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RESEARCH ARTICLE

Occurrence of organic micropollutants and heavy metals in the soil after the application of stabilized sewage sludge

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Abstract

Stabilized sewage sludge could be used as a fertilizer in the soil because of nitrogen, phosphorus, and other micronutrition material to improve the properties of the soil. In this study, clover and fodder corn plants were planted in the soil to which stabilized sewage sludge was applied. The soil was analyzed and evaluated by collecting samples to determine changes in the application fields before plantation and one year after the harvest. To monitor changes in the soil and after the application of stabilized sludge to the soil, the stabilized sewage sludge was evaluated for the suitability of parameters such as heavy metals, halogenated organic compounds including adsorbable organic halogens (AOX), nonylphenol ethoxylates (NPE), polycyclic aromatic hydrocarbon compounds (PAH), polychlorinated biphenyls (PCB), di(2-ethylhexyl) phthalate (DEHP), linear alkylbenzene sulphonates (LAS), polychlorinated dibenzo-p-dioxins and dibenzo-p-furans (PCDD/F), and pathogens and other parameters containing organic matter (OM), pH, conductivity (EC), total nitrogen (TN), and total phosphorus (TP). After the sludge application, the heavy metal content increased, except for Zn. The application of stabilized sludge as a soil conditioner and humus caused a significant increase in the TN and TP content since the product increased the harvest that increased soil nutrition. After the sludge application to the soil, the concentration of organic micropollutants in the soil did not change. The concentration of AOX and PCDD/F in the soil increased by 141% and 125%, respectively.

Keywords Agriculture · Biosolid · Clover · Fodder corn · Wastewater

Introduction

Stabilized sewage sludge varies with raw wastewater characterization and process type, so the amount of sludge changes. The sludge has high organic matter content with macro and micro plant nutrition matter and can be applied to the soil under control, as has been determined in some studies [1]. Furthermore, if stabilized sewage sludge contains heavy metals, organic pollutants, and pathogenic organisms and is used in soils, it can harm the environment, as shown by some researchers [2, 3]. In many studies conducted, it has been determined that stabilized sewage sludge could be used as a fertilizer in the soil because of nitrogen, phosphorus, and other micronutrition material to improve the physical and chemical properties of the soil [4, 5]. According to the EU Directive 86/278/EEC, the national legislation, which has been established in the member states, prohibits the use of sludge in agriculture if the concentrations of heavy metals exceed specific limit values, but it sets no limits for organic contaminants [6].

The limit values for heavy metals in sludge defined in the regulations of different countries are given in Table 1 [7]. The majority of the EU countries in Table 1 adopted even more stringent limits for sludge use in agriculture by setting lower limits for heavy metals compared to the European Directive 86/278/EEC (CEC, 1986) [8].

To control the solubility of metals in the sewage sludge that is applied to the soil, both the sludge and soil pH must be monitored continuously. Therefore, the Turkish legislation states that the pH of the soil must be 6.5 or higher. This parameter restricts the behaviors of heavy metals. After the application of sewage sludge to the soil at increasing doses, there is a decrease in pH, an increase in conductivity, an increase in TN, and an increase in reactive phosphorus, as indicated by the studies. In soils treated with sewage sludge, pH is the most effective parameter for the bioavailability of heavy metals. According to the researcher (2001), if EC

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Table 1 Limits of heavy metals for sludge use in agriculture

State	Heavy Metals (mg/kg of dry sewage sludge)						
	Pb	Cd	Cr	Cu	Ni	Zn	Hg
Directive:86/278/EEC	750–1200	20–40	-	1000–1750	300–400	2500–4000	16–25
Turkey*	750	10	1000	1000	300	2500	10
Austria (Voralberg)	150	4	300	500	100	1800	4
Belgium	800	10	500	600	100	2000	10
Bulgaria	800	300	500	1600	350	3000	16
Denmark	120	0.8	100	1000	30	4000	0.8
Finland	100	1.5	300	600	100	1500	1
France	800	20	1000	1000	200	3000	10
Germany	900	10	900	800	200	2500	8
Italy	750	20	-	1000	300	2500	10
Netherlands	100	1.25	75	75	30	300	0.75
Romania	300	10	500	500	100	2000	5
USA (part 503)**	840	85	3000	4300	420	7500	57

is <2 dS/m, the salinity effect can be ignored, but if it is between 2 and 4 dS/m, salinity can be restricted for very sensitive products [9]. An increase in EC, when the parameters are around (2.55–2.75 dS/m), meaning high salinity content, suggests that it results from the intense application of sewage sludge to the soil [10]. It was revealed the application of the high heavy metal content of sludge to the same field for a long period could cause an increase in the heavy metal content in the soil to produce an important risk factor due to the restricted operational ability of such elements in the soil [11].

