

Article

Risk Assessment of Soil Contamination with Heavy Metals from Municipal Sewage Sludge

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Featured Application: The work can be used to establish new guidelines for the natural use of municipal sewage sludge.

Abstract: Sewage sludge (SS) is a by-product of processes conducted during the treatment of wastewater. It can be used in many different ways. One of them is the use of SS in agriculture as an organic fertiliser, but the main criterion for such use is the heavy metals (HMs) content. Knowledge of the total content of HMs in SS does not translate into the danger it may pose. The toxicity of metals is largely dependent on their mobility. The mobility of SS from three different wastewater treatment plants (WWTP) of the Świętokrzyskie Voivodeship, which were characterised by an increased zinc content, was examined in this study. The aim of the study was to prove whether the high level of zinc in SS actually disqualifies the possibility of its natural use. Calculations were made for five environmental hazard indicators: the geoaccumulation index of heavy metals in soil (I_{geo}), potential environmental risk indicator (PERI), risk assessment code (RAC), environmental risk factor (ERF), and the authors' own environmental risk determinant (ERD) indicator. The obtained results show how important mobility analysis is when assessing the possibility of natural use of SS.

Keywords: heavy metals; sewage sludge; soil; risk assessment; speciation



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1. Introduction

Municipal sewage sludge is an unavoidable by-product of wastewater treatment. An increase in the amount of sewage sludge produced has been observed for many years. The amount of generated sewage sludge is only 1–3% of the volume of flowing sewage [1]. Nevertheless, in the case of improper management, sludge may pose a potential threat to the environment. Sewage sludge contains, among others, heavy metals and pathogenic organisms [2,3]. On the other hand, sewage sludge is of practical importance as it contains organic substances and biogenic elements [4]. Sewage sludge can be used in agriculture as a valuable source of nitrogen and phosphorus, for the production of compost, and for the reclamation of degraded areas [5,6]. The choice of the method of sewage sludge management is particularly dictated by its quantity and properties [7]. Moreover, it is subject to legal regulations. In Poland, the Act on Waste [8], the regulation of the Minister of the Environment on municipal sewage sludge [9] and the regulation of the Minister of the Economy on the criteria and procedures for allowing waste to be deposited in a given type of landfill [10] are in force in this respect. In Europe, the limits of heavy metals in terms of natural use are regulated by the Council Directive of 12 June 1986 on the protection of the environment, in particular of the soil, when sewage sludge is used in agriculture [11]; in the United States, it is the Code of Federal Regulations [12]; for China, the 2002 “Standard for Discharging Pollutants into Urban Wastewater Treatment Plants” [13]; and for areas in southern Africa, “Guidelines for the Utilisation and Disposal of Wastewater Sludge” [14]. The limits for the heavy metal content of sludge for natural use are shown in Table 1.

Table 1. Normative limit values for heavy metals in sewage sludge for natural use.

Metal	Permissible Values for Heavy Metals Intended for Natural Use, mg·kg ⁻¹ d.m.					
	Poland Regulation [8–10]	EU Directive 86/278/EEC [11]	Chinese Regulation GB 18918–2002 [13]		USA Regulation 40 CFR Part 503, 503.13 [11]	South African Guideline (Pollutant Class a) [14]
			pH < 6.5	pH > 6.5		
Cd	20	20–40	5	20	39	40
Ni	300	300–400	100	200	420	420
Zn	2500	2500–4000	500	1000	2800	2800
Cu	1000	1000–1750	250	500	1500	1500
Cr	500	-	600	1000	-	1200
Pb	750	750–1200	300	1000	300	300

The source of heavy metals in sludge is wastewater, which is mainly generated by plants using galvanic processes, steel pickling, and the recycling of lead batteries. Table 2 shows the industries that are the source of heavy metal emissions to the environment. Moreover, heavy metals come from domestic sewage, surface run-off, and corrosion of sewage pipes [15].

Table 2. Industry branches which are a source of heavy metal emissions to the environment [16,17].

Metal	Industry Branches
Cadmium	Galvanising plants, manufacture of dyes, batteries, accumulators, paints and plastics, polymer stabilisers, chemical industry, manufacture of plant protection products, graphic and printing works
Lead	Production of dyes, accumulators, batteries, fertilisers, automotive industry, energy industry, plant protection products, electrochemical industry
Chromium	Electroplating; tanning; wood impregnation; textile, dye and plastic manufacturing; printing and graphic arts plants
Copper	Metallurgical, paint, textile, plant protection products and fertiliser industries
Mercury	Production of batteries, phosphoric acid, caustic soda, pulp mills, production of plant protection products, metallic mercury
Nickel	Electroplating industry, paper industry, refineries, steelworks, fertiliser factories
Zinc	Production of batteries, paints, textile industry, plastics, polymer stabilisers, printing and graphic arts

