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Present restrictions of sewage sludge application in agriculture within the European Union

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Abstract: The use of sludge in agriculture within the European Union (EU) is currently regulated only by the limits of heavy metals (Cd, Cu, Hg, Ni, Pb and Zn) listed in Council Directive 86/278/EEC. This document is now more than 30 years old. Several European countries have introduced more stringent requirements in comparison with the directive, and have adopted limits for concentrations of other heavy metals, synthetic organic compounds and microbial contamination. The paper provides an overview of the current limits of these substances in sewage sludge and concentration limits of heavy metals in soil intended for sludge application, together with applicable laws and regulations in European Union countries. There is a need to update these regulations taking into account the current risks associated with the application of sludge to agricultural land, with the possibility of using ecotoxicological tests to assess the risks. A wide range of technologies for sewage sludge processing is used in EU countries. The predominant choice is a direct application in agriculture followed by composting. The use of sewage sludge in agriculture in 2014 and 2015 in 13 EU countries that provided data amounted to 22.6% (2014) and 22.1% (2015) of produced sludge and 23.3% (2014) and 23.1% (2015) of sludge disposed. It is also highly variable within EU countries ranging between zero (Malta, Slovenia, Slovakia) and 80% (Ireland). Over 50% of sewage sludge is used in agriculture in Bulgaria according to 2015 data.

Keywords: agricultural land; European countries; legislation; limit; sewage sludge disposal

Application of sewage sludge or compost blended with sludge to soil used for crop production is of great importance with regard to the supply of organic matter and nutrients, especially nitrogen and phosphorus (Mantovi *et al.* 2005; Fytili & Zabaniotou 2008; Singh & Agrawal 2008; Urbaniak *et al.* 2016). Further it contains high concentrations of Ca and Mg, but the K content is low (Singh & Agrawal 2008). The increase of organic matter can improve physical (water retention, soil texture, water infiltration, bulk density, porosity) and chemical soil properties (higher cation exchange capacity (McBride *et al.* 1997; Shuman 1998), pH). The biological properties (Moss *et al.* 2002; Andreoli *et al.* 2007), which are necessary for long-term soil fertility (Draeger *et al.* 1999) and nutrient availability due to the aliphatic compounds with low molecular weight which strongly interact with soil minerals, are also positively affected (Hue & Ranjith 1994). The higher availability of organic matter and nutrients also increases the activity of soil enzymes as well as soil microbial activity and soil microbial biomass growth (Singh & Agrawal

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2008). The factors that affect the bioavailability of elements in soil include the source of biological waste, its character, composition and processing, soil pH, organic matter content, condition of the soil, chemistry of elements, plant species and cultivars, growth phase and others (Bertran *et al.* 2004; Mantovi *et al.* 2005; Warmar & Termeer 2005). It is also important to take into account the sludge to soil ratio (Singh & Agrawal 2008).

In general, sludge is characterized by considerable variability in nutrient content depending on the wastewater source and treatment process (Moss *et al.* 2002). Organic nitrogen and inorganic phosphorus constituted the majority of total nitrogen (TN) and total phosphorus (TP) content in sewage sludge (Singh & Agrawal 2008). Uggetti *et al.* (2010) published the percentage ratio of three nutrients in the sewage sludge of a wastewater treatment plant (WWTP) for 1500 population equivalent (PE): total nitrogen 9.76%, total phosphorus 2.68% and potassium (K) 0.27% of total solids. Singh and Agrawal (2008) presented a summary of surveys in Thailand, Spain and India. The range of total nitrogen, total phosphorus and potassium content as a percentage did not vary very much with values between 2.5 and 3.4% of TN, 1.06–1.34% of TP and 0.2–0.42% of K. Gascó and Lobo (2007) published the amount of TN in sewage sludge 1.5% and amount of TP 1.2 g/kg.

Although nutrients are essential for plant growth, when applied excessively (especially nitrogen and phosphorus) they may accumulate in soil and can be leached and transported by drainage systems (particularly nitrogen) or can be transported by water erosion (phosphorus bound to soil particles) and pose a risk to surface water and ground water (Hernandez *et al.* 1999; Walter *et al.* 2000; Korboulewsky *et al.* 2002).