The concentrations of heavy metals (Zn, Cr, Cu, Ni, Pb, and As) in the sewage sludge of Xuchang city, China, were evaluated as the contamination levels and environmental risk for agriculture [12]. The agricultural soil should always be treated with a composted product, and biological properties should be enhanced because there will be no adverse impact on the mentioned properties and the legislation regarding the concentration of heavy metals in the waste to be added is considerably less strict [13].

The limits of heavy metals in the sludge-treated soil (mg/kg dry soil) in the EU member countries and Turkey are presented in Table 2. As can be seen in Table 2, different heavy metal limit values are applied depending on the pH value of the soil.

The suggested limit values for organic compounds were included in the third draft of the Working Document on Sludge (European Commission, 2000a) and the national legislation of EU countries [14] and Turkish directive (Table 3). The most common limit is the limit for AOX, which is the same for all countries that have introduced the mentioned limit. The value is 500 mg per kg DM of sludge. The other most common limits are for PCDD/F, PAHs, and PCBs.

In a study, it was determined that when urban sewage sludge was applied to a limy soil at different levels, the

Table 2 Limits of heavy metals in the soil

State	Heavy Metals (mg/kg dry soil)						
	Cd	Cr	Cu	Hg	Ni	Pb	Zn
Directive: 86/278/EEC	1–3	-	50–140	1–1.5	30–75	50–300	150–300
(6 < pH < 7)	1	60	50	1	50	70	150
Turkey (6 ≤ pH < 7)	1.5	100	100	1	70	100	200
Turkey (pH ≥ 7)	1.5	-	50	1	20	50	200
Belgium	2	200	140	1	-	100	250
Bulgaria (6 < pH < 7.4)	0.5	30	40	0.5	15	60	150
Denmark	0.5	200	100	0.2	60	60	150
Finland	2	150	100	1	50	100	300
France	1.5	100	60	1	50	100	200
Germany	1.5	-	100	1	75	100	300
Italy	0.8	100	36	0.3	35	85	140
Netherlands	3	100	100	1	50	50	300
Romania (pH > 6.5)							

Table 3 Limits of the selected organic micropollutants for sludge use in agriculture (in mg/kg DM of sewage sludge, except for PCDD/F, which is in ng toxic equivalency (TEQ/kg DM)

State	Organic Micropollutants						
	AOX	DEHP	LAS	NP/NPE	PAH	PCB	PCDD/F
EC: (2000)[a]	500	100	2600	50	6	0.8	100
Turkey	500	100	2600	50	6	1	100
Austria (Carinthia)	500	4	300	500	6	1	50
Belgium	–	–	–	–	–	0.8	–
Bulgaria	–	–	–	–	6.5	1	–
Denmark	–	50	1300	10	3	–	–
Portugal			5000	450	6	0.8	100
France	–	–	–	–	2–5	0.8	–
Germany	500	–	–	–	–	0.2	100
Sweden	–	–	–	50	3	0.4	–
Luxembourg	–	–	–	–	20	0.2	20
Romania	500	–	–	–	5	0.8	–

sludge affected the pH, electrical conductivity (EC), cation exchange capacity (CEC) values and organic matter (OM), total nitrogen (N), reactive phosphorus (P), extractable potassium (K), and nitrate (NO_3^-) parameters. It was also determined that the sludge application affected the ammonium (NH_4^+) parameter but only at a statistically insignificant level [15].