Heavy metals entering the environment affect all links in the food chain, from microorganisms, through plants and animals, to humans [18,19]. Therefore, the accumulation of heavy metals in the ground is particularly dangerous [20]. Heavy metals are divided into two groups. The first one, including cadmium, lead, and mercury, is characterised by high toxicity to humans and animals, but lower toxicity for the growth and development of plants. In excess, the metals of the second group, i.e., copper, zinc, and nickel, are more toxic to plants than to animal and human organisms. The increased content of heavy metals can adversely affect the biological properties of soil, cause changes in the food chain, have a toxic effect on plants, and contaminate groundwater. When the permissible content level is exceeded, heavy metals reduce soil fertility, inhibit soil enzymatic activity, and change soil acidity [21]. Heavy metals can be removed from sewage sludge by chemical or biological methods. Chemical methods include ion exchange, adsorption, electrochemical treatment, and membrane filtration. Biological methods include activated sludge processes and biosorption. Although biological methods are low-cost and environmentally friendly techniques, they need large areas and proper maintenance and operation [22–24].

The aim of the study was to analyse sewage sludge from three different wastewater treatment plants located in central Poland in terms of the content of heavy metals, their mobility, and the risk of heavy metals accumulation in soil. The geoaccumulation index

(I_{geo}), the potential environmental risk index (PERI), the risk assessment code (RAC), the environmental risk factor (ERF), and the environmental risk determinant (ERD) proposed by the authors were calculated based largely on the analysis of the mobility of heavy metals.

2. Materials and Methods

2.1. Materials

Sewage sludge (S1, S2, S3) was collected from wastewater treatment plants located in the Świętokrzyskie Voivodeship (Table 3). According to the proximity principle, locations for natural sewage sludge disposal sites (P1, P2, P3) were selected as close as possible to the wastewater treatment plants (Figure 1). For S2 and S3, the same location of sludge utilisation was chosen due to the location of these sewage treatment plants. The characteristics of the soils in the accepted locations are presented in Tables 4 and 5. The sorption properties of the soils are arranged according to their type. A poor sorption complex is characterised by sandy soils, while organic soils have a particularly high sorption capacity. Linde considers that the organic substance in an acidic environment is a more active sorbent for heavy metals than most mineral compounds [25]. Soils have an acidic pH in the range of 4.1 to 4.5. A low pH affects the increased mobility of cadmium and copper. An acidic reaction, a low humus content, and a poor sorption capacity influence increased lead migration. The sorption of zinc depends on the reaction and the granulometric composition. It forms a permanent bond with the soil organic substance, which results in its accumulation. The content of organic substances and the low pH level of the soil boost zinc migration in the soil. The zinc content is high in the analysed soil; however, these are typical values characteristic of Polish soils. Nickel is strongly linked to the organic substance, and its solubility increases with acidity. Due to the susceptibility of nickel to the organic substance, its high mobility is maintained in many soils, even under alkaline conditions [26].

Table 3. Characteristics of wastewater treatment plants [27].

Location and Type of WWTP	Equivalent Number of Residents	Sewage Sludge Treatment	Location of Potential Disposal Sites of Sewage Sludge	Distance from WWTP to Potential Disposal Site of Sewage Sludge, km
Daleszyce mech.-biol. SBR	5000	Oxygen stabilisation	P1—Wola Kopcowa	13.5
Skarżysko-Kamienna mech.-biol. hybrid	59,500	Fermentation	P2—Wąchock	13.8
Starachowice mech.-biol. SBR	95,000	Fermentation	P3—Wąchock	7.6

mech.-biol.—mechanical-biological; SBR-sequential biological reactor WWTP-The mobility of SS from three different wastewater treatment plants.

Table 4. Content of the element from the heavy metal group in agricultural soils of Świętokrzyskie Voivodeship, $\text{mg}\cdot\text{kg}^{-1}$ d.m. [28].

Heavy Metal	Location of Potential Disposal Sites of Sewage Sludge	
	P1	P2 = P3 *
Cu	22.4	3.0
Cr	0.37	3.3
Cd	7.4	0.09
Ni	4.6	2.4
Pb	4.6	10.8
Zn	39.6	19.3

* P2 = P3—the same location of sludge utilisation.

