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Research Article

Ecological risk assessment of domestic sewage sludge: a case study

Elif YAKAMERCAN[1], **Ahmet AYGÜN**[1,*]

[1] *Environmental Engineering Department, Bursa Technical University, Bursa, Turkey*

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ABSTRACT

This study aims to evaluate potential ecological risks and heavy metal pollution in sewage sludge. For these purposes, domestic sewage sludge samples were collected for a period of one year from a wastewater treatment plant in Bursa, Turkey and analyzed for heavy metals. The average heavy metal content of the sewage sludge was wherein decreasing order of Zn>Cu>Ni>Cr>Pb>As>Se>Cd. As a whole, the concentration of heavy metals was below the limit values indicated within the agricultural land application legal standards. Correlation analysis showed a very strong correlation observed between Ni and Cr. Determining to pollution degree and potential ecological risks, some indices such as Enrichment factor (EF), Single-factor pollution index (PI), Geoaccumulation index (Igeo), Nemerow synthetic pollution index (PN), Contamination factor (Cf), Integrated pollution degree (Cd), Pollution Load Index (PLI), Monominal potential ecological risk (ER), Potential ecological risk index (RI), and the Probability of toxicity (mERM-Q) were used in this study. Based on the pollution index calculations, Zn and Se posed the highest contamination while As and Cd posed the lowest contamination. The mERM-Q values indicated that the probability of toxicity varied from 21 to 49%, while ecological indices indicated that ER (2.0–23.7) and RI (67.3–106.2) values were lower than a threshold value for all samples.

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INTRODUCTION

A high amount of sludge is continuously produced as an unavoidable end product in wastewater treatment plants [1]. In the relevant literature, it is estimated that sewage sludge production is more than 22 million tonnes in the world, annually [2]. Although this high amount seems to be a problem, the sludge can be used for different beneficial purposes. Landfilling, soil application, incineration, and sea dumping are the well-known sludge disposal methods for a long time, among them landfilling is the most common disposal method and generally, it is used in developing countries due to the most economical way but it is not a beneficial purpose [3–4]. Incineration and land application of the sewage sludge are classified as a beneficial usage

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***Corresponding author.**
*E-mail address: ahmet.aygun@btu.edu.tr

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while during the incineration process air pollution occurs and also secondary pollutants can be generated [5].

Moreover, reuse options such as agricultural applications are the most ecologically and sustainable way [6–7]. The main cost of the land application of sewage sludge is transportation also finding a suitable area is also important [8]. The presence of a free field around the treatment plant for the use of treatment sludge also reduces the cost of transportation.

Sewage sludge can be used as a fertilizer (as soil additive) in agricultural areas due to its high organic and nutrient content, therefore, more than 50% of produced sludge is used in agricultural purposes in Europe [9]. It is reported that the land application of sludge increases the yield of crops and enhance soil properties [10–11]. However, some legislations have been imposed to control agricultural applications of sewage sludge [12–14].

In addition to providing nutrient sources for plants, sewage sludge may contain potentially bioaccumulative, toxic and carcinogenic elements such as heavy metals [4,8]. The heavy metal content of sewage sludge depends on the origin of the raw wastewater and the treatment methods [15]. In general, the heavy metal content of domestic sewage sludge is lower than industrial sludge [16–17]. Thus domestic sewage sludge can be more suitable for agricultural usage than municipal or industrial sludge.

Heavy metals can accumulate in the soil in long term applications of sewage sludge and can cause irremediable damage to the soil [15]. The optimum sewage sludge application dosage is also important and in literature, it is reported that no pollution occurs in soil and plants in sludge land application up to 30 t ha $^{-1}$ [18]. Land application of sewage sludge also causes both human health and environmental risks [19]. To prevent possible environmental problems in applications, the information on heavy metals concentration was not enough alone, a proper risk analysis should also be addressed before sewage sludge application in soil. In the relevant literature, there are a lot of studies based on the human health risk of sewage sludge land application [20]. Heavy metals are the most important pollutants and sensitive indicators for monitoring changes in the environment [21]. The behavior of heavy metals affects water chemistry [22]. On the other hand, heavy metals have the bioaccumulation potential and incorporated into the food chain in land application, thus ecological risk determination gains significance in terms of sustainability [4].

To determine the ecological risks and pollution potential of land application of the sewage sludge, some indices are used such as Enrichment factor (EF), Geoaccumulation index (I_{geo}), Single-factor pollution index (PI), Nemerow's synthetic pollution index (PN), Potential ecological risk index (RI), Sediment quality guidelines, and the Probability of toxicity (mERM-Q) [23]. PI and I_{geo} indicate the accumulation level of heavy metals in samples and, PN indicates the heavy metal pollution level of the samples, but toxicity is not considered these indices while RI and mERM-Q focus on the toxicity of heavy metals [23–25]. mERM-Q is among the most widely used sediment quality assessment indices [26]. In literature, although there are comprehensive studies about the potential ecological risks of soil, sediment and biochar [27–31], there are the limited studies about ecological risk assessment for use of sewage sludge in agricultural areas [4,9,32]. To the authors' best knowledge, this is the first study on sediment quality guidelines, EF, and mERM-Q index calculations on determination of the sewage sludge land application potential. Many indices were developed for evaluating the environmental risks of toxic metals in surface sediments [23].

