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Research paper

The seasonal changes of heavy metal content as limiting factor of sewage sludge re-use as fertilizer ingredient, based on data from WWTPs in Poland

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Abstract: In Europe, half of the amount of sewage sludge from wastewater treatment plants (WWTPs) is used in agriculture, in Poland about 27%, respectively. Fertilizing products (compost or organic-mineral fertilizers) produced from sewage sludge are promise alternative for directly use of no-treated waste. The paper presents seasonal changes of Cu, Pb, Ni, Zn, Cr, Cd and Hg content in sewage sludge from different regions of Poland collected during one year. It was noted wide range of the metal amount in dependence on season and WWTPs capacity. The Cu concentration was from 0.4 to 784 $mgkg^{-1}$ d.m., Ni from 0.25 to 1281 $mgkg^{-1}$ d.m., Cd from 0.005 to 14.85 $mgkg^{-1}$ d.m., Pb from 0.11 to 306.2 $mgkg^{-1}$ d.m., Hg from<0.001 to 2.3 $mgkg^{-1}$, Cr from 0.23 to 854 $mgkg^{-1}$ and Zn from 11 to 4669 $mgkg^{-1}$, respectively. It has been shown that Cu, Cd, Pb, Hg and Cr content did not exceed the permissible levels for agricultural purposes according to Polish law. There was no clear correlation between the heavy metal amount and the WWTP capacity, expressed as population equivalent (PE). However some increase trend in the case of Cu, Ni, Hg and Cr concentration at the higher PE value was found. It has been concluded that in spite relatively low level of heavy metals in Polish WWTPs, it may be limiting factor for production high quality fertilizers on the base of sewage sludge.

Keywords: sewage sludge, heavy metals, seasonal changes, population equivalent (PE)

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1. Introduction

Huge amounts of sewage sludge are produced annually by municipal wastewater treatment plants (WWTPs) around the world [1], and its disposal is a serious challenge [2]. It is estimated that the global production of sewage sludge reaches the amount 200 million Mg per year [3], and Europe, North America, and East Asia are the main "producers" of sewage sludge in the world [1]. There is a very large variation in the sludge mass among European countries [4]. Also in Poland, an increase of sewage sludge amount generated in municipal WWTPs has been observed for many years [5]. This is mainly due to the increase in the volume of sewage entering the treatment plant as a result of the expansion of sewage networks [6] and the more effective technologies used for wastewater treatment [5]. The amount of sewage sludge generated in Poland in 2021 amounted to 699.670 Mg of dry mass [7]. The most commonly ways of sewage sludge management are – the use in agriculture for the cultivation of plants not intended for consumption and fodder (marked as R10 according to Polish law), use for the production of compost (marked as R3 according to Polish law) and thermal processing (marked as D10 according to Polish law). There are many other sludge technologies that exploit its energy or chemical value, mainly thermal processes [8]. However, in the case of municipal sewage sludge the application on surface [9] is frequently better alternative than thermal processing [10]. There are several advantages of such management, which leads to matter recovery and contributes to the development of a circular economy [11]. A significant content of organic matter in sewage sludge (about 50% of their solid fraction) can improve the physical, chemical and biological properties of the soil and lead to significantly increase yields when used as fertilizer [3]. Organic matter plays an important role in soil aggregation. It improves soil porosity [12] and increases water retention [13].

The addition of organic matter promotes the decomposition of soil substances and establishes a microbiological balance [14]. The sludge contains nutrients beneficial for fertilization: organic matter, fertilizing elements and microelements (iron, manganese, copper, molybdenum) [13] necessary for plants [15], which can contribute to increasing plant production [16] and improve soil quality [17–21]. Sewage sludge applicated on/to the agriculture soil modifies its physicochemical and biological properties [22], and finally after appropriate processing, can replace chemical fertilizers, which may cause acidification of the soil [13]. These benefits can be limited by the presence of contaminants [19], mostly heavy metals, due to their inactive nature, are stable for long time [23]. There are however several other contaminants, which may occur in sewage sludge – chemicals (trace elements – Se, Ag, Ti; nanoparticles; polyaromatic hydrocarbons; polychlorinated biphenyl; perfluorinated surfactants, polycyclic musks, siloxanes, pesticides, phenols, sweeteners, personal care products, pharmaceuticals, benzotriazoles) and biological (*Legionella, Yersinia, Escherichia coli* O157:H7) [24, 25].

It is commonly believed that sludge from large wastewater treatment plants, due to the high content of pollutants (including heavy metals), should be thermally treated. The sewage sludge from smaller treatment plants can be used for agricultural purposes or for reclamation of degraded soils. Results from Germany indicated that the total content of many metals in sewage sludge has significant decreased in last decades. Comparative data from 1977–2012 showed decrease ranging from 64 to 95%. Only in the case of copper this value was smaller and amounted to 22% [24]. Therefore, if sewage sludge are used for agricultural purposes, in the case of heavy metals it is important to know the total content of elements as well as the form in which they occur and their possible variability during the year. Other authors suggest that the determination of metal forms allows for estimation of assimilability of trace elements by plants [26–28].