The amount of nitrogen and phosphorus in the sludge that will be available through the mineralization of organic matter depends on the crop that was previously cultivated (nutrients from crop residues) and the type of soil before applying sludge to the land, which should be considered on a case-by-case basis. Mineralization is quicker on sandy soil than on clay soil. Nitrogen available through mineralization will result in lower fertilizer requirements. In the first year after the application of biowaste, the available nitrogen value is 50% (Andreoli *et al.* 2007).

Phosphate rock is listed as one of the 20 critical raw materials for the European Union in the "Report on critical raw materials for the EU" published by the European Commission in 2014 (European Commission 2014). Many studies and guidances deal with improving nutrient use efficiency (Cassman *et al.* 2002; FAO 2008; Roberts 2008) in agroecosystems and crop systems.

Sludge phosphorus reaching bioavailability ranging from 40% to 80% (Andreoli *et al.* 2007) represents a renewable source of phosphorus. The efficiency of chemical fertilization becomes very low, mainly because of the high soil capacity to fix P. The efficiency of P fertilizers is impacted by soil characteristics, including pH and acidity (FAO 2008; Mooso *et al.* 2013). Andreoli *et al.* (2007) noted that between 5% and 30% of the total phosphorus applied through chemical fertilizer is used by plants. This fact can result in frequent fertilization (P being the most commonly applied nutrient through chemical fertilizers in many places) leading to increased costs of agricultural management.

Sewage sludge may also contain a wide spectrum of harmful toxic substances such as heavy metals, polycyclic aromatic hydrocarbons (PAHs), polychlorinated dibenzo-p-dioxins and dibenzo-p-furans (PCDD/Fs), polychlorinated biphenyls (PCBs), di(2-ethylhexyl) phthalate (DEHP), polybrominated diphenyl ethers (PBDEs), detergent residues, pharmaceuticals, personal care products, endogenous hormones, synthetic steroids and others (Singh & Agrawal 2008; Smith 2009; Fijalkowski *et al.* 2017)

The release of heavy metals contained in sewage sludge and their adsorption and speciation in soil are influenced by soil pH, cation exchange capacity, organic matter content, mobility and form of specific metals (Silveira *et al.* 2003; Singh & Agrawal 2008).

Low doses of sewage sludge did not cause a significant increase in heavy metal concentrations (Singh & Agrawal 2008) and low metal sludge has beneficial effects on microbial biomass, organic carbon and on soil microbial activity (Usman *et al.* 2012). Excessive application of sewage sludge to soil has been found to increase the bioavailability of heavy metals (Singh & Agrawal 2008) that have a negative effect on soil (Usman *et al.* 2012).

The concentrations of organic contaminants in sewage sludge are often correlated with their concentrations in influent wastewater. They are also affected by the sludge characteristics (pH, organic matter, cation concentration), physicochemical properties of the compounds (molecular weight, hydrophobicity, water solubility, acid dissociation constant,

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resistance to biodegradation) and the operational parameters of the WWTP (presence or absence of primary sedimentation, hydraulic residence time in different tanks, sludge residence time in bioreactors, methods of sludge stabilization) (Fijalkowski *et al.* 2017).

The aim of this study is to compare the present restrictions set by legislation dealing with the quality of sewage sludge in EU countries with regard to further sludge utilization in agriculture.

Restrictions given by the content of pollutants in sewage sludge

The application of sludge to soil poses a threat by the potential long-term accumulation of toxic elements in soil (Singh & Agrawal 2008) and their uptake by crop plants.

However, currently the land application of sewage sludge is regulated only by the concentration of heavy metals specified in Council Directive 86/278/EEC (CEC 1986), which is outdated and does not reflect current needs of ensuring the safety of sludge application in agriculture. The directive should support the safe use of sewage sludge in agriculture and regulate its use in order to prevent harmful effects on soil, vegetation, animals and humans.