The aims of this study are (*i*) to evaluate the heavy metal (Cu, Cd, Zn, Pb, Ni, Cr, Se and As) content in sewage sludge samples collected from domestic WWTP (Yenişehir, Bursa) in terms of the regulations (national (TR) and international (EU and EPA)) on the land application; (*ii*) to define the relationship between heavy metals statistically; (*iii*) to assess the heavy metal pollution and ecological risks of heavy metal in sludge with using several indices; (*iv*) to compare and evaluate the indices results.

MATERIAL AND METHODS

Study Area

Yenişehir is located in the eastern part of Bursa Province, Turkey with a population of 53,921, and covers 720 km^2 of a total surface area. The main economic activity of this rural district is agriculture and livestock, hence, there is not an industrial discharge to the treatment plant. Beet, pear, beans, wheat and peaches are the main agricultural products in Yenişehir [33]. The annual rainfall and temperature were 58.74 mm and 14.6 °C, respectively. The dry season is described between April and September, while the wet season is between October and March.

Sampling of Soil and Sewage Sluge

Soil samples were collected from an agricultural site in Yenişehir (North-Western Turkey), is located between 40° 17′ 13″ of the northern latitudes and 29° 38′ 46″ of eastern latitudes, from the surface layer based on Turkish National Standard TS 9923 [34]. The top 10 cm of the soil layer were sampled using a hand shovel. 3–5 subsamples were mixed to obtain a bulk sample. The soil samples were kept under 4° C in plastic containers and transported to the laboratory. Field moist soil was homogenized, sieved (2 mm), air-dried, and milled in compliance with TS 10308 ISO11464 for chemical analysis [35]. The soil background contents of As, Cd, Cr, Cu, Fe, Hg, Ni, Pb, Se and Zn are 8.74, 4.18, 34.30, 63.45, 25.63, 0.97, 39.75, 11.35, 1.35, 122.59 mg/kg, respectively.

In this study, sewage samples were collected from the Yenişehir wastewater treatment plant (WWTP) which is located between 40° 15′ 05″ of the northern latitudes and

29° 40′ 25″ of eastern latitudes. The background soil type is alluvial and the heavy metal content of this soil was used in calculations.

The WWTP was operated as A^2O process and sludge dewatered with a decanter. In this study, dewatered sludge was used for the characterization of heavy metals. The volumetric flow rate and daily sludge production of the WWTP are 5,420–6,878 m^3 day^{-1} and 20,471–24,258 kg day^{-1}, respectively. Sewage sludge samples were collected during 2019 for dry and wet seasons from the domestic wastewater treatment plant two times a month and 24 samples were evaluated totally. All collected samples were kept at 4 °C due to inhibition of biological activity until the laboratory analyses, especially for nutrients and organic content.

Determination of Psyhcochemichal Charachteristics of Sewage Sludge and Total Heavy Metal Concentrations

Physicochemical properties of sludge were determined via dry matter (TS 9546 EN 12880) [36], organic matter (TS 9546 EN 12880) [36], Total Kjeldahl Nitrogen (TKN) (TS 8337 ISO 11261) [37], and Total Phosphorous (TP) (TS 8338) [38]. The conductivity and pH value of samples were determined by using a digital calibrated multiparameter (WTW 330i, Germany).

The heavy metal concentration of sewage sludge and soil were determined by the inductively coupled plasma mass spectrometry (ICP/MS). Firstly, collected sludge samples were dried, then homogenized at 1000 rpm for 4 minutes by a blender. Microwave-assisted acid digestion system (Berghof Microwave MWS-3) was used for the digestion of 1 g of a sample by the microwave-assisted acid (3 mL of HCI + 9 mL of HNO_3) according to EPA Method 3051A. After the digestion process, heavy metal concentrations of the samples determined by an ICP/MS (iCAP Qc, Thermo Fisher Scientific, Bremen, Germany). In this study, for each heavy metal, five replicate results were obtained and the average heavy metal content was calculated per kg of sludge or soil as a dry weight basis.

Pollution Indices and Potential Ecological Risk Assessment

Pollution indices which are widely used were preferred to evaluate the pollution and ecological risk potential of heavy metals in sewage sludge samples. Summary information for the pollution indices used in this study was given in Table 1.

Statistical Analysis

All calculations were performed using MS Excel, R Software (3.6.1), and Origin 9 Pro. Descriptive statistics (mean, median, maximum, minimum, and standard deviation) were calculated. The confidence interval of all statistical analyses was selected as 95% ($p<0.05$). T-test or Mann Whitney U test was applied to explain the seasonal differences. To determine the relationship between heavy metals of sewage sludge, Spearman's correlation coefficients (r) were calculated. A very strong correlation is observed when the absolute $|r|$ value is close the 1, strong correlation is considered when $|r|$ value is between 0.7 to 1 and moderate correlation is considered value is between 0.5 to 0.7. Hierarchical cluster analysis (HCA) was applied to determine the groups of heavy metals in sewage sludge and also their pollution (I_{geo} and C_f) and risk indices (ER) the HCA results were demonstrated as dendrograms. Cluster analysis is based on a single linkage method thus squared Euclidean distance which is used to determine the degree of the similarities between analyzed groups calculated [39].