2. Objective

Sewage sludge generated in sewage treatment plants is characterized by soil-forming and fertilizing properties. However, their improper management may lead to wastage of resources and serious pollution of the ground and water environment. One aspect of assessing the impact of sewage sludge on the environment is the content of heavy metals. Even though this is a standard analysis performed in sewage treatment plants, extensive comparative analyzes are rarely performed [15, 27].

The aim of the study was to determine the content of heavy metals (such as Cu, Pb, Ni, Zn, Cr, Cd and Hg) depending on the amount of sewage reaching the treatment plant, expressed as a population equivalent number (PE). During one year, changes in the concentration of heavy metals were monitored, which also allowed the influence of the seasonality factor to be determined. This is the first comparative analysis in many years based on data obtained from several dozen facilities throughout the country.

3. Methods

3.1. Data collection

In the selected WWTPs, sewage sludge tests with reference methods were analysed with a frequency depending on the WWTP load, expressed as the number of population equivalent (PE), i.e., at least:

– once every six months – with PE up to 10,000, i.e., at least twice a year;
– once every four months – with PE over 10,000 to 100,000, i.e., at least 3 times a year;
– once every two months – with PE over 100,000.

The analysis covered data obtained from selected WWTPs belonging to all size groups expressed as PE. Thus, these facilities differed in the volume of inflowing and treated sewage, as well as in the technological solutions implementing the treatment stages. Mechanical and biological treatment with the use of activated sludge dominates among the examined objects, as shown in Table 1.

Table 1. Estimation of sewage sludge treatment and disposal in analysed WWTPs

The size of the WWTP expressed in PE	**<2000**	**2000–9999**	**10000–14999**	**15000–99999**	**>100000**
Number analyzed WWTPs	5	19	4	22	14
Sewage sludge treatment:					
– aerobic stabilization	2	3	–	4	–
– anaerobic stabilization	–	–	1	8	10
– thickening, dewatering	3	17	1	16	14
– solar drying	–	–	–	1	1
Sewage sludge management:					
– composting	–	4	–	3	1
– use in agriculture,	–	12	2	11	4
– use for reclamation for agriculture purposes	3	2	1	1	2
– use for growing plants from which compost is produced	1	–	–	4	4
– thermal treatment	–	–	–	1	1
– transfer to an external company	1	1	–	–	–
– process R3[1]	–	3	1	5	4

[1] According to Polish law process R3 is concerning on recycling or recovery of organic substances that are not used as solvents (including composting and other biological conversion processes).

3.2. Data analysis

The Cu, Cd, Ni, Hr, Cr, Zn and Pb concentration in dry matter [d.m.] of sewage sludge was used for the analysis. The results obtained from WWTPs were grouped accordingly to number of population equivalent (PE) and compared with the permissible levels according to the applicable law regulations.

For statistical analyses and determination of relationship between data, two types of formula were used: Pearson correlation coefficient and R^2 determination coefficient.

4. Results and discussion

4.1. Selected problems of sewage sludge in legal regulations

For the analysis and assessment of the quality of sewage sludge, the Polish Regulation of the Minister of Environment of February 6, 2015 on municipal sewage sludge [29], which defines the permissible content of heavy metals in municipal sewage sludge before its use and Regulation of the Minister of Climate and Environment of December 31, 2021 amending the regulation on municipal sewage sludge [30] and Directive 86/278/EEC [31] were used.

Moreover, the permissible levels of metal content were taken into account according to global regulations, as shown in Table 2.

Table 2. Comparison of permitted heavy metal content in municipal sewage sludge between Polish, EU, China and US regulations

Metals	**The metal concentration [mgkg^{-1} d.m.]**								
	Polish regulation [2015, 2021]			**EU Directive 86/278/EEC**	**Proposed changes ENV/E.3/LM**		**GB18918 -2002[1]**		**EPA US[2]**
	in agriculture and for reclamation for agriculture purposes	**for reclamation of degraded terrains for non-agriculture purposes**	**other**	**in agriculture**			**pH ⩽ 6.5**	**pH ⩾ 6.5**	
	2015	**2015**	**2015**	**–**	**2015**	**2025**	**2002**		**1993**
Cd	20	25	50	20–40	5	2	5	20	85
Cu	1000	1200	2000	1000–1750	800	600	250	500	4 300
Ni	300	400	500	300–400	200	100	100	200	420
Pb	750	1000	1500	750–1200	500	200	300	1000	840
Zn	2500	3500	5000	2500–4000	2000	1500	500	1000	7500
Hg	16	20	25	16–25	5	2	5	15	57
Cr	500	1000	2500	–	800	600	600	1000	–

[1] General Inspection and Quarantine of China (2009) Standardization administration of China, GB 24188-2009 Quality of sludge from municipal wastewater treatment plant Beijing. There are monitored: As, Cd, Cr, Cu, Hg, Ni, Pb, Zn.

