the long term.

Nitrogen has an important role in the life cycle of the plant; it is a component of structural compounds, energy carriers and compounds that regulate the metabolism of the plant. It affects the growth rate and the amount of biomass produced by the plant. However, it is relatively difficult to specify the directions of the transformations of this component in the soil. This is due to the diversity of both the forms of the nitrogen compounds and the processes occurring between them [56].

On the basis of Figure 4, it can be concluded that the content of total nitrogen in the tested composts is significantly lower than the content in sewage sludge. This is due to low nitrogen content in the fly ash and nitrogen losses during the composting process. The

increase in the ash content in the compost mass does not significantly affect the content of total nitrogen in the compost mass.

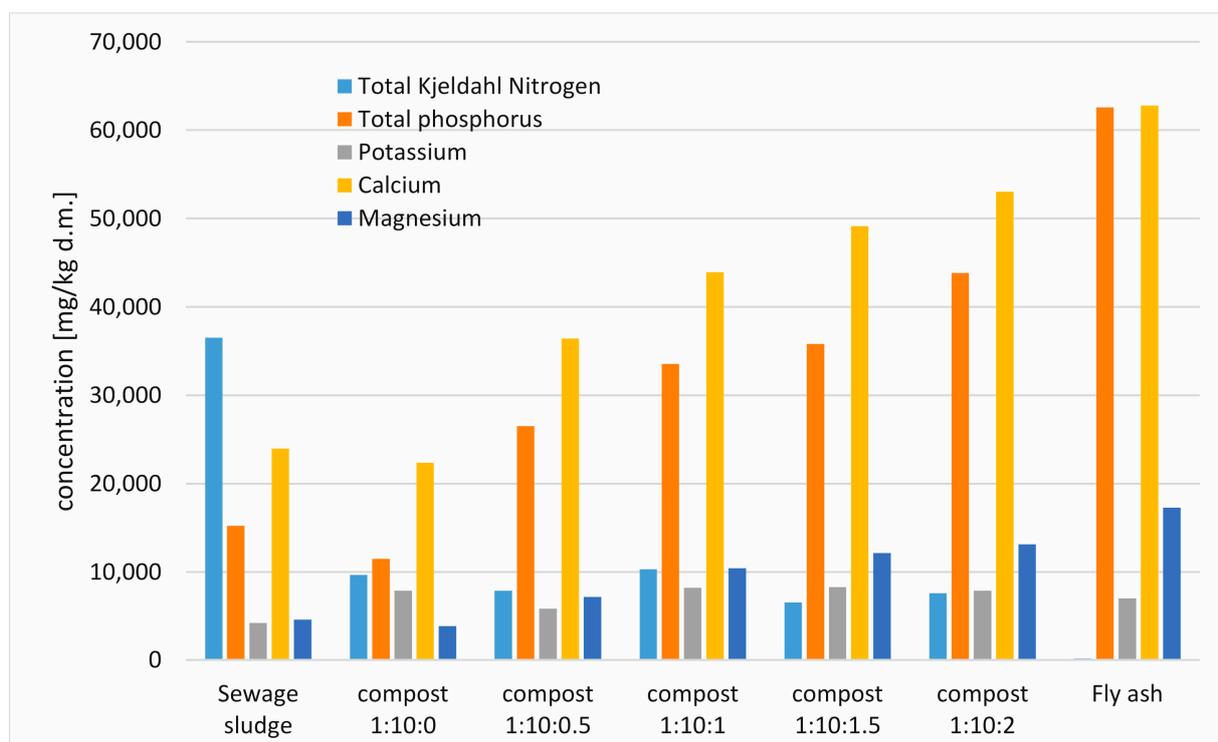


Figure 4. The content of fertilizing substances in compost (whey straw as a structural material), sewage sludge and fly ash—average of three replications.

The form of ammonium nitrogen is well retained in the soil, is easily absorbed by plants and facilitates the uptake of phosphorus and silicon. It is slowly and evenly absorbed by plants, and also helps to build a strong root system [56]. The content of ammonium nitrogen in the tested composts ranges from 0.32 to 0.95% d.m. As in the case of total nitrogen, the content of ammonium nitrogen in the compost is clearly lower than the content in the sewage sludge, and the dose of added ash does not significantly affect its value in the compost.