Among other things, the Directive defined the rules for sampling and analysis of sludge and soil; it also sets the limits for concentrations of heavy metals in sludge-treated soil. All EU Member States have transposed the limits of European Council Directive 86/278/EEC (CEC 1986) into their own legislation (Table 1).

Most of the EU countries presented in Table 1 adopted even more stringent limits for sludge use in agriculture by setting lower limits for heavy metals in comparison with European Directive 86/278/EEC (CEC 1986). In the case of cadmium, it is 18 out of 27 countries. In the case of copper, it is 14 out of 27 countries. In the case of mercury, it is 19 out of 27 countries. In the case of nickel, it is 16 out of 27 countries. In the case of lead, it is 14 out of 27 countries. In the case of zinc, it is 10 out of 27 countries. It is obvious that lower limits have been adopted by the EU countries mainly for the most toxic heavy metals – mercury and cadmium (in 67% of the 27 analysed EU countries). On the contrary, for zinc the limit values recommended by the Directive have been adopted (in 33% of the 27 analysed EU countries). We also analysed for how many heavy metals of the Directive group (6 metals – Cd, Cu, Hg, Ni, Pb, Zn) each of the EU member countries had adopted lower limit values. There are two main groups of countries: the first is the group of countries (9 countries out of 27) adopting lower limit values for all heavy metals, the second is the group of countries (7 countries out of 27) adopting the limit values of the Directive for all heavy metals. The third largest group (4 countries out of 27) adopted lower limit values for 5 heavy metals. It seems that there were two approaches there – adopting the Directive recommendations or taking a stricter approach, in some countries resulting in a very low ratio of sewage sludge being used in agriculture (Table 6, Figure 1). There is no difference between old EU member countries and new EU member countries (entering the EU after 2004).

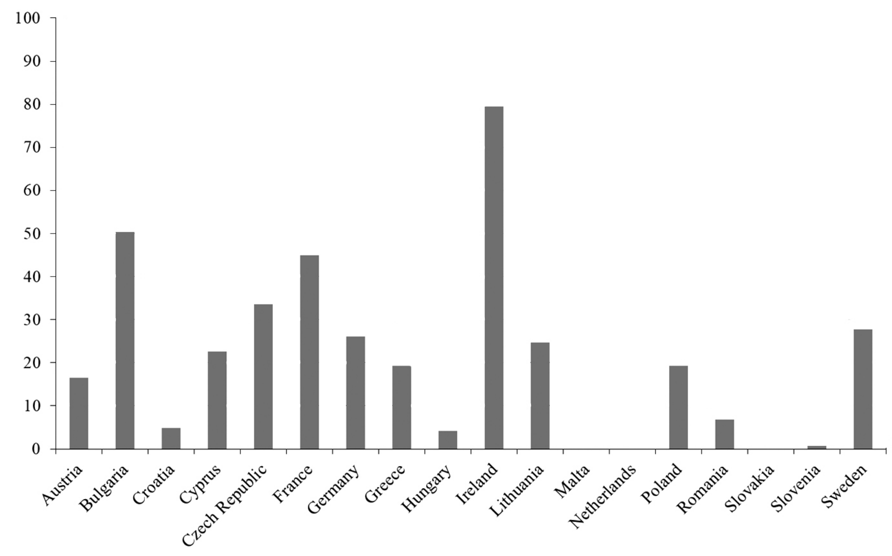

Figure 1. Percentages of total sludge used in agriculture in selected 18 EU countries in 2014

https://doi.org/10.17221/36/2018-SWR

Table 1. Limits of Cd, Cu, Hg, Ni, Pb and Zn for sludge use in agriculture (mg/kg DM of sewage sludge)