[2] EPA U S. (1993) Code of Federal Regulations Title 40, Part 503. U. S. There are monitored: As, Cd, Cu, Ni, Pb, Zn, Hg.

Sewage Sludge Directive 6 (Council Directive 86/278/EEC) [31], was to ensure the proper use of sewage sludge in agriculture and prevent its harmful impact on soil, vegetation, animals and humans. It requires that sludge be used in such a way as to take into account the needs of plants and not to deteriorate the quality of soil, surface water and groundwater. The use of the potential of sludge to recycle valuable plant nutrients and as an effective soil enrichment will help not only in the sustainable management of this waste, but also in minimizing the negative effects associated with its traditional disposal [32, 33].

Currently, the sludge directive is considered outdated and does not ensure the safety of using sludge on agricultural land [21]. In the ongoing consultations, many public authorities, associations and entrepreneurs from all over the Union opted for the need to amend the oldest legal act. Among the proposals there is a postulate that sewage sludge should be used in such a way as to take into account the demand for nutrients [34]. There are a number of points and issues that require careful analysis, clarification and inclusion in new regulations. This applies mainly to new pollutants, including pharmaceuticals [35] and microplastics, but also to organic pollutants [36], including PAHs [37] and traditionally determined in sewage sludge, e.g., heavy metals. Moreover, in the USA there is no requirement to analyse sewage sludge for the presence of organic chemicals [38]. In Canada, no restrictions have been proposed for most trace organic pollutants [39]. Sewage sludge from domestic wastewater treatment is believed to be a diffuse point source of heavy metal contamination [40]. Therefore, some European countries introduced stricter requirements in relation to those contained in EU regulations [21]. In the case of heavy metals, their excessive accumulation in the soil may threaten its fertility [41], and in extreme cases lead to the degradation of the entire soil ecosystem [42]. The scale of the problem is influenced, among others, by the composition of sludge, soil properties, or the method of sewage sludge treatment and utilization [43], therefore it is important to carefully assess these parameters before deciding on the use of sludge or developing recommendations regarding the dose [22] and the frequency of their application to the soil. The content of heavy metals in the sewage sludge is closely related to the industrial characteristics of the region. It has been noted that higher levels of these metals occur in places with higher industrial development [44], including Hg and Cd content, which depends on the coal industry [5]. Particular attention should be paid to cadmium, which is one of the most toxic heavy metals with teratogenic and mutagenic effects. This element inhibits the activity of soil bacteria and plant growth, and above all, it poses a serious threat to food safety [42]. In many countries, the revision of standards for sludge management is mentioned [46].

The current Polish National Waste Management Program 2022 indicates that one of the priorities in the management of municipal sewage sludge should be organic recycling, including composting of sludge with other waste to obtain material after the composting process used for fertilization, as well as mineral recycling with phosphorus recovery.

The revised Directive should establish a hierarchy of desirable uses for sludge that encourages preferential use in brownfield restoration, forestry and energy crops, before considering use on land producing food or feed [47].

The issue of using sewage sludge in agriculture is very complex and carries many risks. To avoid these risks, it is important to develop common European legislation as well as national legislation. Therefore, it is necessary to take into account issues related to the increase in the rate of sludge production and ways to reduce them, emerging pollutants and their fate, recovery of carbon, phosphorus, nitrogen and energy for reuse as the most current research trends, as in China [48]. It is necessary to have appropriate safety measures to prevent possible contamination of surface and groundwater and to avoid toxic effects on soil, plants, animals and people.

4.2. Management methods in relation to the sewage sludge generated

In total, in 2021, the analyzed treatment plants generated 73,514.57 Mg dry matter of sewage sludge requiring management. The largest share was the use in agriculture, up to 36.8%, and the R3 recovery process, amounting to 27.2%. A negligible share was recorded in the case of composting – 2.8% and thermal transformation – 1.3%. Taking into consideration the share of the different management methods in dependence on population equivalent (PE) it was found that the sewage sludge was used in agriculture mainly in small and medium-size wastewater treatment plants (Fig. 1). At the largest analysed facilities (> 100000 PE), the other way of sewage sludge management was preferred – thermal treatment, utilization by external companies.