RESULTS AND DISCUSSION

Physicochemichal Characteristics of Sewage Sludge

The physicochemical properties of sewage sludge samples were shown in Table 2. pH is an important determinant of metal mobility in soil, and increasing values of this parameter causes decreased heavy metal migration and enhances released content [40]. In this study, the annual pH value of sludge samples ranged from 6.32 to 8.58. These values are in the same range as previous studies for sewage sludge land applications in other regions [15,41–42].

OM and nutrient content are important for determining the sewage sludge nutritive value for plants in land application [10]. In this study, the sewage sludge, organic matter content was variable from 61.6 to 71.3%. These values were higher than organic soil range, thus the situation indicated that samples were rich in terms of organic matter [29]. Nutrient concentrations were characterized by TKN (9.28–46.62 g/kg), and TP (8.86–13.97 g/kg), those provide high benefit for agricultural usage when compared to commercial fertilizers.

EC is an indicator of salt content, higher values may result in adverse effects on soil, and secondary salinization may occur high EC values can be harmful for plant [42]. In this study, EC values ranged from 0.99 to 1.9 mS/cm, which were far from the risk of salinization.

When the obtained results are compared with previous studies, OM parameter is higher than relevant literature [43–44], while other parameters such as an EC is found to be lower than the values given in previous studies [42,45]. TKN and TP values are similar to slightly above the previous studies [46]. As clearly seen from the relevant examples, these differences are common in sewage sludge samples and mainly resulted from the influent wastewater characteristics and treatment methods [3].

Heavy Metal Concentrations in Sewage Sludge

The heavy metal content of sewage sludge must be determined before the land application due to their potential ecological risks [9]. Sewage sludge samples were collected in dry and wet seasons to obtain an annual and also

Table 1. Pollution indices and description

Indices	Equation	Class	Desciption
Geoaccumulation Index (I_{geo}) [23]	$I_{geo} = \log_2\left(\frac{C_n}{B_n \times 1.5}\right)$ C_n: Measured heavy metal concentration in sewage sludge (mg/kg) B_n: Background soil heavy metal concentration (mg/kg)	$I_{geo} \leq 0$ $0 < I_{geo} \leq 1$ $1 < I_{geo} \leq 2$ $2 < I_{geo} \leq 3$ $3 < I_{geo} \leq 4$ $4 < I_{geo} \leq 5$ $5 < I_{geo}$	Uncontaminated Uncontaminated to moderately contaminated Moderately contaminated Moderately to heavily contaminated Heavily contaminated Heavily to extremely contaminated Extremely contamination
Single-factor pollution index (PI) [23]	$PI = \frac{C_s}{C_n}$ C_s: Measured invidual heavy metal concentration in sewage sludge or regulation standard (mg/kg) C_n: Background soil or regulation limit value of invidual heavy metal concentration (mg/kg)	$PI \leq 1$ $1 < PI \leq 2$ $2 < PI \leq 3$ $3 < PI \leq 5$ $5 < PI$	No contamination Low level Moderate level Strong level Very strong level
Nemerow synthetic pollution index (PN) [23]	$PN = \sqrt{\frac{P_{i,ave}^2 + P_{i,\max}^2}{2}}$ $P_{i,\,ave}$: The average value of single factor pollution index $P_{i,\,max}$: The maximum value of single factor pollution index	$PN \leq 0.7$ $0.7 < PN \leq 1$ $1 < PN \leq 2$ $2 < PN \leq 3$ $3 < PN$	Safety Warning line of pollution Slight pollution Moderate pollution Heavy pollution
Contamination factor (C_f) [23]	$C_f = \frac{C_S}{C_n}$ C_s: Measured individual heavy metal concentration in sewage sludge (mg/kg) C_n: Background soil individual heavy metal concentration (mg/kg)	$C_f < 1$ $1 < C_f \leq 3$ $3 < C_f \leq 6$ $6 < C_f$	Low degree Moderate degree Considerable degree High degree
Integrated pollution degree (C_d) [23]	$C_d = \sum_{i=1}^{n} C^i{}_f$	$C_d < 5$ $5 < C_d \leq 10$ $10 < C_d \leq 20$ $20 < C_d$	Low contamination Moderate contamination Considerable contamination High contamination
Pollution Load Index (PLI) [23]	$PLI = \left(C_{f_1} \times C_{f_2} \times C_{f_3} \ldots \times C_{f_n}\right)^{\frac{1}{n}}$ n: Number of heavy metals C_f: Contamination factor	$PLI < 1$ $PLI = 1$ $1 < PLI$	Denote perfection Only baseline levels of pollution Deterioration of soil quality
Monominal potential ecological risk (ER) [25]	$ER = T_r \times P_i$ P_i: Single factor pollution index T_r: Metal toxic response factor Zn (1), Cr (2), Cu (5), Ni (5), Pb (5), As (10), Cd (30)	$ER < 40$ $40 \leq ER < 80$ $80 \leq ER < 160$ $160 \leq ER < 320$ $320 \leq ER$	Low risk Moderate risk High risk Very high risk Extremely high risk
Potential ecological risk index (RI) [25]	$RI = \sum ER_i$ ER_i: Monominal potential ecological risk	$RI < 150$ $150 \leq RI < 300$ $300 \leq RI < 600$ $690 \leq RI$	Low risk Moderate risk High risk Very high risk
Enrichment factor (EF) [23]	$EF = \frac{\frac{C_s}{LV}\text{ sample}}{\frac{C_n}{LV}\text{ background}}$ LV: Reference metal (Fe) concentration C_s: Measured individual heavy metal concentration in sewage sludge (mg/kg) C_n: Background soil individual heavy metal concentration (mg/kg)	$0.5 < EF < 1.5$ $EF > 1.5$	Heavy metal in the soil is caused by natural processes Heavy metal contamination from anthropogenic activities