In the case of declaring the total nitrogen in compost for all composts obtained, we obtain values of total nitrogen significantly above the minimum value (0.3% m/m—mass of the component to the mass of the mixture) specified in the Regulation of the Minister of Agriculture and Rural Development on the Implementation of Certain Provisions of the Act on Fertilizers and Fertilization [55].

Phosphorus introduced into the soil is removed from it with the growth of plants or because of erosion, surface runoff or leaching. According to the literature, it can only be supplemented from external sources [57].

The presence of easily available phosphorus compounds in the soil determines the development of the root system. Phosphorus even stimulates root growth. The deficiency of this component, as well as many others, means that the nitrogen taken up by plants is not fully used to create a crop. Phosphorus in a plant cell is found in, among other forms, inorganic form as a fixed pool of metabolic phosphorus (Pi). It is currently used in the processes of phosphorylation and ATP synthesis. It is a component of the high-energy bond contained in the adenosine triphosphate (ATP) molecule, which is the basic source of energy for all biochemical reactions taking place in the living organism.

The content of total phosphorus in the obtained composts is in most cases higher than the content in sewage sludge. This is due to the high content of phosphorus in the fly ash. In the case of declaring phosphorus in the compost, for all composts obtained, we obtain

phosphorus values above the minimum value (0.2% m/m—weight of the component converted to phosphorus pentoxide P_2O_5 to the weight of the mixture) specified in the Regulation of the Minister of Agriculture and Rural Development on the Implementation of Certain Provisions of the Act on Fertilizers and Fertilization [55].

Potassium is an element responsible for many processes, e.g., photosynthesis processes, energy transformations, water management and osmotic processes, regulation of the daily cycle of stomatal work, ion uptake from the soil and nitrogen management. The interaction between nitrogen and potassium boils down to an increase in yields under the influence of nitrogen fertilization, and thus to a greater uptake of potassium by plants. Potassium deficiency drastically reduces nitrogen uptake, which, depending on the plant species, affects the realization of its yield potential to varying degrees [58].

The obtained test results show that the potassium content in the compost is higher than the content in the sewage sludge. The source of the higher potassium content in composts is the addition of fly ash. In the case of declaring potassium in the compost, for all composts obtained, we obtain potassium values above the minimum value (0.2% m/m—mass of the component converted to potassium oxide K_2O to the compost mass) specified in the Regulation of the Minister of Agriculture and Rural Development on the Implementation of Certain Provisions of the Act on Fertilizers and Fertilization [54].

For plants, calcium is a macroelement, i.e., it is present in an amount of more than 0.1 to 5% of dry mass. Therefore, it is one of the most important elements for plants, which, in the case of agriculture, often determines the quality and size of crops. Calcium has both a structural role and the function of a universal information transmitter. In the plant, it is very poorly reutilized, which means that the symptoms of its deficiency are observed in the form of the curling of the edges of young leaves or a weakening of the root system, which is then covered with mucus. It is also a key component that regulates the functions of the plasma membrane. Calcium deficiency causes severe damage to plants, up to the atrophy of membranes. It also takes part in controlling the activity of many key metabolic enzymes. The above information shows that calcium is of both structural and physiological importance for plants [59].

The obtained test results show that in most cases the calcium content in the compost is higher than the content in the sewage sludge. The addition of fly ash contributes to higher calcium content in composts.

Magnesium is an important plant nutrient, one which is necessary for the construction of, among other substances, chlorophyll. Magnesium deficiency in plants affects their negative growth and yield. This component participates in the process of photosynthesis and is an activator of many biochemical and enzymatic processes. In addition, it participates in the synthesis of proteins and fats and has a positive effect on the accumulation of sugar in the root. Importantly, magnesium reduces the susceptibility of plants to diseases, especially to tassel, and at the same time stimulates the yield-forming effect of nitrogen [60,61].

The obtained test results show that in most cases the magnesium content in the compost is higher than the content in the sewage sludge. The addition of fly ash contributes to higher magnesium content in composts.

The ratio of carbon to nitrogen (C/N) in the tested compost samples ranged from $1:23 \div 32$, with an average value of 1:29.