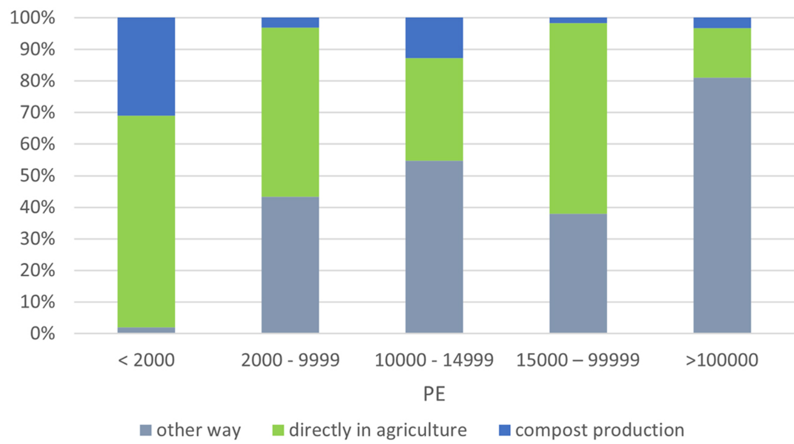


Fig. 1. The share of the management of sewage sludge in dependence on population equivalent (PE)

The chemical form of heavy metals determines their mobile and immobile mobility [49], and only sequential analysis allows us to estimate the percentage of metals in the mobile form, available to plants [50]. Particularly important are metals associated with the water-soluble and exchangeable fraction and the fraction of metals bound in carbonates and labile organic compounds. The research by Jakubus and Czekała [51] showed that the tendency to create such fractions increased in the following order: Cu<Cr<Zn<Cd<Ni.

4.3. The range of the metal content in sewage sludge in dependence on PE

The obtained results (Table 3, Fig. 1) indicate quite wide ranges of the content of individual elements in the dry mass of sediments and the calculated standard deviations, which are a measure of the dispersion of the obtained data.

One of the main sources of higher levels of heavy metal contamination in wastewater are industrial wastewater [2], but also rainwater and meltwater intakes in the sewage system [52]. Therefore, the content of selected heavy metals in sewage sludge can be varied. Generally the total concentration of heavy metals in sewage sludge decreased significantly in last decades [24]. However, there are no comparative studies showing the dependence of the content of heavy

Table 3. The range of concentration of selected heavy metals in dependence on PE

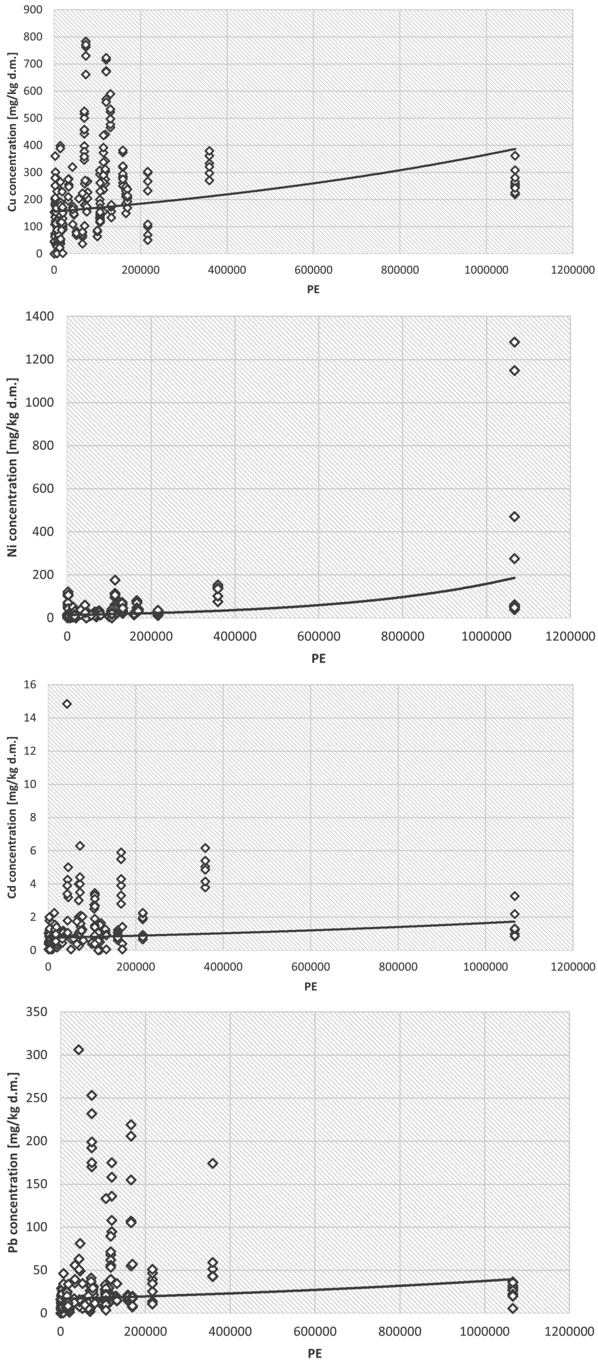
Metals	**Ranges of values and average contents [mgkg^{-1} d.m.] in sewage sludge at PE**				
	<2000	**2000–9999**	**10000–14999**	**15000–99999**	**>100000**
Cd	< 0.05 – 1.02	0.05–1.8	0.4–2.26	0.05–14.85	0.025–6.17
	0.52	0.75	0.88	2,02	1.57
Cu	0.4–154	1.0–360	35.8–398	2.9–784	50.6–723
	65.3	155.1	140.6	208.6	271.9
Ni	0.25–111	0.75–121	4.22 – 119.2	0.4 – 60.22	6.25–1281
	21.4	19.5	16.5	18.6	60.8
Pb	0.11–16.1	0.3–46.2	6.92–19.4	1.0–306.2	3.65–175
	7.4	14.1	12.9	42.2	41.5
Zn	3.06–10506	2.6–2100	134–809	11–1414	218–4669
	1685	596	400	725	1195
Hg	< 0.005 – 0.23	< 0.01 – 0.98	0.04–0.25	0.074–2.01	0.05–2.0
	0.123	0.185	0.163	0.623	0.592
Cr	< 0.5 – 139	0.23–89.4	6.46–129	2.9–112.84	9.03–854
	28.7	22.1	36.3	28.1	88