seasonal characterization of land application potential. Total heavy metal concentration in samples compared the soil background and legal standards are demonstrated in Table 3. These concentrations were within the legal standards for sewage sludge using in agricultural land.

The analysis shows that the Zn had the highest annual concentration in sewage sludge and followed by Cu, Ni, Cr, Pb, As, Se and Cd (Table 3). Similar studies also reported the concentration of the Zn and Cu were the highest and Cd was the lowest one in sewage sludge from municipal wastewater treatment plants [46–47].

Generally, municipal sewage sludge contains lower heavy metal content than industrial sewage sludge. The heavy metal concentrations in this study consistent with previous studies reported for municipal sewage sludge, but lower than industrial sludge due to the significant contribution of industrial processes on wastewater characteristics [16,21].

Average levels of almost all heavy metals, except As and Cd, in sludge samples were higher than corresponding values in background soil (Yenişehir). Toxic elements such as Pb and Cd are not expected to enrich from the sewage sludge disposal onto land ecosystems, but the environmental processes such as dry and wet atmospheric deposition and industrial discharges are well-known sources of those heavy metals in urban and background soil [48]. Some heavy metals were higher in wet season such as Ni and Se. Similar to previous studies, there was not a significant difference ($p>0.05$) between wet and dry seasons [21,49]. As it is stated above, atmospheric deposition resulting the enhancement in the urban run-off flows may be another reason of the seasonal fluctuations of heavy metal species in influent wastewater to the plant, and in the sludge samples indeed [50].

Table 2. Physicochemical properties of sludge

Parameters	Max.	Min.	Mean
pH	8.58	6.32	7.44
Electrical conductivity (EC) (mS/cm)	1.89	0.99	1.45
Organic matter (OM) (%)	71.30	61.60	63.90
Dry matter (DM) (%)	19.27	15.15	17.64
Total Kjeldahl Nitrogen (TKN) (g/kg DM)	46.62	9.28	18.60
Total phosphorus (TP) (g/kg DM)	13.97	8.86	10.93

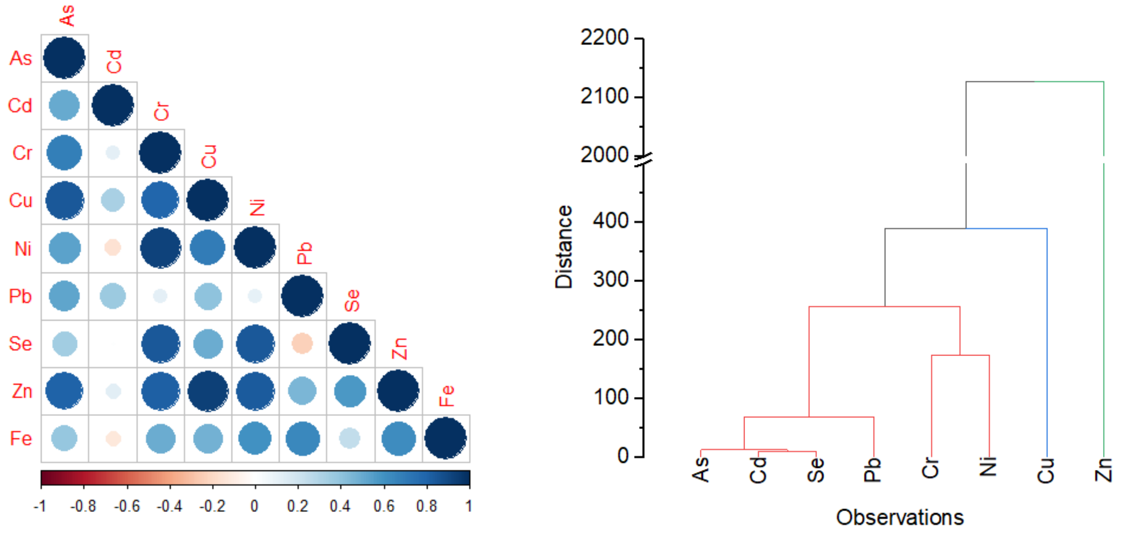
The main sources of heavy metals could be run-off flow of urban streets and corrosion within the sewage system besides the reason of the high Zn concentration is galvanized water supply pipes [40]. The concentrations of these metals depend on raw wastewater characteristics and treatment process [3].