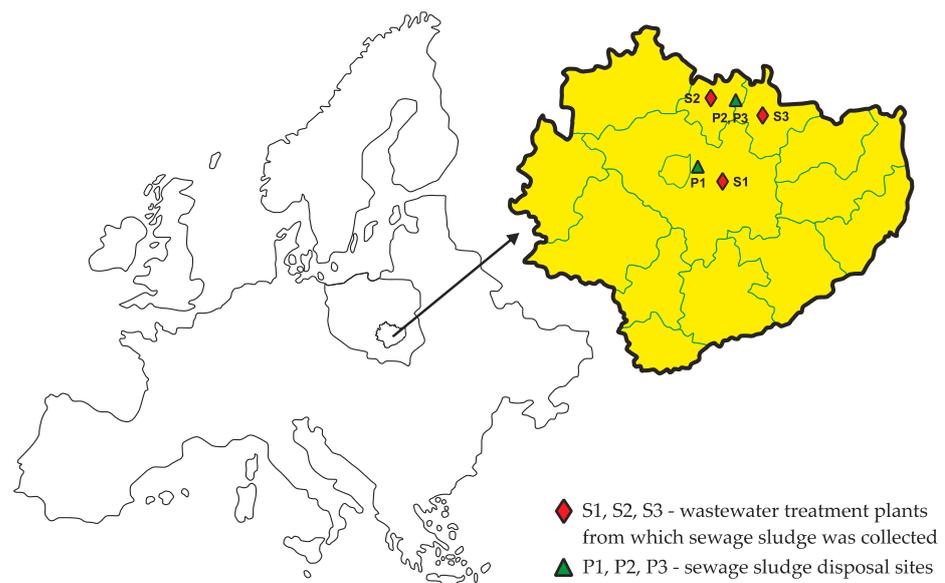


Figure 1. Location of potential disposal sites of sewage sludge and wastewater treatment plants.

Table 5. Characteristics of analysed potential disposal sites of sewage sludge [28].

Point	Soil type	Bonitation Class	Soil Species	Soil Species	pH	Caries, %	C _{organic} , %	N _{total} , %	C/N Ratio
P1	leached brown soils	IVa	5 (rye good)	light clay sand	4.5	1.08	0.63	0.08	7.9
P2 = P3	clay soils	IVa	5 (rye good)	clay sand	4.1	0.99	0.57	0.08	7.2

2.2. Mobility of Heavy Metals of Sewage Sludge

In sewage sludge, metals are dissolved, precipitated, coprecipitated with metal oxides, absorbed, or assimilated with biological residues. They can take the form of oxides, hydroxides, sulphides, sulphates, phosphates, silicates, organic compounds in the form of humic complexes, and compounds with complex sugars [20]. Heavy metals may belong to four different mobility fractions, depending on their migration capacity. Speciation of heavy metals allows them to be separated into the individual forms in which they occur [29,30]. In order to determine the fraction in which the metals occur, a sequential extraction according to the BCR (European Community Bureau Reference) procedure was applied [31,32]:

Stage I: extraction CH_3COOH —for determination of the content of assimilable and carbonate-bound metals (fraction FI—interchangeable; mobile);

Stage II: extraction $\text{NH}_2\text{OH}\cdot\text{HCl}$ —for determination of the content of metals associated with amorphous iron and manganese oxides (fraction FII—reductive; mobile);

Stage III: extraction $\text{H}_2\text{O}_2/\text{CH}_3\text{COONH}_4$ —for determination of the content of the organometallic and sulphide fraction (fraction FIII—oxidising; potentially mobile);

Stage IV: mineralisation of the residual fraction with a mixture of concentrated acids (HCl , HF , HNO_3)—for determination of the content of metals bound to silicates (fraction FIV—residual; immobile).

Sludge samples (S1, S2, S3) taken from all facilities before the hygienisation process were used for mobility tests performed by the BCR sequential extraction. A PerkinElmer Optima 8000 inductively coupled plasma optical emission spectrometer (ICP-OES) (Waltham, MA, USA) was used to determine heavy metal content in the extracts. Every process of determination was repeated 4 times.

2.3. Contamination Assessment Methodology

Geoaccumulation index of heavy metal in soil (I_{geo})

The I_{geo} is used in order to assess the degree of accumulation of heavy metals. At first, the I_{geo} was used for the ecological risk assessment of sediments [33]. It was also used for the assessment of the contamination of soil [34], sewage sludge [3], and sewage sludge ash [35]. The I_{geo} is described in the equation [36,37]:

$$I_{geo} = \log_2 \frac{C_n}{1.5 \cdot B_n} \quad (1)$$

where:

C_n —content of a given element from the group of heavy metals contained in sewage sludge, $\text{mg} \cdot \text{kg}^{-1}$ d.m.; B_n —content of a given element from the group of heavy metals present in the soil, $\text{mg} \cdot \text{kg}^{-1}$ d.m. (Table 6). The constant value 1.5 is introduced for better analysis of the natural variability of the content of the chosen substance in the environment.

Table 6. B_n and T_r^i of heavy metals (HMs) from sewage sludge (SS).

B_n^* , $\text{mg} \cdot \text{kg}^{-1}$	Cu	Cr	Cd	Ni	Pb	Zn
B_{S1}	4.6	7.4	0.37	4.6	22.4	39.6
$B_{S2} = B_{S3}$	3.0	3.3	0.09	2.4	10.8	19.3
T_r^{i**}	5	2	30	5	5	1

* $B_n = C_R^i$ —values determined on the basis of the report on the realisation of stage III of the procurement [26]. Measurement points were located adequately in relation to the analysed wastewater treatment plants ** on the basis of [36].