Some European countries have introduced more stringent requirements compared to the directive and have adopted limits for concentrations of other heavy metals, synthetic organic compounds, and microbial contamination. The suggested limit values for particular synthetic organic compounds, such as halogenated organic compounds, linear alkylbenzene sulphonates (LAS), phthalates, nonylphenol, PAHs, PCBs, and PCDD/Fs, were included in the third draft of the Working Document on Sludge (European Commission, 2000a) and the national legislation of EU countries [14]. For the purpose of reducing the possible adverse impacts due to the agricultural usage of biosolids, specific actions must promote the achievement and utilization of sewage sludge with low heavy metal content and the identification of soils that are more sensitive to heavy metal accumulation, which may cause the ecotoxicity or increased metal content in crop products [16]. Despite the fact that sewage sludge represents a rich source of nutrients for arable farming and soil improvement, it may also be a source of pollutants [17]. Moreover, sewage sludge can contain a wide spectrum of harmful toxic substances, e.g. heavy metals, PAHs, PCDD/Fs, PCBs, DEHP, polybrominated diphenyl ethers (PBDEs), detergent residues, pharmaceuticals, personal care products, endogenous hormones, synthetic steroids, etc. [18]. (Wilson et al., 1994) concluded that the application of sludge to land at typical application rates would increase the soil VOC content, but this elevation was unlikely to create a cause for concern [19]. However, it is recommended to monitor field samples. As a well-known main source of contaminants, the industry is replaced by households at an increasing rate, as in the case of pharmaceuticals (PhCs) and personal care products (PCPs) [20]. The suggested changes in the EU Directive require the control of toxic organic micropollutants in sewage sludge applied in agriculture, such as PAHs, PCBs, PCDD/PCDF, DEHP, NPE, AOX, and LAS [21]. The results demonstrate that the combined impact of PAHs and heavy metals on microorganism activity in the soil and on some plants at an early stage of their development may be stronger than in soils amended with HM or PAH separately. The reaction of the tested organisms was related to soil properties, PAH concentration, time, and plant species. Mature plants (maize) were insensitive to the applied levels of both groups of contaminants [22]. As for organic contaminants, the 3rd draft of the "Working Document on Sludge" (European Commission, 2000) suggested limit values for the so-called sum of halogenated organic compounds (AOX), linear alkylbenzene sulphonates (LAS), di(2-ethylhexyl)phthalate (DEHP), nonylphenol and nonylphenol ethoxylates (NP/NPE), polynuclear aromatic hydrocarbons (PAH), polychlorinated biphenyls (PCB), and polychlorinated dibenzo-p-dioxins and -furans (PCDD/F) [23]. The importance of the phthalate plasticizer compound, DEHP, in sewage sludge for human health and the environment requires careful and appropriate interpretation. Recently, DEHP has been downgraded as a carcinogen by IARC. Moreover, the US ATSDR reached a conclusion that there were no pieces of evidence to show that DEHP was an endocrine disrupter in humans at the levels in the environment, which is reinforced by the comparatively rapid degradability of DEHP in the soil and the absence of significant transfers of DEHP to food crops. The risk assessment of DEHP in the sludge-treated agricultural soil is suggested to clarify the effect of the said plasticizer compound on human health, soil quality, and the environment [24]. The resulting organic contaminants in sludge would make a further contribution to the technical evidence base to protect human health and the environment in case of recycling sewage sludge biosolids in agriculture as soil improvers and fertilizers [25]. An investigation in the real environment is more difficult, and the clear-cut interpretation of the results is frequently impossible due to the variety of factors that influence changes in the concentration of PAHs [26]. Future studies must focus on establishing the PAH concentrations in soils and vegetation in the most impacted zones and on enlarging the study to perform measurements of PAH levels in the air in order to observe the relationship between the different media [27]. In the soil, the biogenic production of PCB and PCDD/F is possibly negligible in comparison with the anthropogenic input [28]. Regardless of the AOX concentrations in the wastewater influent and the treatment efficiency, the AOX in the sludge produced is formed mainly inside the wastewater plant. The AOX concentration in the sludge exposed to the sun for a period of three months was decreased to two-thirds of its original concentration. In order to reduce potential environmental and health hazards, it is suggested that sludge should be exposed to the sun for several months before its use in agriculture [29].

In the present study, the usability of the sewage sludge, which was stabilized and dewatered from the Konya WWTP, one of the largest capacity treatment plants in Turkey, was evaluated for its use in the soil. In this context, the characterization of the WWTP sludge, the soil properties in the application fields, the application of sludge to agricultural fields according to the Turkish legislation, and changes in the treated soil one year after the sludge application were investigated.