The distribution fittings of heavy metals were inspected, then Spearman correlation coefficients (r) of the heavy metals were assessed to explain inter-relations among elemental species (Fig. 1a). There were both positive and negative correlations among heavy metals in the samples. The multivariate analyses (cluster and principal component analysis) showed that very strong correlation observed between Ni and Cr, and also Cr and Cu. Chromium was also shown a moderate correlation with Zn ($p<0.05$), and the observed relationships were also reported in similar studies [47].

The high correlation coefficients indicate that the statistically related heavy metals in sewage sludge probably have common sources according to their covariances. To evaluate the sources of the heavy metals, hierarchical cluster analysis (HCA) was applied to samples. There are three singletons in a cluster which were showed Fig. 1b. Similar to the previous study based on the elemental characterization of municipal sewage sludge samples in China, Zn and Cu were placed in a single cluster and related to other clusters, but in this study, Zn had the highest distance while the

Table 3. Heavy metal in sludge samples and heavy metal limits for land application of sludge

Heavy metals (mg/kg)	Value				Heavy metal limits for land application of sludge		
	Min.	Max.	Ave.	Std.	EU [12]	TR [13]	USEPA [14]
As	6.7	9.8	8.6	1.0	–	–	41
Cd	3.3	4.3	3.8	0.3	10	10	39
Cr	34.9	102.2	65.4	24.9	1000	1000	–
Cu	117.8	197.6	151.4	21.2	1000	1000	1500
Fe	8444	11254	9385.7	919.9	–	–	–
Ni	40.3	262.7	89.0	66.5	300	300	420
Pb	21.3	39.6	27.4	4.6	750	750	300
Se	5.2	10.2	6.3	1.4	–	–	100
Zn	566.8	900.3	716.8	99.5	2500	2500	2800



(a) (b)

Figure 1. a) Spearman correlation and b) Hierarchical cluster analysis of the heavy metals on sewage sludge samples comparison of the indices.

previous one, and Cu had the highest distance [46]. Also, Cr-Ni and As-Cd- Se were placed in one cluster indicating all they are sharing the common drivers in their vicinity in municipal sludge. Almost similar clusters were observed in both dry and wet season samples, except Cu, was not placed a single cluster in the wet season indicating that this element may have variable sources in two seasons such as atmospheric deposition, agricultural production, traffic intensity on urban streets, etc.

Assessment of Pollution Level and Ecological Risk

Individual and complex indices were used to evaluate the pollution level and ecological risk when the sewage sludge applied to the agricultural soils [9,16]. The enrichment factors (EF), Geo-accumulation index (I_{geo}), Single-factor pollution index (PI) and Contamination factor (C_f) as the individual indices while Nemerow synthetic pollution (PN) index and pollution load index (PLI) as complex indices were applied in this study to evaluate the accumulation levels of the heavy metals. Besides, Potential ecological risk index (RI) and the probability of toxicity (mERM-Q) indices were used to assess the ecological risk of heavy metals in the domestic sewage sludge.

Enrichment Factor (EF)

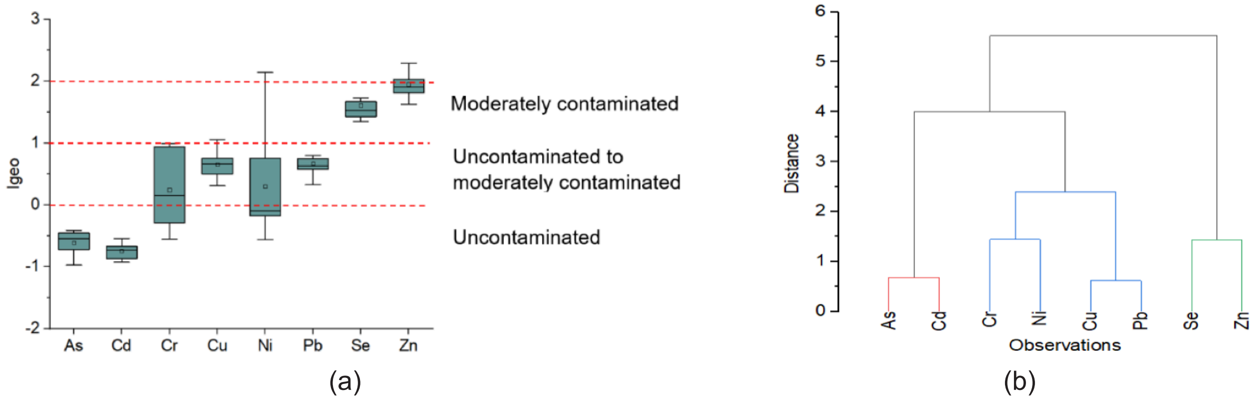
Enrichment factor was calculated to evaluate the possible impact of anthropogenic activity [23]. Due to its stability in soil, iron (Fe) was selected as a reference element for enrichment calculations [51]. All EF values were lower than 0.5, this indicated that no enrichment would be occurred due to the application of sewage sludge to background soil.