metals on the equivalent number of inhabitants (size of the sewage treatment plant). Larger treatment plants are believed to contain higher concentrations of metals, which is related to the presence of industrial wastewater (as mentioned earlier). Higher concentrations of metals in sludge may also be related to the way they are processed in the technological cycle of sewage treatment plants. Processes such as anaerobic digestion, commonly used in medium and large facilities, contribute to a significant reduction in sludge mass. In result, heavy metals due to no biodegradation are concentrated [53] and their concentration is higher.

Cadmium (Cd) content ranged from 0.025 to 14.85 mgkg^{-1} d.m. In the case of samples from most small and medium-sized WWTPs, the metal concentration varied 0.05-2.26 mgkg^{-1} d.m., and the average values did not exceed 2 mg Cdkg^{-1} d.m.

Copper (Cu) content ranged from 0.4 to 784 mgkg^{-1}, usually up to 200 mg. Sewage sludge from municipal wastewater usually contains low concentration of this metal, while the inflow of industrial wastewater can cause its significant increase in the sludge. This was reflected in the obtained data, because in sludge from treatment plants of large and very large industrial cities, the copper content is higher. However, all samples did not exceed the level allowed for use in agriculture, which is equal 1000 mg kg^{-1} d.m. Cu.

The content of nickel (Ni) in the sewage sludge was quite different (0.25–1281 mgkg^{-1} d.m.), with an average value equal 27.36 mgkg^{-1} d.m. Only four facilities had results exceeding 100 mgkg^{-1} d.m., which can be connected with the developed industry in the given cities. In the case of one WWTP high results of nickel content was obtained, and it was one of the smallest treatment plants. It be found that wastewater reached this type of WWTP was heavily contaminated with nickel, chromium and zinc compounds (Fig. 2).


Cu concentration [mg/kg d.m.]
PE
Ni concentration [mg/kg d.m.]
PE
Cd concentration [mg/kg d.m.]
PE
Pb concentration [mg/kg d.m.]
PE

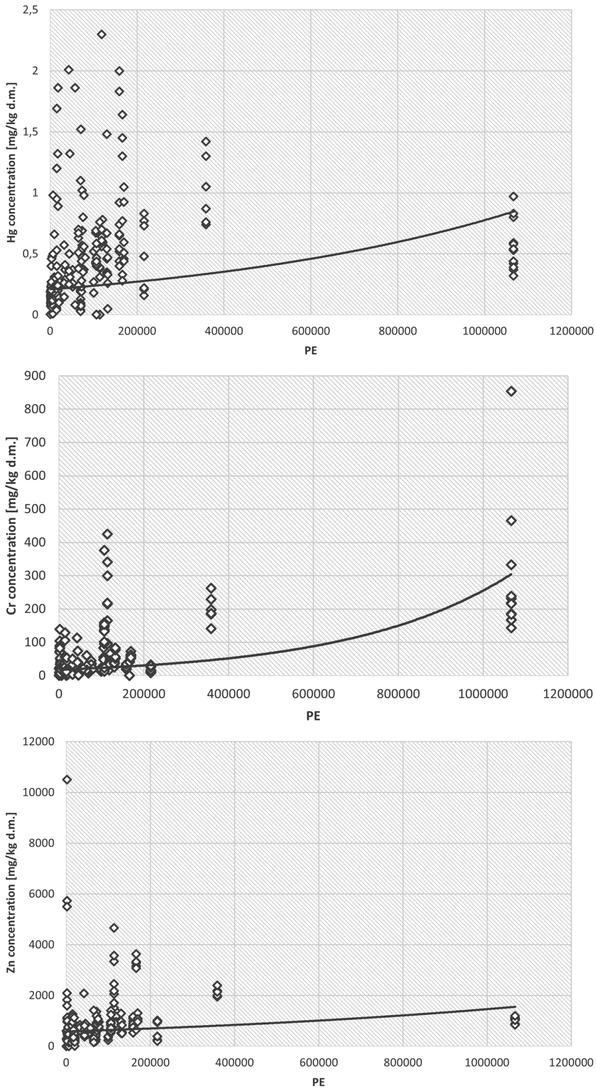


Fig. 2. The range of selected heavy metals content in sewage sludge from analysed WWTPs in Poland in dependence on population equivalent (PE) value

Lead (Pb) contents in sewage sludge ranged from 0.11 to 306.2 mgkg^{-1} d.m., with an average of 23.62 mgkg^{-1} d.m.