State	Cd	Cu	Hg	Ni	Pb	Zn
Directive 86/278/EEC	20–40	1000–1750	16–25	300–400	750–1200	2500–4000
Austria (Burgenland)						
Quality class I[a]	2	300	2	60	100	1000
Quality class II[a]	10	500	10	100	500	2000
Austria (Carinthia)[b]						
Class I[b]	0.7	70	0.4	25	45	200
Class A[b]	1	150	0.7	60	150	500
Class AB[b]	2	300	2	60	150	1200
Class B[b]	2.5	300	2.5	80	150	1800
Austria (Lower Austria)						
Quality class I[c]						
Quality class II	2	300	2	60	100	1500
Austria (Salzburg)	application of sewage sludge and its mixtures is prohibited[d]					
Austria (Steiermark)	2	300	2	60	100	1200
Austria (Tyrol)	application of sewage sludge and products with their content on farmland is prohibited					
Austria (Upper Austria)	5	400	7	80	400	1600
Austria (Vienna)	application of sewage sludge is prohibited[e]					
Austria (Voralberg)	4	500	4	100	150	1800
Belgium	10	600	10	100	500	2000
Bulgaria	30	1600	16	350	800	3000
Croatia	5	600	5	80	500	2000
Cyprus	20–40	1000–1750	16–25	300–400	750–1200	2500–4000
Czech Republic	5	500	4	100	200	2500
Denmark	0.8	1000	0.8	30	120	4000
Estonia	20	1200	20	400	900	3500
Finland	1.5	600[f]	1	100	100	1500[f]
France	20	1000	10	200	800	3000
Germany[g]	10	800	8	200	900	2500
Greece	20–40	1000–1750	16–25	300–400	750–1200	2500–4000
Hungary	10	1000	10	200	750	2500
Ireland	20	1000	16	300	750	2500
Italy	20	1000	10	300	750	2500
Latvia	10	800	10	200	500	2500
Lithuania	6.0	600	6.0	300	500	2000
Luxembourg	2.5	700	1.6	80	200	3000
Malta	5	800	5	200	500	2000
Netherlands	1.25	75	0.75	30	100	300
Poland	20	1000	16	300	750	2500
Portugal	20	1000	16	300	750	2500
Romania	10	500	5	100	300	2000
Slovakia	10	1000	10	300	750	2500
Slovenia	1.5	300	1.5	75	250	1200

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Table 1 to be continued

State	Cd	Cu	Hg	Ni	Pb	Zn
Directive 86/278/EEC	20–40	1000–1750	16–25	300–400	750–1200	2500–4000
Spain						
pH of soil < 7	20	1000	16	300	750	2500
pH of soil > 7	40	1750	25	400	1200	4000
Sweden	2	600	2.5	50	100	800
United Kingdom	use of sewage sludge limited by the maximum allowable concentrations of Cd, Cu, Hg, Ni, Pb and Zn in soil intended for sludge application					

[a]The limits are set with regard to the application rate of the maximum allowable annual pollutant load: Class I from 4.17 t DM/ha·year (at limited concentrations of Cu or Ni in sludge) to 12.5 t DM/ha·year (at limited concentrations of Cr, Pb, Cd and Hg in sludge), Class II 2.5 t of DM/ha·year

[b]Division into classes according to limits and application rates of the maximum allowable pollutant load per 2 years: Class I 10 t DM/2 years, Class A 8 t DM/2 years, Class AB 6 t DM/2 years, Class B 4.8 t DM/2 years; exceeding the limit values for one parameter at a maximum of 25% is permissible; if exceeding is induced by geogenic loads, in exceptional cases the controller may mark the application as admissible

[c]Sludge of Class I must not exceed the regional average value in DM for these indicators: Zn, Cu, Cr, Pb, Cd, Hg and AOX in the upper soil layer (for arable soil to a depth of 25 cm, for pastures to a depth of 10 cm); at the same time they must not exceed the value of Class II for these indicators

[d]This restriction is not applicable to agricultural wastewater based mixtures, three-month matured sanitized sludge from septic tanks and sludge of the wastewater treatment plants of structures that are located in extreme positions if there is no sludge application restriction; compost with an admixture of sludge can be used only on soil that is not intended for food production

[e]Only hygienically safe products containing treated sludge marketed as fertilizers, compost and soil can be applied

[f]For plants it is required that the load of Cu and Zn will not be more than doubled if the lack of nutrients is in the soil, and fertilizers are applied to them; sewage sludge can be applied as a fertilizer, i.e. treated sewage sludge, sludge from septic tanks or sludge mixtures; however, it must not exceed the maximum concentration in the soil listed in Table 5

[g]In the case of soil that is classified as light soil with a clay content below 5% at 5 < pH < 6, the maximum allowable concentrations of Cd and Zn in sludge are Cd 5 mg/kg DM and Zn 2000 mg/kg DM

In the group with the lower limit values, there are 5 old countries and 4 new countries. In the group with limit values given by the Directive, there are 4 old countries and 3 new countries.