Geo-accumulation Index (I_{geo})

The Geo-accumulation index (I_{geo}) for land application of sewage sludge revealed a different degree of heavy metal contamination. The annual mean I_{geo} index results, given in Fig. 2, showed that the agricultural soil was uncontaminated with respect to, As and Cd. The average I_{geo} values of heavy metals were in the decreasing order of Zn>Se>1>Pb>Cu>Ni>Cr>0>As>Cd. This index indicated that the soil was classified as "uncontaminated to moderately contaminated" with Cr, Cu, Ni and Pb. Besides, the pollution level of Zn and Se was "moderately contaminated". Based on these results, it was indicated that Zn (1.95±0.21) caused the highest, and As (−0.61±0.19) caused the lowest contamination in sewage sludge. All of the I_{geo} values in this study were lower than or consistent with previous study [9].

On the comparison of the dry and wet seasons in terms of I_{geo}, generally in dry season I_{geo} values, except Ni and Se, higher than wet season due to sludge heavy metal content, although there was not a significant difference between dry and wet seasons ($p>0.05$).

Hierarchical Cluster analysis was used to determine the groups of heavy metals in terms of I_{geo} index. Two clusters can be distinguished; As-Cd and Cu-Pb were the most similar to each other because they had the lowest distance. As, Cd, Cr, Ni, Cu and Pb can be a group in one cluster, while Se and Zn can be another cluster. The cluster formed for heavy metal concentrations was found to be different from



(a) (b)

Figure 2. Geoaccumulation index values (a) and HCA cluster (b).

the cluster for the I_{geo} index. This difference can arise from soil background content.

Single-factor Pollution Index (PI) and Nemerow Synthetic Pollution (PN) Index

In this study, the single factor pollution index was calculated based on two different approaches for C_s value [9, 23]. Firstly, soil background values were used as C_s and PI ranged from 0.9 to 5.85. Single-factor pollution index values of As and Cd were lower than one this indicated that no contamination occurs. Zn represented the highest pollution potential, whereas Cr showed the lowest to the soil when sewage sludge applied. Secondly, C_s is used as the standard of the individual heavy metal, mg/kg, according to standards for pollutants in sludge from agricultural use [12–14].

In the USEPA regulation on the application of sludge to the land, there is no restriction for Cu, and EU and TR for As and Se [12–14]. The annual single factor pollution index ranged from 0.05 to 0.17 for USEPA and 0.03 to 0.34 for TR and EU 3 $^{rd.}$ Draft based calculations. All calculated PI values were lower than the suggested value for that no contamination occurs when sewage sludge applied to land. All Nemerow synthetic pollution (PN) index values were lower than 0.7 wherein the safety class. The PN values of heavy metals were in the following decreasing order Ni>Cd>Zn>Cu>Cr>Pb for TR and EU also this order Ni>Zn>As>Cu>Pb>Cd>Se was for USEPA rule regulation.

It has been observed that there is a significant difference in PI and PN values calculated using two different C_s values. The reason for this difference was that the background heavy metal concentration of the soil was considerably lower than the standard values of application of the sludge in the soil.

Contamination Factor (CF) and Pollution Load Index (PLI)

From the results of C_f evaluation, it was revealed that the land application may result "moderately contamination" due to Cr, Cu, Ni, Pb and Se; and "contamination" due to Zn and Se (Fig. 3). Also, As and Cd were expected to result "low contamination" in the soil. Higher C_f values depend on the lower background soil heavy metal content in this study and Zn concentration in sewage sludge was far more than average soil background value.

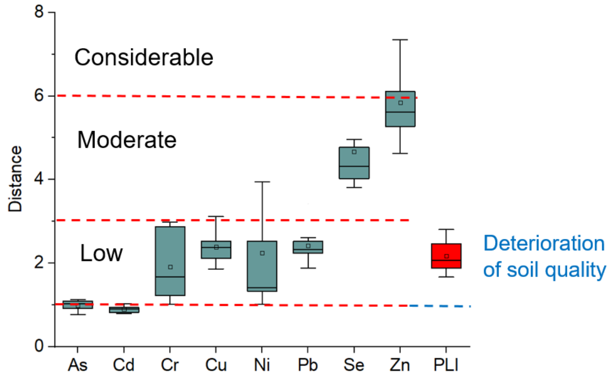


Figure 3. Contamination factor and PLI.

Integrated pollution degree (C_d) values were the sum of individual C_f values. Considering this degree varied from 16.69 to 28.72 with an annual average of 21.35 (high contamination). Besides, C_f and C_d values were lower than previous study [16].