The content of zinc (Zn), which is a metal commonly found in the environment, in the examined samples was the highest as compared to other metals and was characterized by a very large diversity (2.6–10506 mgkg^{-1} d.m.). It should be noted, however, that the maximum value was obtained only once from the given range. Among the WWTPs from the group of PE below

2000, there was one facility mentioned in the analysis of nickel content, whose sewage sludge is characterized by an exceptionally high content of zinc, amounting to 10506 mg/kg d.m. The values obtained from the previous determinations were about 50% lower, however, it is still two-fold more for the permissible zinc content in sewage sludge when used in agriculture. Further analysis by the authors showed that in the sludge samples taken from this WWTP in 2022, a zinc content was much lower and equal 279 $mgkg^{-1}$ d.m. The cause of significant short increase in Zn concentration may have been connected with local incidentally source.

Mercury (Hg) content during 2021 ranged from 0.005 to 2.3 $mgkg^{-1}$ d.m. reaching an average value 0.3372 mg/kg d.m., which proves a low level of sewage contamination with mercury compounds.

Chromium (Cr) content in sewage sludge ranged from 0.23 to 854 $mgkg^{-1}$ d.m., with an average of 40.64 $mgkg^{-1}$ d.m. and are similar to the copper content.

The concentrations of individual metals were compared with the maximum values allowed by the applicable regulations and with the results of analyzes of sewage sludge from literature reports. It was shown that the concentrations of Cu, Cd, Pb, Hg and Cr did not exceed, and sometimes were several times lower than, the limit values of heavy metals in agricultural sludge. The analysed samples of sewage sludge contained, on average, about 12 times less chromium, 20 times less lead, 17 times less cadmium, 6 times less copper and 47 times less mercury than the limit values specified in the regulation.

Few times, nickel and zinc values exceeding permissible level have been recorded. In the case of zinc from the all analyzed objects, three WWTP clearly exceeded the permissible content of 2500 mg Zn kg^{-1} d.m. In one WWTP, after six analyses carried out during the year was found that in five cases the permissible level were exceeded by an average of 32%; while in the other WWTP's there were three exceedances, but by an average of 54.5%. In the third facility, three tests were carried out during the year and in all case very high zinc contents were recorded. Considering the nickel content, the limit value (300 $mgkg^{-1}$ d.m.) was found to be exceeded in two determinations for the largest of the analyzed treatment plants.

Taking into consideration analysed elements individually, in the case of copper, nickel, mercury and chromium, there was an upward trend in the content of these components with increasing PE value (Fig. 2). Although correlation between the minimum and maximum content of heavy metals and the size of the WWTP expressed as population equivalent (PE) was not clear. Moreover, it was observed that sewage sludge from small and medium-sized WWTTPs more frequently met the appropriate quality standards for agricultural application.

As mentioned above, statistical analyses did not confirm that level of heavy metals in sewage sludge was dependent on population equivalent. Both correlation coefficient and R^2 value were low, which indicates that the observations are far from the model's predictions (Table 4). The lowest value of correlation coefficient was obtained in the case of Pb (below 0.1); the highest – in the case of Cr (0.6).

Table 4. The Pearson correlation coefficient and R^2 determination value on dependence selected heavy metals on PE

Coefficient	Cu	Ni	Cd	Pb	Hg	Cr	Zn
Pearson correlation	0.151	0.524	0.119	0.055	0.194	0.629	0.100
R^2 determination	0.023	0.273	0.013	0.003	0.036	0.394	0.010

4.4. The range of the metal content in sewage sludge in dependence on season

Taking into account the aspect of seasonality, attention was paid to changes in the content in the periods of determination. The greatest possibilities of analysis are provided by treatment plants from the group of PES above 100,000, for which determinations were made min. every two months, and in some cases monthly. For these WWTPs, the arithmetic mean content of heavy metals in January–February, May–June and July–August are arranged in the following order: Zn>Cu>Cr>Pb>Ni>Cd>Hg. The sequence and concentration ranges are consistent with previous studies of sewage sludge in Poland [27] and other countries [40, 54]. A certain deviation can be noticed in the autumn months, for which the following order of metal content was obtained: Zn>Cu>Ni>Cr>Pb>Cd>Hg. This may be a signal for changes in content influenced by the season.

Considering the average content of heavy metals in particular research periods, it can be concluded that in most cases (copper, nickel, chromium, zinc) the lowest content occurs in the months at the turn of spring and summer. At the same time, the highest contents of these elements were recorded in late autumn and early winter. The largest changes in metal content over 2021 were recorded for nickel, where an almost threefold increase was observed. In other cases, changes in the content of individual elements by an average of 20% were noted. Since for each component there may be different contents during the year and different trend lines (Fig. 3), it is certainly not possible to generalize seasonal changes in content and on this basis determine the suitability of sewage sludge [54].