The United Kingdom was not included in the assessment, because it took a different approach to setting limit values. Use of sewage sludge is limited by the maximum allowable concentrations of Cd, Cu, Hg, Ni, Pb and Zn in soil intended for sludge application.

In addition to the limit values for heavy metals listed in Directive 86/278/EEC (CEC 1986), several countries introduced limit values for other elements (Table 2) – chromium (22 countries), arsenic (7 countries), molybdenum (2 countries), cobalt and selenium (Hungary for both metals), as well as for other categories of pollutants usually detected in the sludge as organic micropollutants (Table 3) and pathogens (Table 4).

Except for Hungary which set a limit value of 1 mg per kg dry matter (DM) for hexavalent chromium (Cr^{VI}) (Table 2), EU countries have not set any limit values for heavy metal ions which are more reactive and more toxic to plants.

Proposed limit values for certain synthetic organic compounds (Table 3) such as halogenated organic compounds, linear alkylbenzenesulphonates (LAS), phthalates, nonylphenol, PAHs, PCBs and PCDD/Fs were included in the third draft of the Working Document on Sludge (European Commission 2000a) and national legislation of EU countries. Due to the fact that the treated sludge can contain significant amounts of pathogens, depending on the treatment procedure used, limits for bacteria as indicators of faecal contamination were proposed (Table 4) in the third draft of the Working Document on Sludge (European Commission 2000a). Compliance with these indicators is necessary to eliminate the negative effects of sludge due to the presence of contaminants, in particular the concentration of heavy metals Zn, Cu, Pb, Hg, hazardous organic micropollutants and pathogenic microorganisms.

https://doi.org/10.17221/36/2018-SWR

Table 2. Limits of Cr, As, Co, Mo, and Se for sludge use in agriculture (mg/kg DM of sewage sludge)

State	Cr	As	Co	Mo	Se	State	Cr	As	Co	Mo	Se
Austria (Burgenland)						Estonia	1200				
Quality class I[a]	100					Finland	300				
Quality class II[a]	500					France	1000				
Austria (Carinthia)[b]						Germany	900				
Class I[b]	70					Greece					
Class A[b]	70					Hungary	1000 (1 mg/kg Cr^{VI})	75	20	50	100
Class AB[b]	70					Ireland					
Class B[b]	100					Italy					
Austria (Lower Austria)						Latvia	600				
Quality class I[c]						Lithuania	400				
Quality class II						Luxembourg	100				
Austria (Salzburg)	application of sewage sludge and its mixtures is prohibited[d]					Malta	800				
Austria (Steiermark)	70					Netherlands	75	15			
Austria (Tyrol)	application of sewage sludge and products with their content on farmland is prohibited					Poland	500				
Austria (Upper Austria)	400					Portugal	1000				
Austria (Vienna)	application of sewage sludge is prohibited[e]					Romania	500	10		50	
Austria (Voralberg)	300					Slovakia	1000	20			
Belgium	500					Slovenia	200				
Bulgaria	500	25				Spain					
Croatia	500					pH of soil < 7	1000				
Cyprus						pH of soil > 7	1500				
Czech Republic	200	30				Sweden	100				
Denmark	100	25				United Kingdom					

[a]The limits are set with regard to the application rate of the maximum allowable annual pollutant load: Class I from 4.17 t DM/ha·year (at limited concentrations of Cu or Ni in sludge) to 12.5 t DM/ha·year (at limited concentrations of Cr, Pb, Cd and Hg in sludge), Class II 2.5 t of DM/ha·year