Materials and methods

The study area

In the current study, the usability of the sewage sludge stabilized in anaerobic digestion from the Konya (Turkey) WWTP with a flow rate of 200.000 m^3/day was evaluated. The Konya WWTP has the following processes: pre treatment, four-stage Bardenpho process, UV dsinfection, sludge thickeners, sludge stabilization and sludge dewatering. Sludge stabilization was carried out by operating four anaerobic digesters, each with a volume of 7000 m^3. The mixing process required for stabilization was carried out with the mechanical mixer in the digester. The sludge taken from the thickeners was heated by means of heat exchangers and the digester was operated at a temperature of 36–38 °C under mesophilic conditions. SRT was 17 days in the each digesters. Stable sludge was dewatered to a solids content of 25–28% in the sludge dewatering unit.

Data description and analysis

In two different lands (Konya, Turkey), sewage sludge was applied to the soil within the legislative limits (Table 4). Clover was cultivated in application Field-1 of 24,565 m^2, and fodder corn plants were cultivated in application Field-2 of 37,334 m^2. The stabilized sewage sludge applied soil was analyzed and evaluated by collecting samples to determine changes in the main control parameters in the application fields before plantation and one year after the harvest. Before the application, sludge and soil samples were taken to determine the main control parameters for the conformity with the values specified in the legislation. The representative soil samples from the application field taken from different locations at a depth of 10–25 cm were prepared by mixing twenty-five samples [30].

During this study, the following analysis methods of the parameters were used; pH: TS 836 method, OM: TS 8336 method, TN: SM 4500 method, TP: Olsen method, Heavy Metals: EPA 3051 A:2007 method, EC: TS ISO 11265 method, *E. coli*: SM 9221:F:2012 method, PAH: EPA 3540C:1996/EPA 8270D:2007 method, PCB: EPA 3665A:1996/EPA 3540C:1996 method, AOX: TS EN ISO 9562 method, PCDD/F: EN 1948–1,2 method, LAS: PR.504.301 + SM 5540 C method, NPE: CEN/TS 16182:2012 method, DEHP: EPA 8270 D:2007 method.

Results and discussion

After the application of the stabilized sewage sludge to the soil, clover and fodder corn were planted in the field. One year after the harvest, samples were taken from the same field and analyzed to determine the accumulation alteration of the main control parameters of the soil. The analysis results of the composed sample taken from the stabilized sludge produced are given in Table 4.

The limit values for heavy metals, halogenated organic compounds, dioxins, pathogens, etc. in sludge defined in the European Union (EU) [8], EC: (2000)[a] and Turkish regulations are presented in Table 4. Heavy metals in stabilized sewage sludge were determined to be very low in comparison with the allowable limits: The Konya WWTP does not accept industrial wastewater, and as a result, the heavy metal parameters there are at very low levels. The halogen organic compounds needed to be controlled in stabilized sludge are also considerably below the allowable limits: AOX 2.51 mg/kg, NPE < 2 mg/kg, PAH < 0.1 mg/kg, PCB < 0.1 mg/kg, LAS < 22 mg/kg, DEHP <22 mg/kg, and dioxins PCDD/F 0.44 mg/kg. As one of the main control parameters to permit

Table 4 The characterization of stabilized sewage sludge and its comparison with the EU and TR legislation

Parameters	Unit	EU Directive: 86/278/EEC	Turkey Directive: 2010/27661.	Stabilized sewage sludge analysis results
Heavy Metals				
Pb	mg/kg	750–1200	750	24.22
Cd	mg/kg	20–40	10	2.53
Cr	mg/kg	–	1000	154.12
Cu	mg/kg	1000–1750	1000	143.40
Ni	mg/kg	300–400	300	49.12
Zn	mg/kg	2500–4000	2500	1005
Hg	mg/kg	16–25	10	0.33
Halogenated Organic Compounds		EC: (2000)[a]		
AOX	mg/kg	500	500	2.51
NPE	mg/kg	50	50	< 2
PAH	mg/kg	6	6	< 0.1
PCB	mg/kg	0.8	1	< 0.1
LAS	mg/kg	2600	2600	< 22
DEHP	mg/kg	100	100	< 22
Dioxins				
PCDD/F	ngTE/kg	100	100	0.44
Pathogen				
E.Coli	%Removal	–	99.00	99.40
Others				
OM	%	–	≥ 40	52.37
pH		–	6–8.5	7.66
EC	dS/m	–	–	3.71
TN	mg/kg	–	–	22842
TP	mg/kg	–	–	7627