Under the provisions of the Directive, the use of sludge in soils intended for the cultivation of fruit and vegetables, which are in direct contact with the soil and are eaten raw, is prohibited, and this prohibition applies for a period of 10 months before the harvest of these plants and during the harvest itself. This creates some kind of difficulty in getting rid of deposits on a regular basis. As already mentioned, the sludge with the best characteristics in terms of heavy metal content in most cases of the analyzed treatment plants was produced in May–June, so after the fields were used and the crops harvested, this sludge could be managed. However, in the case of sludge produced in the autumn and winter, it would have no chance of being used.

The analyzed data indicate in the vast majority of cases that the content of heavy metals in the sewage sludge does not limit the possibility of their use for land reclamation for non-agricultural purposes and in adapting land to specific needs resulting from waste management plans, spatial development plans or decisions on land development and development conditions, for the cultivation of plants intended for the production of compost, for the cultivation of plants not intended for consumption and for the production of food.

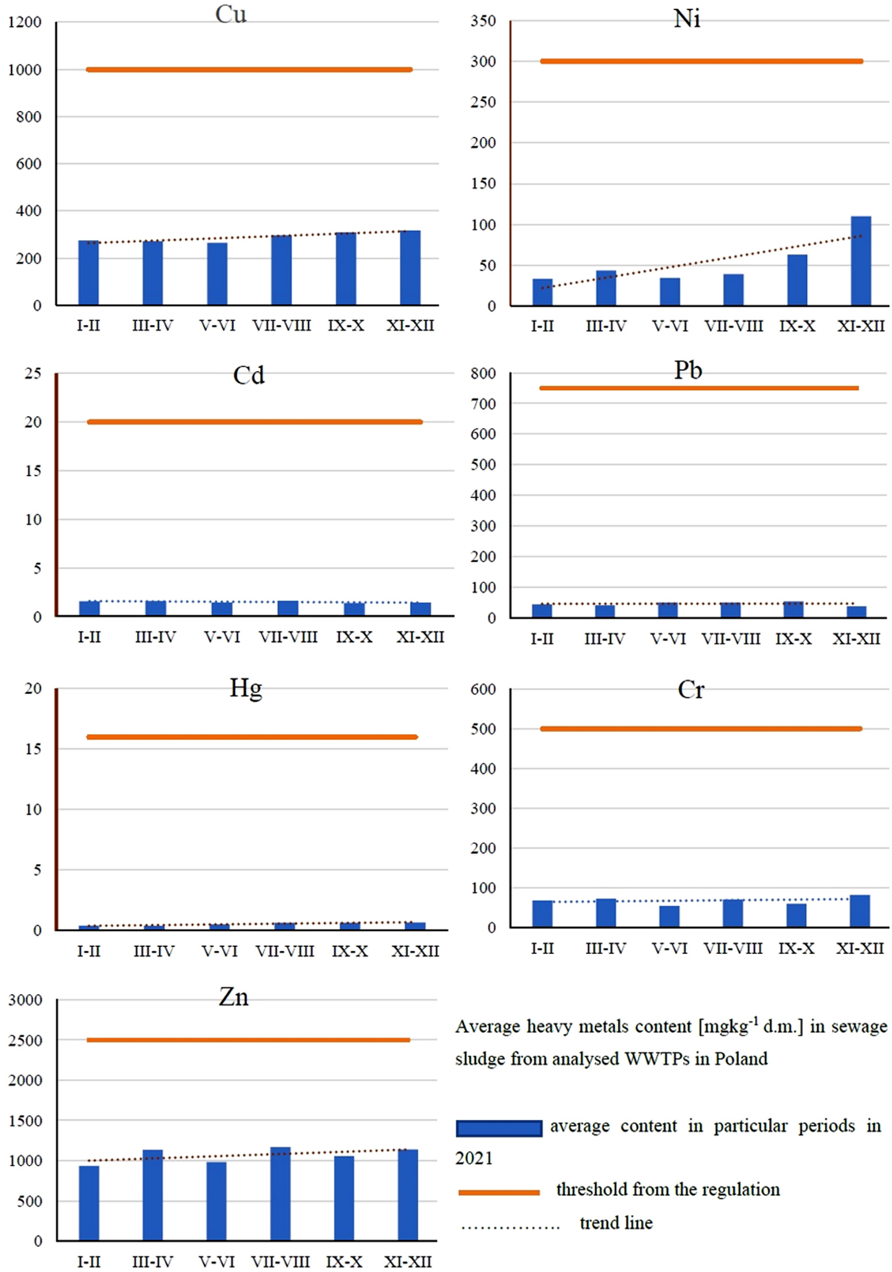


Fig. 3. Seasonal changes in the content of heavy metals in sewage sludge from analysed WWTPs in Poland (the orange line represents the threshold from the regulation, the dotted line indicates the trend line calculated on the base mean values)