The classification of the heavy metals I_{geo} is: < 0 —no pollution; $0-1$ —no pollution, moderate pollution; $1-2$ —moderate pollution; $2-3$ —moderate or high pollution; $3-4$ —high pollution; $4-5$ —high or very high pollution; >5 —very high pollution [37].

Potential environmental risk indicator (PERI)

The PERI is a measure of the environmental risk of soil contamination with heavy metals and is described in the following formulas [36,38]:

$$C_f^i = \frac{C_D^i}{C_R^i} \quad (2)$$

where:

C_f^i —pollution factor; C_D^i —concentration of the i -th element from the group of heavy metals present in sewage sludge, $\text{mg} \cdot \text{kg}^{-1}$ d.m.; C_R^i —concentration of the i -th element from the group of heavy metals in the soil, $\text{mg} \cdot \text{kg}^{-1}$.

$$E_r^i = T_r^i \cdot C_f^i \quad (3)$$

where:

E_r^i —indicator of the potential ecological risk of the i -th element from the group of heavy metals; T_r^i —toxicity factor of the i -th element from the group of heavy metals. Heavy metals differ in degree of toxicity, which takes into account the toxicity factor (T_r^i): lead—5; cadmium—30; chromium—2; copper—5; nickel—5; zinc—1 [37].

The sum of indicators of potential ecological risk of the contamination with heavy metals (HMs) from sewage sludge (SS) in the ground is defined by the formula [37]:

$$PERI = \sum_{i=1}^n E_r^i \quad (4)$$

Table 7 shows the degree of the potential environmental risk in relation to the PERI value.

Table 7. Interpretation of the potential environmental risk indicator (PERI) value [38–41].

E_r^i	PERI	Potential Environmental Risk
<40	<150	Low
40–80	150–300	Medium
80–320	300–600	High
>320	>600	Very high

2.3.1. Risk Assessment Code (RAC)

The RAC was also used to assess the environmental risks posed by heavy metals. The RAC was used to assess soil contamination with heavy metals from sewage sludge and sewage sludge ashes [42]. The RAC takes into account the percentage of heavy metals present in the mobile fraction (F1). The risk level can be classified into 5 categories: <1—no risk; 1–10—low risk; 11–30—medium risk; 30–50—high risk; >50—very high risk [36]. It is defined by the formula [42,43]:

$$Rac = \frac{F1}{HM} \cdot 100\% \quad (5)$$

where:

F1—concentration of heavy metal in acid-soluble/free fraction, $\text{mg} \cdot \text{kg}^{-1}$; HM—total heavy metal concentration, $\text{mg} \cdot \text{kg}^{-1}$.

2.3.2. Environmental Risk Factor (ERF)

The ERF takes into account the proportion of heavy metals in mobile fractions (FI+FII) to their content in stable fractions (FIII + FIV). It is determined by the formula [39]:

$$ERF = \frac{F_1 + F_2}{F_3 + F_4} \quad (6)$$

where:

F₁—fraction FI; F₂—fraction FII; F₃—fraction FIII; F₄—fraction FIV.

The classification of the ERF results is: $0 < ERF \leq 0.4$ —low risk; $0.4 < ERF \leq 1$ —medium risk; $1 < ERF$ —high risk [39].

2.3.3. Environmental Risk Determinant (ERD)

Analysing the mobility of heavy metals, one can conclude that only the FIV fraction is fully stable and does not migrate to the environment. The fractions FI and FII are considered mobile, while FIII can be mobile under certain conditions, i.e., when microorganisms fully process organic matter in the soil and when a storm occurs under the influence of ozone. Metals in water-soluble compounds and metals associated with carbonates are considered the most mobile. Metals bound to iron and manganese oxides are released to the environment much more slowly. Under certain conditions of pH and oxidation-reduction potential, FII bound metals can show significant bioavailability [44]. The environmental risk assessment is based on the first three fractions, taking into account the level of individual predisposition of each fraction to release heavy metals into the soil environment. The ERD determines the content of an element in the heavy metal group, depending on its content in the four fractions. Each fraction is assigned an appropriate weight depending on the 0–1 scale. The authors proposed the use of the ERD indicator because none of the indicators using the issue of mobility take into account the weight of individual fractions. It should be taken into account that FI, FII, and FIII fractions are mobile, but the FI fraction is much more mobile than FII and FIII, which takes into account the formula for the ERD indicator. The weighting ranges adopted are proposed on the basis of an analysis of the scale of the other indicators. Its determinant is described by the formula:

$$ERD = F_{p1} + F_{p2} + F_{p3} \quad (7)$$

where:

$F_{p1} = F_1; F_1$ —metal content in fraction FI on a scale of 0–1; $F_{p2} = F_2^2; F_2$ —metal content in fraction FII on a scale of 0–1; $F_{p3} = F_3^3; F_3$ —metal content in fraction FIII on a scale of 0–1.