the use of stabilized sewage sludge in soil conditioning, the *E. coli* removal was realized at 99.40%, and this value also approves the use of sludge. In the Konya WWTP, mesophilic anaerobic digesters are used for the stabilization of sludge. It is observed that the anaerobic sludge digesters operated at high efficiency provided high rates of pathogenic removal. The OM 52.37% and pH 7.66 parameters are other parameters the sludge has to conform with according to the limits specified in the legislation. The legislation does not specify limits for some parameters. However, in soil application, they are mentioned as the parameters that should be under control. These parameters are determined as follows: EC 3.71 dS/m, TN 22842 mg/kg, and TP 7627 mg/kg. It was determined that the stabilized sludge used in this study conformed with the EU and Turkish legislation and the parameters to be controlled were within the limits for sludge use in the soil.

Although standard parameters are not specified in the legislation, TN and TP with the EC values affecting the behavior of heavy metals must be monitored. In this study, the soil parameters were evaluated and controlled before and after the stabilized sewage sludge application to monitor the changes. The main control parameters (Pb, Cd, Cr, Cu, Ni, Zn, Hg, AOX, NPE, PAH, PCB, LAS, DEHP, PCDD/F, OM, TN, TP, EC, pH) were checked in the soil before and after the application in two different locations, as given in Table 5. Because the pH ≥ 7 value was found in the application fields, the respective parameter and limit criteria were used as a reference.

The heavy metal and organic matter parameters were discussed for the data obtained from the two different fields chosen. With the treatment sludge application, it was noted that the organic matter content of the soil, as well as NH_4, NO_3, available P, exchangeable K, Ca, Mg, Na and available Fe, Cu, Mn, Zn and B contents increased [31]. They reported that heavy metal concentrations in the soil increased and this event could be due to the effect of increasing soil pH and retention and adsorption [32]. The total amount of phosphorus in the soil may be high, but it is usually in a from that cannot be used by the plant [33].

When the 24565 m^2 area where clover was planted was analyzed, heavy metals were observed to alter as follows: an

Table 5 Soil analysis results before and after the application of stabilized sewage sludge

Parameters	Unit	Field 1			Field 2			Turkey Legislation limit value (pH ≥ 7)
		Before the application	After the application	Change (%)	Before the application	After the application	Change (%)	
Pb	mg/kg	10.50	11.46	9	11.75	12.43	6	100
Cd	mg/kg	0.25	0.28	12	0.25	0.34	36	1.5
Cr	mg/kg	26.00	56.33	117	36.75	52.90	44	100
Cu	mg/kg	11.25	15.54	38	13.77	14.80	7	100
Ni	mg/kg	30.75	56.38	83	36.75	38.69	5	70
Zn	mg/kg	31.75	19.64	−38	37.25	22.48	−40	200
Hg	mg/kg	0.25	0.32	28	0.25	0.29	16	1
AOX	mg/kg	< 20	44.30	122	< 20	33.51	68	-
NPE	mg/kg	< 0.37	< 0.37	-	< 0.37	< 0.37	-	-
PAH	mg/kg	< 0.05	< 0.05	-	< 0.05	< 0.05	-	-
PCB	mg/kg	< 0.004	< 0.004	-	< 0.004	< 0.004	-	-
LAS	mg/kg	< 2	< 2	-	< 2	< 2	-	-
DEHP	mg/kg	< 0.75	< 0.75	-	< 0.75	< 0.75	-	-
PCDD/F	ng TE/kg	0.34	0.82	141	0.59	1.33	125	-
OM	%	1.87	3.08	65	0.84	4.69	458	≤ 5
TN	ppm	2220	13193	494	3970	9883	149	–
TP	ppm	2.03	13.00	540	2.24	20.80	829	–
EC	µs/cm	151	117	−22	146	180	23	–
pH	–	7.97	8.15	2	7.50	7.86	5	–

increase of 9% in Pb,12% in Cd,117% in Cr,38% in Cu,83% in Ni, 28% in Hg, and a decrease of 38% in Zn were determined. The only decrease was in Zn, and the maximum increase was in Cr. After the application, it was determined that the heavy metal values were considerably below the allowable limits.