[b]Division into classes according to limits and application rates of the maximum allowable pollutant load per 2 years: Class I 10 t DM/2 year, Class A 8 t DM/2 year, Class AB 6 t DM/2 year, Class B 4.8 t DM/2 year; exceeding the limit values for one parameter at a maximum of 25% is permissible; if exceeding is induced by geogenic loads, in exceptional cases the controller may mark the application as admissible

[c]Sludge of class I must not exceed the regional average value in the DM for these indicators: Zn, Cu, Cr, Pb, Cd, Hg and AOX in the upper soil layer (for arable soil to a depth of 25 cm, for pastures to a depth of 10 cm); at the same time they must not exceed the value of Class II for these indicators

[d]This restriction is not applicable to agricultural wastewater based mixtures, three-month matured sanitized sludge from septic tanks and sludge of the wastewater treatment plants of structures that are located in extreme positions if there is no sludge application restriction; compost with an admixture of sludge can be used only on soil that is not intended for food production

[e]Only hygienically safe products containing the treated sludge marketed as fertilizers, compost and soil can be applied

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

State	AOX	DEHP	LAS	NP/NPE	PAH	PCB	PCDD/F (ng TEQ/kg DM)	$C_5 – C_{40}$
EC (2000)[a]	500	100	2600	50	6[f]	0.8[j]	100	
Austria (Carinthia)[b]	500				6[f]	1	50	
Austria (Lower Austria)								
Quality class I[c]								
Quality class II	500							
Austria Steiermark)[d]	500				6[g]			
Austria (Voralberg)						0.2[k]	100	
Austria (Upper Austria)	500							
Belgium						0.8[j]		
Bulgaria					6,5	1		
Croatia						0.2[l]	100	
Czech Republic	500				10[h]	0.6[j]		
Denmark		50	1300	10	3[f]			
France					fluoranthene 5, benzo(b)fluoranthene 2.5, benzo(a)pyrene 2	0.8[j]		
Germany	500					0.2[m]	100	
Hungary					10[g]	1[j]		4000
Luxembourg					20[g]	0.2[k]	20	
Portugal			5000	450	6	0.8	100	
Romania	500				5[i]	0.8[j]		
Sweden[e]				50	3[f]	0.4[j]		

AOX – absorbable organic halogen; DEHP – di(2-ethylhexyl)phthalate; LASs – linear alkylbenzene sulphonates; NP/NPEs – nonylphenols and nonylphenol ethoxylates; PAHs – polycyclic aromatic hydrocarbons; PCBs – polychlorinated biphenyls; PCDD/Fs – polychlorinated dibenzo-p-dioxins and dibenzo-p-furans

[a]Proposed but withdrawn (European Commission 2000a)

[b]Applies to all classes

[c]Sludge of class I must not exceed the regional average value in DM for these indicators – Zn, Cu, Cr, Pb, Cd, Hg and AOX in the upper soil layer (for arable soil to a depth of 25 cm, for pastures to a depth of 10 cm); at the same time it must not exceed the value of class II for these indicators

[d]These limits apply to sewage sludge that comes from WWTPs for more than 30 000 PE

[e]The limits set in the context of a voluntary agreement on quality assurance between the Swedish Environmental Protection Agency – Naturårdsvertket and the Federation of Swedish Farmers (LRF) and the Swedish Water and Waste Water Association (VAV)

[f]Sum of acenaphthene, fluorene, phenanthrene, fluoranthene, pyrene, benzo(b+j+k)fluoranthene, benzo(a)pyrene, benzo(ghi) perylene, indeno(1,2,3-c,d)pyrene

[g]Sum of 16 US EPA PAU (naphthalene, acenaphthylene, acenaphthene, fluorene, fenanthrene, anthracene, fluoranthene, pyrene, benz(a)anthracene, chrysene, benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(a)pyrene, dibenzo(a,h)anthracene, indeno(1,2,3-c,d)pyrene and benzo(ghi)perylene)

[h]Sum of anthracene, benzo(a)anthracene, benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(a)pyrene, benzo(ghi)perylene, fenanthrene, fluoranthene, chrysene, indeno(1,2,3-cd)pyrene, naphthalene, pyrene