The classification of the ERD results is: $0 < ERD \leq 0.35$ —low risk; $0.35 < ERD \leq 0.6$ —medium risk; $0.6 < ERD \leq 0.8$ —high risk; $0.8 < ERD$ —very high risk.

3. Research Results and Discussion

Table 8 shows the results of the speciation analysis of heavy metals in sewage sludge. The cadmium, nickel, and copper content of the sewage sludge from all three wastewater treatment plants did not show any values exceeding the acceptable limits given in the literature [8–14]. For chromium, this value was exceeded only for S2, as the value was $2760.3 \text{ mg}\cdot\text{kg}^{-1} \text{ d.m.}$, and was several times higher than the limit values. A high lead content was found in S1, equalling $427.10 \text{ mg}\cdot\text{kg}^{-1} \text{ d.m.}$ All tested sewage sludge, on the other hand, was characterised by a high zinc content, exceeding the limit values indicated in the regulation of the Minister of the Environment on municipal sewage sludge [9]. Zinc belongs to the migrating elements, and has a relatively low degree of toxicity to humans and animals. However, high doses of zinc cause damage to many biochemical processes, and it is deposited in the kidneys, the liver, and genital glands [45].

Table 8. Chemical speciation of heavy metal * in sewage sludge, $\text{mg}\cdot\text{kg}^{-1}$.

Heavy Metal	Fraction I	Fraction II	Fraction III	Fraction IV	$\sum F1 \div F4$
Sewage sludge—S1					
Cu	0 ± 0.1	0 ± 0.1	14.6 ± 0.9	6.5 ± 0.4	21.1
Cr	13 ± 0.9	4.2 ± 0.2	29.4 ± 1.6	59.1 ± 2.3	105.7
Cd	1.2 ± 0.1	1.5 ± 0.1	3.7 ± 0.1	3 ± 0.1	9.4
Ni	2 ± 0.2	0.9 ± 0.1	6.1 ± 0.5	7.7 ± 0.6	16.7
Pb	2.5 ± 0.2	0 ± 0.2	16.2 ± 0.3	408.4 ± 9.1	427.1
Zn	509.9 ± 9.0	447.3 ± 9.2	1119 ± 15.0	693.2 ± 8.4	2770.0
Sewage sludge—S2					
Cu	0 ± 0.1	0 ± 0.1	9.5 ± 2.7	12.3 ± 1.9	21.8
Cr	5 ± 0.4	2.3 ± 0.1	1284 ± 12	1469 ± 47	2760.3
Cd	0.3 ± 0.1	0.7 ± 0.2	1.1 ± 0.3	10 ± 1.7	12.1
Ni	4.5 ± 0.1	1 ± 0.1	14.1 ± 0.3	8.9 ± 0.1	28.5
Pb	0.7 ± 0.2	1 ± 0.2	0 ± 0.1	29.6 ± 7.8	31.3
Zn	152.9 ± 0.7	144.5 ± 0.2	537.5 ± 23.6	4516 ± 91.0	5351.0
Sewage sludge—S3					
Cu	0.9 ± 0.2	1.2 ± 0.2	124 ± 0.8	69.9 ± 0.6	196.0
Cr	2.1 ± 0.2	0.9 ± 0.1	78.1 ± 0.8	61.5 ± 0.5	142.6
Cd	0.1 ± 0.1	0.3 ± 0.1	0.9 ± 0.2	1.1 ± 0.3	2.4
Ni	7.9 ± 0.3	3.2 ± 0.2	9.8 ± 0.5	23 ± 0.9	43.9
Pb	7.8 ± 0.8	1 ± 0.2	4.5 ± 0.5	43.5 ± 4.5	56.8
Zn	79 ± 1.0	275 ± 3.0	1491 ± 21.0	932 ± 82.0	2777.0

* \pm standard deviations.

When assessing the value of the I_{geo} of heavy metals in the soil, it can be concluded that the dominant heavy metals causing a high risk of contamination are cadmium, zinc, and chromium (Figure 2). Zinc, the content of which exceeded the limit values outlined by legal acts, reached a very high level of potential risk in all three treatment plants. It should be taken into account that the I_{geo} compares the heavy metal content of the sewage sludge to that of potentially used soils. The difference between the two values was very high due to the low values of heavy metals in soils.