3.3. Heavy Metals in Compost

Restoring to the soil the components contained in sewage sludge is right not only from the economic point of view, but also for maintaining or restoring ecological balance. Often, the organic and mineral composition of sewage sludge is comparable to the soil's organic substance, called humus, which makes it possible to use it in nature [62]. However, in addition to checking the content of nutrients and fertilizing substances in the sewage sludge, it is also extremely important to test for the presence of heavy metals in it.

The toxic effect of heavy metals consists in causing disturbances in the enzymatic system of living organisms, which in turn leads to changes during biochemical and physio-

logical processes. In extreme cases, these processes cause the death of cells and tissues by reducing the permeability of cell membranes and limiting the rate of photosynthesis [63].

Heavy metals are present in various amounts and forms in sewage sludge and fly ashes. Their mobility depends on the fraction of the solid matrix, but may vary depending on the pH of the soil on which they are applied. Heavy metals are generally toxic to plant, animal and human organisms. They can reach the level of phytotoxicity, and thus cause inhibition of plant growth. In addition, fertilization of soils with sewage sludge with a high level of heavy metals may also cause a decrease in its enzymatic activity [64].

Table 3 shows the concentrations of heavy metals in the composts, sewage sludge and fly ashes used for the tests. Analyzing the presented data, the content of heavy metals (except mercury) in the fly ash is significantly higher than their content in the sewage sludge. The addition of fly ashes to the composted material may therefore result in higher concentrations of metals in the compost. At the same time, studies of heavy metal concentrations in water extracts showed a lower mobility of the metals contained in the ashes. This phenomenon is beneficial from the point of view of environmental impact.

Table 3. Comparison of the results of tests of the content of heavy metals in compost and their permissible content specified in the Polish Regulations.

Indicator	Unit	Sewage Sludge	Fly Ash	Compost—Weight Proportions: Straw:Sewage Sludge:Fly Ash					[6]	[55]
				1:10:0	1:10:0.5	1:10:1	1:10:1.5	1:10:2		
Chromium	mg/kg d.m.	32.4 ±4.9	412 ±62	35.0 ±5.25	142.0 ±21.3	203.0 ±30.4	251.0 ±37.6	282.0 ±42.3	500.0	100.0
Chromium (water extract)	mg/L	<0.003	0.01 ±0.002	0.024 ±0.004		<0.003		<0.003		
Lead	mg/kg d.m.	30.0 ±3.0	107 ±11	27.2 ±2.7	58.2 ±5.8	64.9 ±6.49	76.2 ±7.6	81.5 ±8.2	750.0	140.0
Lead (water extract)	mg/L	<0.01	<0.01	<0.01	<0.01			<0.01		
Cadmium	mg/kg d.m.	0.817 ±0.123	3.93 ±0.39	0.243 ±0.0364	2.060 ±0.309	1.47 ±0.22	1.50 ±0.225	2.46 ±0.369	20.0	5.0
Cadmium (water extract)	mg/L	0.021	<0.0005	<0.0005		<0.0005		<0.0005		
Copper	mg/kg d.m.	299.0 ±44.9	598 ±60	259.0 ±38.9	447.0 ±67.1	488.0 ±48.8	519.0 ±77.9	544.0 ±81.2	1000.0	-
Copper (water extract)	mg/L	0.946	0.214 ±0.043	0.384 ±0.077		0.242 ±0.048		0.254 ±0.051		
Nickel	mg/kg d.m.	19.3 ±2.9	126 ±13	15.2 ±2.3	59.7 ±8.9	62.5 ±9.38	73.0 ±10.9	94.3 ±14.1	300.0	60.0
Nickel (water extract)	mg/L	0.149	0.01 ±0.002	0.104 ±0.021		0.04 ±0.008		0.052 ±0.01		
Mercury	mg/kg d.m.	0.33 ±0.066	0.09 ±0.02	0.210 ±0.042	0.380 ±0.076	0.220 ±0.044	0.190 ±0.038	0.200 ±0.040	16.0	2.0
Mercury (water extract)	mg/L	<0.0005	<0.0005	<0.0005		<0.0005		<0.0005		
Zink	mg/kg d.m.	825.0 ±124	3159 ±474	828.0 ±124	1595.0 ±239	2010.0 ±302	2255.0 ±338	2346.0 ±352	2500.0	-
Zink (water extract)	mg/L	1.08	0.591 ±0.118	0.404 ±0.081		0.388 ±0.078		0.36 ±0.072		

Relating the concentrations of heavy metals in the obtained composts to the limit values set for the use of municipal sewage sludge in agriculture and for the reclamation of land for agricultural purposes provided in the Regulation of the Minister of the Environment on Municipal Sewage Sludge [7], it can be stated that there are no violations.