When the 37334 m^2 area where fodder corn was planted was analyzed, heavy metals were determined to alter as follows: an increase of 6% in Pb,36% in Cd,44% in Cr,7% in Cu,5% in Ni,16% in Hg and a decrease of 40% in Zn were determined. The only decrease was in Zn, and the maximum increase was in Cr. After the application, it was determined that the heavy metal values were considerably below the allowable limits. Zn concentration in the soil solution decreased with increasing soil pH value. This is because the adsorption of Zn to the soil increases at higher pH [34–37]. Zn, directly available for plant uptake [38–40].

The concentrations of TN and TP before and after the sludge application in Field-1 and Field-2 are given in Fig. 1. After the application of sludge to Field-1, an increase of 494% occurred in TN. In Field-2, an increase of 149% occurred in TN The increase in TN may have resulted from the fact that the bacteria living together with the roots of clover collected the free nitrogen in the air and stored it in the roots. The same bacteria could not cooperate with the fodder corn roots.

After the application of sludge to Field-1, TP increased by 540%. In Field-2, TP increased by 829%. In the application fields, the TP parameter altered very much, and the phosphorus value of the soil improved significantly. The alterations in OM before and after the sludge application in fields are given in Fig. 1. After the application of sludge to Field-1, OM increased by 65% from 1.87% to 3.08%. In Field-2, OM increased by 458% from 0.84% to 4.69%. In the application fields, the OM parameter altered very much, and the OM value of the soil improved significantly. After the application, the parameter value was still below the allowable values specified in the legislation (OM ≤ 5). The increase in OM may have resulted from the plant species and management. In corn harvest, the soil might have transferred many plantation tissues, or the roots of corn might have excreted more assimilated matter than clover. The alterations in EC before and after the sludge application in fields are given in Fig. 1. After the application of sludge to Field-1, EC decreased by 22%. In Field-2, EC increased by 23%. In the application fields, the EC parameter altered according to the soil properties. The EC of sludge is higher than the EC of the soil. The alterations in pH before and after the sludge application in fields are given in Fig. 1. After the application of sludge to Field-1, pH values increased by 2% from 7.97 to 8.15. In Field-2, pH values increased by 5% from 7.50 to 7.86. In the application fields, no significant pH alterations were observed.

PCBs are also hazardous due to their persistence and toxic properties. PAHs have toxicity to humans and