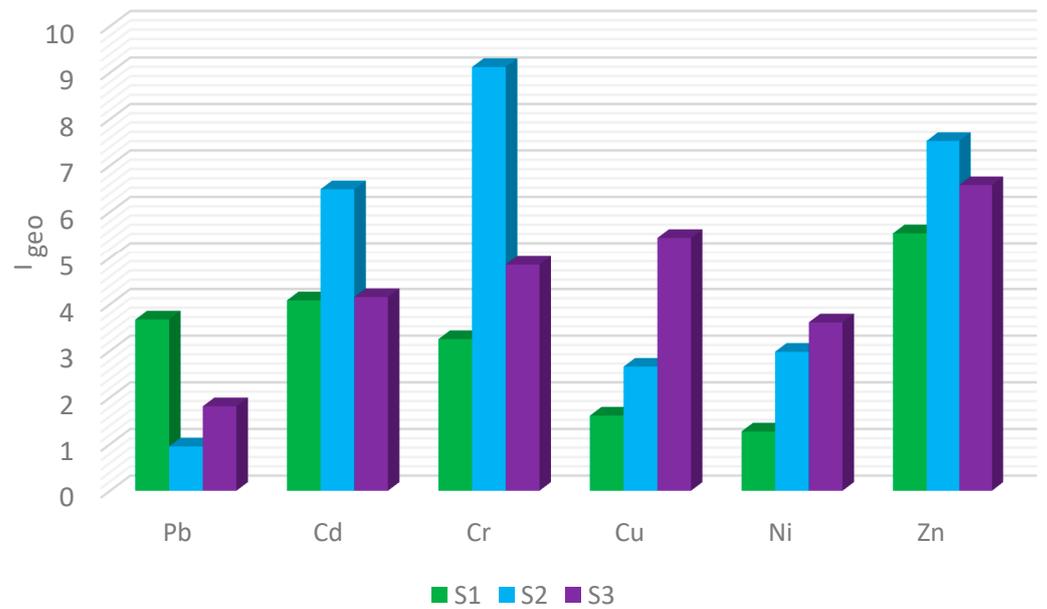


Figure 2. The geoaccumulation index (I_{geo}) of HMs in SS.

The values of the PERI indicate that the main element in the heavy metal group causing a very high PERI value in all three sewage sludges is cadmium (Figure 3). Chromium of S2 and copper of S3 also showed very high potential environmental risk values. Zinc from S1 showed a moderate level of risk, and high for the other two. The main factor distinguishing the potential environmental risk indicator from the other indicators is the variation in toxicity levels of each heavy metal. Zinc showed the lowest toxicity factor of 1, which resulted in lower risk values according to the PERI when compared to the I_{geo} .

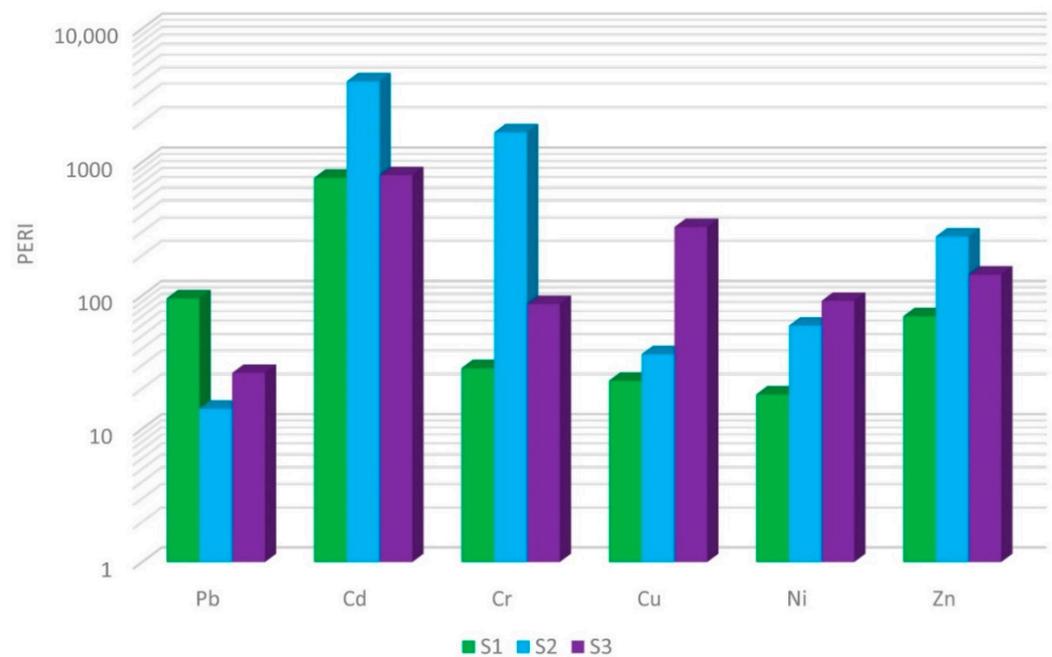


Figure 3. PERI of HMs in SS.

In most cases, the level of the RAC does not show a high environmental risk (Figure 4). This is due to the low share of heavy metals in the most mobile fraction (F1). The zinc content in the first fraction is 18.41% for S1, 2.86% for S2, and 2.84% for S3. Therefore, it can be concluded that a decidedly greater part of zinc is found in stable fractions which cannot

migrate to the environment. This results in a low risk level despite exceeding the values for zinc in relation to the regulation of the Minister of the Environment on municipal sewage sludge [9].