5. Conclusions

Sewage sludge intended for use in agriculture should be treated as fertilisers, therefore the period of their introduction into the soil as well as soil testing should be taken into account [55]. They are carried out each time before directing a given batch of municipal sewage sludge for use on the ground. In addition, the bioavailability of heavy metals should be taken into account, so research should be conducted on the share of individual fractions of a given element simultaneously with the bioavailability of biogenic compounds by plants from this source. Total concentrations of heavy metals only inform about the degree of environmental pollution [26, 56], while a full assessment of their impact on soils includes mobility, bioavailability and ecotoxicity. The migration of metals in soil and absorption by plants depend on the type of metal, physicochemical properties of sewage sludge and soil, and plant species [50]. Attention should also be paid to the possibility of potentially higher accumulation of metals [57] when sludge from the months with the highest content is used repeatedly.

1. The total content of heavy metals in the analyzed sewage sludge does not preclude its agricultural use, because the obtained results were many times lower than the limit values set for the sludge used for this purpose.
2. Exceeding the permissible content of zinc and nickel in the sewage sludge from three sites prevents their natural use.
3. Analysis of the content of heavy metals in sewage sludge in terms of changes during the year showed that the greatest risk of contamination was recorded for nickel, chromium and mercury during the autumn and winter. The risk is definitely lower if the sewage sludge is used immediately after production and stabilized in the summer months.
4. Comparing the content of heavy metals in the tested samples of sewage sludge with the values specified in legal regulations, it can be concluded that in most cases they meet even the more stringent standards.
5. The content of the tested heavy metals in sewage sludge showed considerable variation.

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Sezonowe zmiany zawartości metali ciężkich jako czynnika ograniczającego ponowne wykorzystanie osadów ściekowych do produkcji nawozów na podstawie danych z oczyszczalni ścieków w Polsce

Słowa kluczowe: metale ciężkie, osady ściekowe, zmiany sezonowe

Streszczenie:

W Europie połowa ilości osadów ściekowych z oczyszczalni ścieków jest wykorzystywana w rolnictwie, w Polsce odpowiednio około 27%. Istnieje kilka przepisów prawa krajowego opartych na ramowej dyrektywie osadowej (86/278/EWG) ograniczających poziom zanieczyszczeń (głównie metalami ciężkimi) w przypadku bezpośredniego wykorzystania odpadów na powierzchni gleby. Jednak nowelizacja Dyrektywy Osadowej (i drastyczne obniżenie dopuszczalnych stężeń zanieczyszczeń) może ograniczyć, a nawet utrudnić bezpośrednie wykorzystanie osadów ściekowych do celów rolniczych. Stąd efektywne produkty nawozowe (kompost lub nawozy organiczno-mineralne) pochodzące z osadów ściekowych są dobrą alternatywą dla ponownego wykorzystania nieoczyszczonych odpadów. Na zawartość substancji toksycznych w osadach ściekowych mogą mieć wpływ różne czynniki, w tym obecność ścieków przemysłowych czy wielkość oczyszczalni. W pracy przedstawiono wyniki badań zawartości metali ciężkich (Cu, Pb, Ni, Zn, Cr, Cd, Hg) w osadach ściekowych analizowanych przez certyfikowane laboratoria, udostępnianych przez oczyszczalnie ścieków z różnych regionów Polski, pobranych w ciągu jednego roku. Uzyskane wyniki wskazały na bardzo szerokie przedziały zawartości poszczególnych pierwiastków w suchej masie osadu. Stężenie Cu mieściło się w zakresie od 0.4 do 784 $mgkg^{-1}$ s.m., Ni od 0.25 do 1281 $mgkg^{-1}$ s.m., Cd od 0.005 do 14.85 $mgkg^{-1}$ s.m., Pb od 0.11 do 306.2 $mgkg^{-1}$ s.m., Hg od <0.001 do 2.3 $mgkg^{-1}$, Cr od 0.23 do 854 $mgkg^{-1}$ i Zn od 11 do 4669 $mgkg^{-1}$. Wykazano, że zawartość Cu, Cd, Pb, Hg i Cr nie przekraczała dopuszczalnych poziomów użytkowania rolniczego zgodnie z polskimi przepisami. Nieliczne były przypadki przekroczenia ilości niklu i cynku. Nie stwierdzono wyraźnej korelacji między minimalną i maksymalną ilością metali ciężkich a wielkością oczyszczalni wyrażoną przez równoważną liczbę mieszkańców (RLM). Zaobserwowano jednak pewien trend wzrostowy w przypadku stężeń Cu, Ni, Hg i Cr przy wyższych wartościach RLM. Ponadto odnotowano sezonowe zmiany zawartości metali ciężkich w osadach ściekowych oraz sezonową zmienność poziomów poszczególnych pierwiastków. Stwierdzono, że pomimo stosunkowo niskiego poziomu metali ciężkich, może to być czynnikiem ograniczającym produkcję wysokiej jakości nawozów na bazie osadów ściekowych.

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