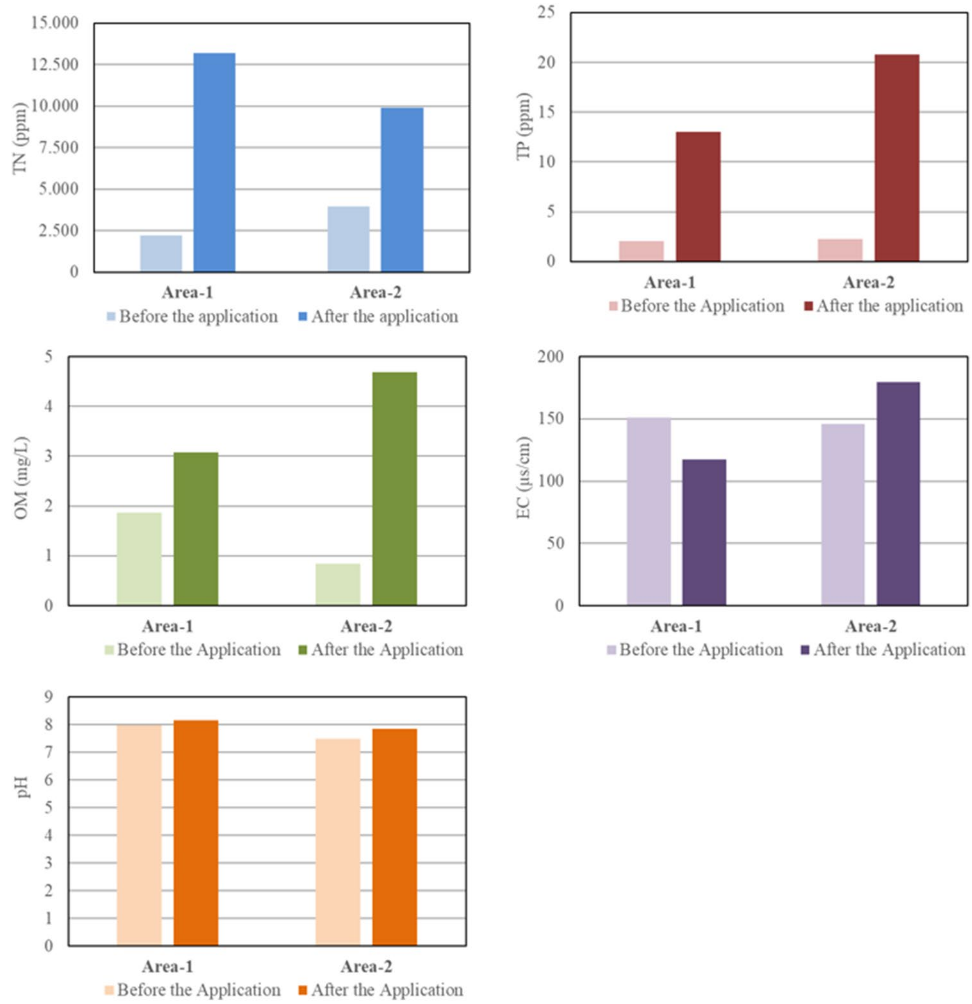

Fig. 1 TN, TP, OM, EC, and pH concentrations before and after the sludge application in Field-1 and Field-2

deteriorative effects on soil organisms and plants [41]. In the past studies on animals with PAHs, it has been observed that they have different toxic properties as well as mutagenic and carcinogenic effects [42]. NPs cause damage by affecting the skeletal and muscular systems in living things of plants it prevents growth by damaging roots and chloroplasts [43]. The important negative health effects of these compounds, known as carcinogenicity, which cause accumulation in the body of people who interact with these environmental environments, make it absolutely necessary to determine the mechanisms, amount of the accumulation and to take the necessary precautions [44].

In Field-1 and in Field-2 where clover was planted, AOX and PCDD/F concentrations were increased. Since NPE, PAH, PCB, LAS and DEHP parameters remained below the LOQ limits, no change was observed (Fig. 2).

Conclusion

By using the stabilized sewage sludge from the WWTP in the soil in real-scale applications, important conclusions were reached when the following data on the main control parameters were received and analyzed. In Turkey, since it is the first and only licensed application, the results of the application are very important for both the government and the municipalities concerning the sustainability of the application.

In this study, stabilized sewage sludge was checked for the conformity to the legislative main control parameters, such as heavy metals, halogen organic compounds (the halogen organic compounds that have to be controlled in stabilized sludge such as (AOX, NPE, PAH, PCB, LAS, DEHP), dioxins (PCDD/F), organic matter, pH, and *E. coliform*. The limit values of sludge were achieved, and it was decided that the sludge could be used in soil conditioning. Furthermore, the parameters not defined in the legislation

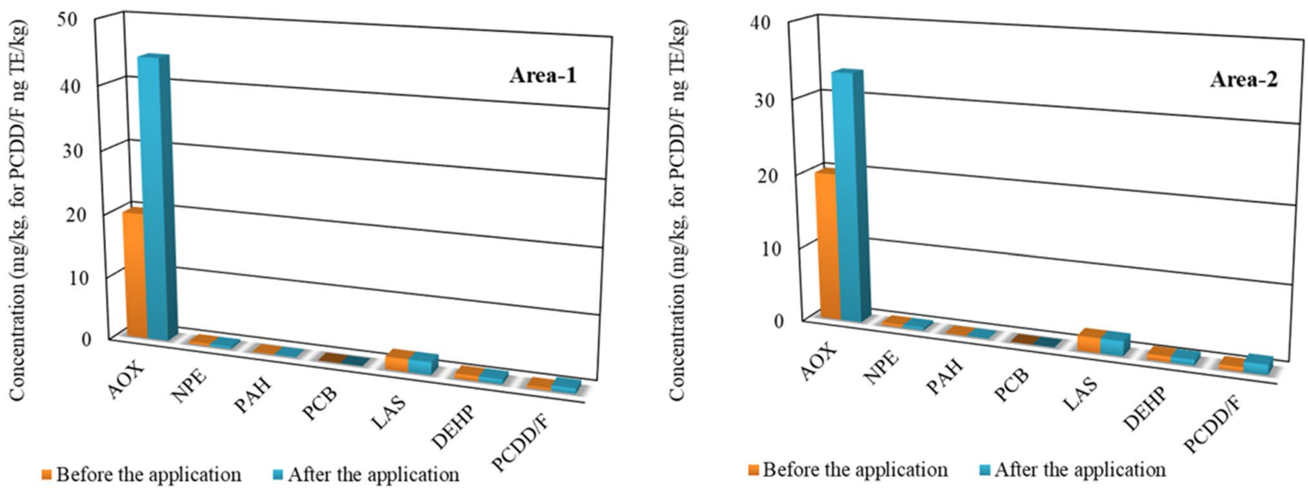

Fig. 2 AOX, NPE, PAH, PCB, LAS, DEHP, and PCDD/F concentrations before and after the sludge application in Field-1 and Field-2