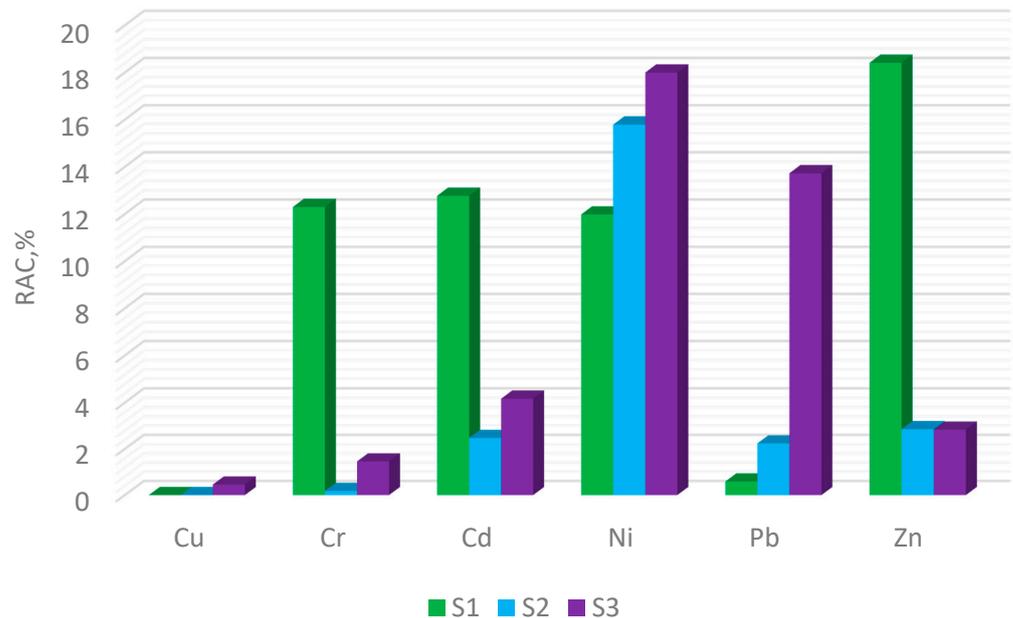


Figure 4. RAC of HMs in SS.

Similar values of the RAC for copper (S1–S3), lead (S1), and zinc (S1) were obtained by Zhang et al. [42]. The RAC with a value indicating a low risk of pollution of the environment with copper is also confirmed by the literature data [39]. Copper is an essential component of many enzymes and proteins, but at high concentrations it can be toxic. Many plants are sensitive to the presence of copper, which can reduce yields. The RAC of nickel for all tested sewage sludge samples, and those presented in [42], belongs to the range indicating a medium risk of contamination of the environment with this metal. On the basis of the RAC of sewage sludge analysed by Tytła [39], and the tested S1–S3 sludge samples, it was discovered that the tested sewage sludge poses a lower risk of a negative influence of nickel on the environment.

The results of the ERF for SS from the analysed facilities are presented in Figure 5. The analysis of the environmental risk factor showed a low level of contamination for all heavy metals from the three facilities, except for zinc for the S1 and cadmium for S2. Zinc in S1 showed a medium risk of contamination. This was due to its increased content in fractions FI and FII. The literature presents the ERF with a high risk of contamination with zinc [39], and a medium risk for cadmium [39].

Analysing the results of the environmental risk determinant, it can be concluded that most metals show a low level of risk (Figure 6). Zinc in all three sewage sludge samples showed a low risk of contamination. The highest indicator level for zinc was obtained for S1. However, the ERD indicator did not show a medium risk when compared to the ERF indicator, due to the fact that the ERF indicator treats the FI and FII fractions equally.