Comparing the concentrations of heavy metals in the obtained composts to the limit values given in the Regulation of the Minister of Agriculture and Rural Development on the Implementation of Certain Provisions of the Act on Fertilizers and Fertilization [55], there are noticeable exceedances for two metals, i.e., chromium and nickel. In the light of the applicable regulations, it can be concluded that the concentration of these two metals in

the fly ash is a factor limiting the amount of fly ash that can be added to the compost mass. Figure 5 shows the concentration of chromium and nickel in the substrates and samples of the resulting compost.

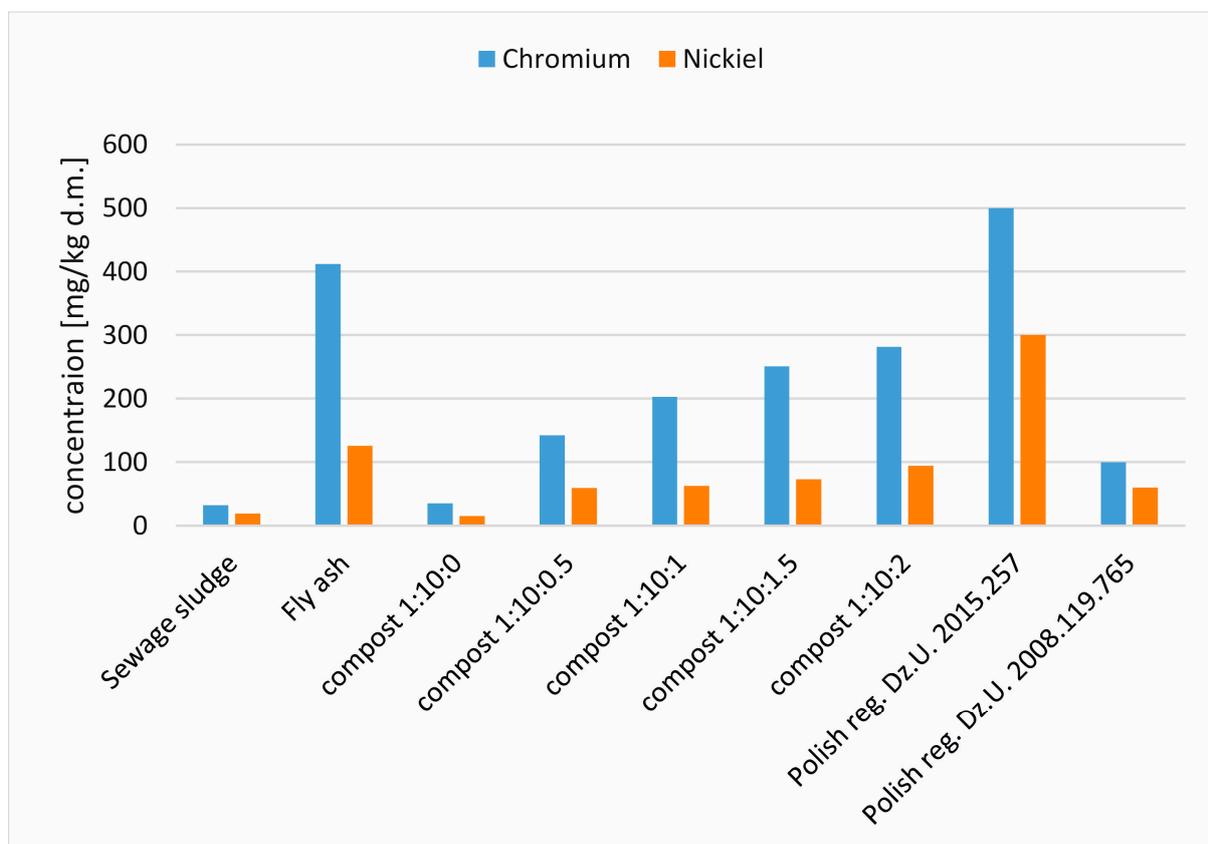


Figure 5. Chromium and nickel concentrations in substrates and composts using straw as a structural material—average of three replications.