To compare the pollution status and heavy metal effect on sludge quality, pollution load index was evaluated. In this study, PLI of all sewage sludge samples were higher than 1 and the annual average was 2.2, indicated that sludge samples polluted the soil considerably. When the seasonal variation of PLI and C_d value was examined, the average value of PLI was higher in the dry season compared to the wet season, whereas in C_d it was the opposite.

Potential Ecological Risk (RI)

Determining the potential ecological risk from heavy metals is important before the sewage sludge land application. In this study, ecological risks were calculated based on Hakanson approach [25], this index was commonly used to determine ecological risk assessment of soil, sediment and sludge due to its high-level precise scale while the main disadvantage of this index is not useable for some heavy metals such as Se [23].

Calculated results were given in Fig. 4a. Toxicity response values were used in the single pollution index calculation. In average cadmium posed the highest potential ecological risk factor (ER: 26.97±2.4) while Cr posed the lowest one (ER: 3.81±1.5). Similar results were obtained by Gomes et al. [4] who reported the highest and lowest ER values were Ni and Cr respectively. On the other hand, maximum ER value for Ni was higher than Cr.

The average ER values were in the decreasing order of Cd>Pb>Cu>Ni>As>Zn>Cr. A similar order was found in the previous study that indicates the calculated ER values of domestic sewage sludge in Poland, were found as decreasing order: Cd>Hg>Pb>Zn>Cu>Ni>Cr [32]. Although sewage sludge was taken from domestic WWTP in these two studies, the WWTP, where sludge samples were taken in the previous study, was located in an industrial area while in this study it was in the rural area this situation can be a reason of the higher ER values in the previous study (0.9–1060) than this study (4.6–33) [52]. On the other hand, in this study, ER value was lower than 40 for all sludge samples. This indicated that sludge samples have not a potential ecological risk for land application.

There were not any significant differences between the wet and dry seasons, and the average ER values were 11.48 and 11.85, respectively. The ER order was similar in dry and wet seasons, however Pb and As posed higher ER in the dry season than the wet season while Ni posed higher ER in the wet season than the dry season.

Potential ecological risk (RI) was the sum of ER, in this study this value varied from 67.34 to 106.2 and RI value for all samples lower than 150 which indicated that low risk occurs when land application of the sewage sludge. These findings were lower than [9,32,53] and consistent with previous studies [4,54].

HCA was applied to heavy metal based on ecological risk indices and dendrograms showed in Fig. 4b. This dendrogram can be divided into three singletons (As, Ni and Cd) and two clusters. Cluster 1 contained Cu-Pb and cluster 2 contained Cr-Zn. Of all heavy metals, ER values Cu and Pb were the more similar ones due to low distance. Besides, the distance between cluster 1 metals was lower than the distance between cluster 2 metals. The Cd singleton had the highest distance, thus it can be higher risk potential than other heavy metals, but all the sludge samples ER value was lower than the threshold. In the literature, HCA study based on ER from municipal sewage, heavy metals reported that one cluster (Zn and Pb) and three singletons (Ni, Cu and Cd) was observed [32]. The difference can be arise from the heavy metal content of the sewage sludges.

Sediment Quality Guidelines and the Probability of Toxicity (mERM-Q)

Sediment quality guidelines are commonly referred to estimate the biological effects of heavy metals in polluted sediments [23]. Biological effects are classified to effect range row (ERL) and effect range median (ERM) which were described in a previous study [26]. To best of the authors' knowledge, it is the first time to use this index in sewage sludge. Based on this study's results, it was indicated that Zn concentration of all sewage sludge samples was higher than ERL and ERM that can be risky for ecosystem while Pb was lower than ERM and ERL. The 70% of samples had higher As concentration than ERL and 10% of samples had lower Ni than ERM.

Also, mERM-Q index was applied to determine the probability of toxicity. The mERM-Q values were ranged from 0.47 to 1.26. These values indicated that the probability of toxicity 21–49 % (medium to high-risk level) when sewage sludge applied to the land. The major risk contributor metals were Ni and Zn besides, minor risk contributors were Cr, As and Pb. In wet season average mERM-Q value was higher than the dry season but significant differences were not observed between seasons (p>0.05).

The soil pollution from sewage sludge land application studies generally used sediment pollution indices [9,25,26,32] thus in this study, sediment quality guidelines applied to determine toxicity profile of sewage sludge.

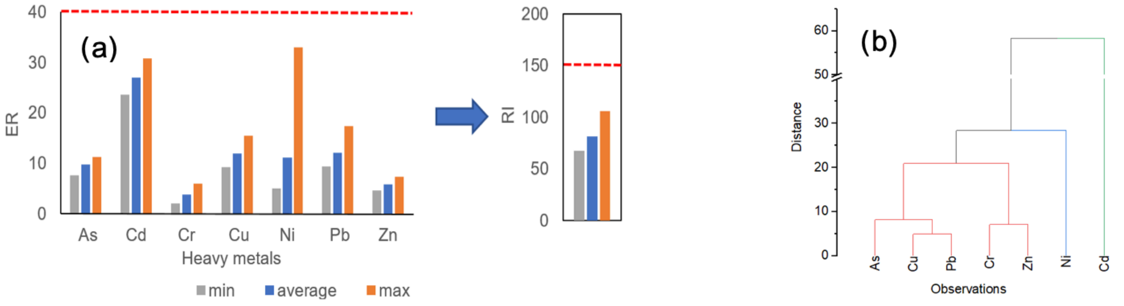


Figure 4. ER and RI values of heavy metals (a) and dendrogram for IR values (b).