for sludge were checked and determined to be within the allowable limits for the soil quality. These parameters were found to be as follows: EC 3.71 dS/m, TN 22842 mg/kg, and TP 7627 mg/kg.

The standard values of pH criteria were pre-determined for the soil that would receive the stabilized sewage sludge application. The soil in the application fields had a pH value of ≥7, and the heavy metals with OM values were considerably below the allowable limits. Furthermore, the TN and TP limits were not given as control parameters, but the analysis was conducted before and after the sludge application because they affected the behavior of the soil like conductivity.

After the sludge application, the heavy metal content increased, except for Zn. Solubility is inversely proportional to the pH of the soil. As pH increases, solubility decreases and vice versa. Clover and corn plants can receive nutrition if the pH of the soil is between 5.5–8. The plants in the study fields could receive Zn from the soil easily since the pH was at this point and the Zn content of the soil decreased. Dalpe et al., stated that maize and alfalfa plants are highly dependent on mycorrhiza [45]. Marschner stated that mycorrhiza contributes to approximately 50% of Zn uptake by plant. [46].

The TN and TP parameters were also checked, although they are not legislative prerequisites. The application of stabilized sludge as a soil conditioner and humus caused a significant increase in the TN and TP content since the product increased the harvest that increased soil nutrition. Nitrogen deficiency is a result of lower OM content in the soil. Therefore, the high nitrogen content of the sludge used for soil conditioning is helpful for soil amendment in general. Phosphorus deficiency causes less crop harvesting of plants in the soil. Therefore, the high phosphorus content of the sludge used for soil conditioning is helpful for the soil amendment in general.

In the application fields, the OM parameter altered very much, and the OM value of the soil improved significantly. After the application, the parameter value was still below the allowable legislation values (OM ≤ 5). However, the determined values were close to the parameter limits. The stabilized sewage sludge used in this study contained over 40% of OM. OM amends the physical, chemical, and biological properties of the soil and helps plants get the nutrition elements. As the OM content of sludge increases, the quality also increases. Stabilized sewage sludge can be used for conditioning the agricultural soil with poor OM content. Although the same sludge was used in both test fields, the EC behavior differed due to the soil properties. The decrease in the clover plantation field may be due to the sprinkler irrigation system used that might have moved the OM below the root level. The EC increase in the fodder plantation field might have resulted from high EC sludge (3700 µs/cm) use and the use of the drip irrigation method that could not wash away the salt from the roots.

During and after the sludge application, no significant changes occurred in pH values, and the plants received the necessary nutrition easily since the plants received nutrition when the pH values were between 5.5 and 8.

After the sludge application to the soil, since NPE, PAH, PCB, LAS and DEHP parameters remained below the LOQ limits, no change was observed. AOX and PCDD/F concentrations increased in Field-1 and Field-2. There are no limit values for organic micropollutants in the EU and TR regulations. Organic halogens have the carcinogenic effects but it is determined that all of the organic pollutants in the stabilized sludge used in this study are below the legal limit values. In addition, it is also determined that the AOX and

PCDD/F parameters of the pollutants in the soil after the application are above the LOQ value and below the permissible limit values for sludge.

According to this study, sludge can be used at least for one-year periods. The monitoring of the land must be performed after the application, and if the main control parameter values are close to the limits, then the land of the application must be changed. The transformation of heavy metals to the food chain must be monitored scientifically, and different scenarios should be developed according to the data obtained. Similar applications in the world and Europe must be followed scientifically, and local application techniques and legislative organization efforts should be made.

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Author contributions Serdar Koyuncu (Ph.D) conducted all the experiments and wrote the manuscript.

Declarations

Conflict of interest The author declare that they have no conflict of interest.

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