3.4. Water Extracts from Compost Mass

A negative impact on the natural environment may be caused primarily by excessive loads of pollutants getting through into surface and underground waters. They cause deterioration of the quality of these waters.

Table 4 lists the BOD₅ and COD concentrations for water extracts from the used substrates and the obtained composts. The test results show that the content of BOD₅ and COD in water extracts from composts is lower than in water extract from sewage sludge. The concentrations of these indicators in water extracts from fly ash are trace, below the determination threshold. The concentration of COD is on average several times higher than BOD₅, which proves that the solution is dominated by organic substances that are not susceptible, or not very susceptible, to biochemical decomposition. The observed concentrations of BOD₅ and COD are significantly lower than in animal fertilizers, where the BOD₅ of pig manure is 4890 mg O₂/L and the COD is 13,080 mg O₂/L [54].

Table 4 summarizes the concentrations of biogenic indicators in water extracts. The obtained test results show that the concentration of total nitrogen in water extracts from composts is lower than in the water extract from sewage sludge. The TKN content in water extracts decreases with the increase of the fly ash content in the compost. This indicates a decrease in the availability of nitrogen for plants. This confirms scientific reports that an increase in the pH of the compost mass may affect the losses and immobilization of nitrogen [40,42,65]. The concentrations of nitrate and nitrite nitrogen in water extracts from the analyzed samples showed insignificant values.

In the tests carried out, a high temperature of the compost of 67 °C was obtained. It can therefore be assumed that it will be sufficient to remove parasite eggs in the event of their occurrence [67]. According to Mininni et al. [5] composting is the best way to remove pathogens.

3.6. Model of Sewage Sludge Composting Process with Ash Addition

From the point of view of the impact of the obtained composts on the environment, the limiting factor is the content of heavy metals. Fly ash is a source of many fertilizing ingredients, but at the same time, it can contain high concentrations of heavy metals. Based on the results, heavy metals that may have the greatest impact on the breach of the environmental requirements according to the Regulation on the Implementation of Certain Provisions of the Act on Fertilizers and Fertilization [55] are chromium and nickel. Therefore, the characteristics of changes in the concentrations of chromium and nickel in the compost depending on the added dose of fly ashes refining the compost mass were determined; the models of changes in the concentrations of these metals are presented in Figure 6.