Evaluation of the Indices

The heavy metal concentration of sewage sludge was not enough for determining the appropriateness or not the agricultural application alone, thus some calculations based on predefined indices were necessary to clarify the pollution, toxicity and risk degree. Although these indices were generally used for determining soil and sediment pollution, nowadays they applied the sewage sludge [9,32].

In this study, though C_f and PI were calculated with the same formula, the results of the indices were evaluated according to their different ranges. Therefore, some differences were observed in the interpretation of the calculated values. For example, C_f and PI for Cr were found to be 1.9, according to the evaluation table of the indices (Table 1), this value was interpreted as moderate pollution in C_f and low pollution in PI.

Although, similar interpretation of results was observed between I_{geo} and C_f calculations the pollution level of some indices varied due to their different calculation methods. During the PI calculations, C_s value used as background soil heavy metal content and legal standards [13]. PI and Nemerow index values based on legal standards were lower than C_s based on background soil. Higher C_f values depend on the lower background soil heavy metal content in this study and Zn concentration in sewage sludge was far more than average soil background value. The sewage sludge land application regulations allow higher heavy metal concentrations than soil background values, thus the differences were originated from this situation.

Based on ecological risk calculations, there is no risk potential was estimated due to the low ER and RI values, on the other hand, pollution indices demonstrate the contamination level was high for some heavy metals such as Zn and Se. The reason for this situation is the soil background contained low heavy metal content. Besides, the contamination degree of the samples based on before mentioned indices can be decreased with mobility and degradation of the heavy metals.

Statistical methods such as hierarchical cluster analysis (HCA), principal component analysis (PCA) and regression analysis (for I_{geo}, C_f, PI, PN, ER indices) can be applied to the comparison of indices. The dendrograms for indices were shown in Fig. 5. The dendrogram based on I_{geo}, C_f, PI, PN, ER indices can be divided into one cluster and singleton. ER index included singleton because ER was used to determine the ecological risk and others are commonly used to determine the pollution degree of heavy metal (except mERM-Q). C_f and PI were the same clusters due to they calculated with the same equation. EF was similar to I_{geo} consistent with the literature due to they both involving soil background values [23]. I_{geo} and mERM-Q were the most similar to each other even though they used different purposes, but they can group in one cluster. PN and PI can be grouped in another cluster because PI (or C_f) values used in PN calculations.

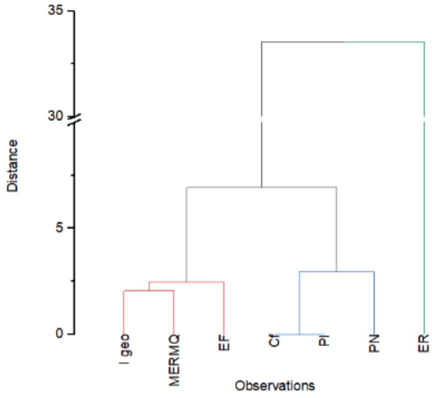


Figure 5. Comparison of indices.

The risk level calculations based on mERM-Q index were found medium to high while the ecological risk calculation from ER and RI demonstrated that there was no risk occur when sewage sludge applied to the land. The reason for this can be due to the ERM values used in the mERM-Q index calculations.

Although the ER and I_{geo} indices are mostly used in the risk and pollution assessment of the sewage sludge in the literature [4,16,19], in this study, these assessments were made from different perspectives by applying the indices.

CONCLUSION

The domestic sewage sludge samples are suitable for use instead of fertilizers in terms of having low heavy metal content (within the permissible values permitted by the legal standards) and high nutrient content. However, it is imperative to determine the potential ecological effects of heavy metal content. According to pollution indices, the pollution degree varied from “uncontaminated” to “considerably contaminant” and Zn caused the highest and As caused the lowest contamination in sewage sludge. Because, soil background value was lower than sewage sludge samples. On the other hand, all samples posed a low risk based on ER and IR indices. To ensure the safetly land application of sewage sludge, health risk assessment is recommended. At the same time, ecological and health risk assessment studies are site-specific, even the heavy metal concentration’s is the same, the ecological and health risks maybe quite different if the sludge applied in different sites.

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AUTHORSHIP CONTRIBUTION

Authors equally contributed to this work.

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DATA AVAILABILITY STATEMENT

The authors confirm that the data that supports the findings of this study are available within the article. Raw data that support the finding of this study are available from the corresponding author, upon reasonable request.

CONFLICT OF INTEREST

The author declared no potential conflicts of interest with respect to the research, authorship, and/or publication of this article.

ETHICS

There are no ethical issues with the publication of this manuscript.

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