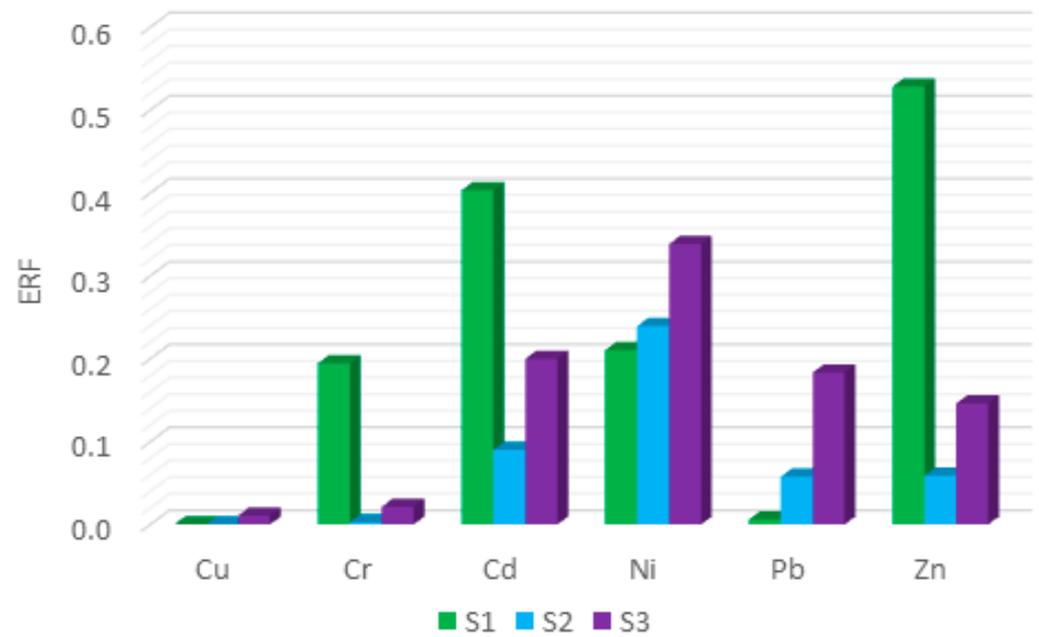


Figure 5. The environmental risk factor (ERF) of HMs in SS.

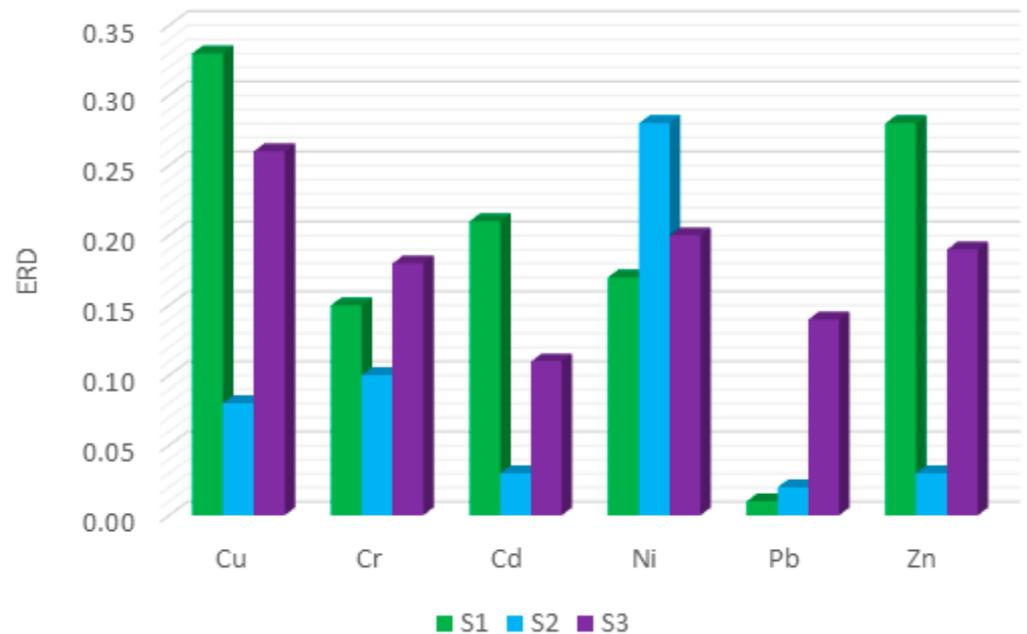


Figure 6. The environmental risk determinant (ERD) indicator of HMs in SS.

4. Conclusions

The tested sewage sludge was characterised by a high zinc content in relation to the applicable legal regulations. However, analysis of the potential environmental risks has shown that despite exceeding the limit values for concentrations of heavy metals in sewage sludge, its use for natural purposes does not necessarily involve a high risk of environmental contamination. A key role is played here by a speculation analysis of heavy metals in sludge. The mobility of HMs is important in terms of the possibility of natural or agricultural use of sewage sludge. Consideration must be given to the extent to which heavy metals in sewage sludge are permanently bound, and whether there are external factors that can influence the change in the speculative fractions in which they are currently found.

The I_{geo} showed a very high or high risk of organic contamination with zinc. On the other hand, the PERI for all three sludge samples showed a very high risk of cadmium contamination, while for zinc it was moderate and high. The RAC for all metals showed mostly a medium to low risk. For zinc, it was low in each case. The environmental risk factor analysis showed low levels of contamination for all three heavy metals, except for zinc for S1 and cadmium for S2.

The authors' own indicator, the ERD, is also based on the issue of metal mobility. It determines the content of a given element from the group of heavy metals, depending on its content in four fractions. It takes into account only three fractions (FI, FII, and FIII), with the highest value of potential contamination assigned to metals contained in the FI fraction and the lowest in the FIII fraction. Zinc in all three sewage sludge samples showed a low risk of contamination. The highest indicator level for zinc was obtained for S1. However, the ERD indicator did not show the average risk when compared to the ERF indicator, due to the fact that the ERF indicator treats the FI and FII fractions equally.

The heavy metal content of FII and FIII fractions, which are conditionally mobile, cannot be ignored. However, they cannot be treated in the same way as the metals in the most mobile FI fraction. The proposal to use the indicator suggested by the authors is a response to the differences in the mobility of heavy metals depending on the form in which they occur in sewage sludge.

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