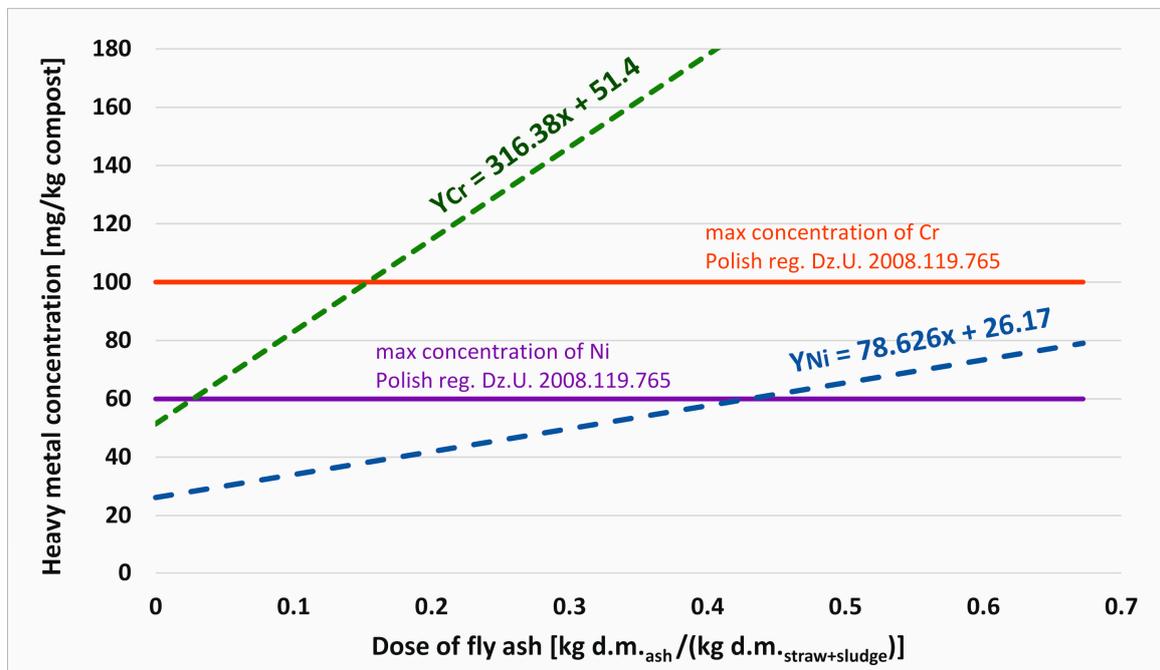


Figure 6. Models of the effect of fly ash dose on the concentration of chromium and nickel in the dry mass of compost (structural material—straw).

The determined characteristics of concentration changes made it possible to determine the equations for increasing the concentration of a given heavy metal in the compost and to determine the maximum doses for which the values of its concentration in the compost will not be exceeded (Figure 6). For chromium, the relationship between the concentration and the dose of added fly ash was defined as:

$$Y_{Cr} = 316.38 \cdot x + 51.4$$

For nickel, the relationship between the concentration and the dose of added fly ash was defined as:

$$Y_{Ni} = 78.626 \cdot x + 26.17$$

where:

x —Ash dose [kg d.m._{ash} / (kg d.m._{straw+sludge})].

Y_{Cr} —Chromium concentration [mg Cr / (kg d.m._{compost})].

Y_{Ni} —Nickel concentration [mg Ni / (kg d.m._{compost})].

Therefore, the maximum doses of ash added to the compost mass, not exceeding the maximum concentrations of heavy metals provided for in the Regulation on the Implementation of Certain Provisions of the Act on Fertilizers and Fertilization [55] should be:

For chromium with straw as a structural material:

$$-0.154 \text{ kg d.m.}_{\text{ash}} / (\text{kg d.m.}_{\text{straw+sludge}}) = 154 \text{ kg d.m.}_{\text{ash}} / (\text{ton d.m.}_{\text{straw+sludge}}).$$

For nickel with straw as a structural material:

$$-0.430 \text{ kg d.m.}_{\text{ash}} / (\text{kg d.m.}_{\text{straw+sludge}}) = 430 \text{ kg d.m.}_{\text{ash}} / (\text{ton d.m.}_{\text{straw+sludge}}).$$

4. Conclusions

It is possible to use fly ashes from sewage sludge combustion for compost mass as an additive for improving compost.

The maximum temperature of the compost mass was around 67 °C at the 6th day of the process.

Fly ash has an alkaline reaction, and its addition increases the pH of the composted material. In the tested range of ash addition, no significant negative effect on the efficiency of the process was observed. The high pH of the composted material may inhibit the effectiveness of biochemical processes, which would result in the extension of the composting time and a limit to the maximum temperatures obtained in the compost mass.

The addition of fly ash affects the growth of fertilizing substances in the compost, in particular, calcium and phosphorus.

Microbiological tests did not show the presence of indicator microorganisms in the tested compost masses.

The increase of fly ash in the compost reduces the concentration of nutrient indicators in the water extract, due to the reduction of their leaching from the compost mass.

However, comparing the concentrations of heavy metals in the obtained composts to the limit values set out in the regulations for organic and organic-mineral fertilizers, their use is limited mainly due to the presence of two heavy metals, chromium and nickel. According to the current regulations, it can be concluded that the concentration of these two heavy metals in the fly ash is a factor determining the amount of fly ash in the compost mass. The most favorable weight proportions of substrates, as limited by higher concentrations of chromium in the fly ash, are as follows:

Whey straw : Sewage sludge: Fly ash
410 kg d.m. : 590 kg d.m. : 154 kg d.m.

Fly ashes can be used to obtain the declared content of fertilizing substances in compost produced from sewage sludge. Each time, chemical analyzes of substrates should be performed in order to properly balance them in the compost mass.

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