[i]Sum of anthracene, benzoanthracene, benzofluoranthene, benzoperylene, benzopyrene, chrysene, fluoranthene, indeno(1,2,3) pyrene, naphthalene, fenanthrene, pyrene

[j]Sum of 7 congeners: PCB 28, 52, 101, 118, 138, 153, 180

[k]Sum of 6 congeners: PCB 28, 52, 101, 138, 153, 180

[l]For each of these congeners: PCB 28, 52, 101, 141, 180

[m]For each congener

https://doi.org/10.17221/36/2018-SWR

Table 4. Standards for maximum concentrations of pathogens in sewage sludge

State	*Salmonella* sp.	Other pathogens
EC (2000)[a]	no occurrence in 50 g	*Escherichia coli* < 500 cfu/g
Bulgaria	no occurrence in 20 g	*Escherichia coli* < 100 MPN/g *Clostridium perfringens* < 300 MPN/g Helminths eggs and larvae, 1unit/kg DM
Czech Republic[b]	no occurrence in 50 g	*Escherichia coli* or Enterococci < 10^3 cfu/g (4 samples from 5) < 5.10^3 cfu/g (1 sample from 5)
Sludge category I[b, c]	no occurrence in 1 g of DM	Thermotolerant coliforms < 10^3 cfu/g DM Enterococci < 10^3 cfu/g DM
Sludge category II[b, c]	not determined	Thermotolerant coliforms 10^3–10^6 cfu/g DM Enterococci 10^3–10^6 cfu/g DM
Denmark[d]	no occurrence	Faecal streptococci < 100/g
Finland	no occurrence in 25 g	*Escherichia coli* < 1000 cfu, < 100 cfu in greenhouse cultivation, where the consumed part is in contact with the substrate
France	8 MPN/10 g DM	Enterovirus < 3 MPCN/10 g DM Helminths eggs < 3/10 g DM
Italy	1000 MPN/g DM	
Lithuania		*Escherichia coli* ≤ 1000 cfu/g *Clostridium perfringens* ≤ 100 000 cfu/g Helminths eggs and larvae, 0 units/kg Enterobacteria, 0 cfu/g
Luxembourg		Enterococci – 100/g Helminths eggs cannot be contagious.
Poland	no occurrence in 100 g	
Portugal	no occurrence in 50 g	Escherichia coli < 1000 cfu/g
Austria (Carinthia)[e]	no occurrence in 1 g	Enterococci < 10^3/g no Helminths eggs
Austria (Lower Austria)	no occurrence in 1 g	*Escherichia coli* < 100 cfu no Helminths eggs
Austria (Steiermark)	no occurrence in 1 g	Enterococci < 10^3/g
Slovakia		Thermotolerant coliforms < 2×10^6 cfu/g DM Faecal streptococci < 2×10^6 cfu/g DM

[a]Proposed but withdrawn (European Commission 2000a)
[b]The number of tested samples is 5
[c]These criteria can be used in the interim period until December 31, 2019
Sludge category I – sludge which can be generally applied to land used for agriculture respecting the other provisions of the Decree
Sludge category II – sludge which can be applied to agricultural land used for industrial crop cultivation or land used for conventional crop cultivation in the autumn; land where sludge category II was applied cannot be used for growing field vegetables, potatoes and intensive orchard cultivation for at least three years after the application of sewage sludge
[d]For treated sludge only
[e]Applies to all classes
DM – dry matter; CFU – colony forming unit; MPN – most probable number; MPCN – most probable cytopathic number

Limits of Cd, Cr, Cu, Hg, Ni, Pb and Zn in sludge-treated soil (mg/kg dry soil) in EU member states are given in Table 5. Valid legislation relating to the use of sewage sludge in agriculture in EU countries is presented in Table S1 in the Electronic Supplementary Material (ESM).

Sewage sludge production and processing

For sludge processing, the size category of the WWTP and particularly purification technology play important roles. In small WWTPs, municipal wastewater usually outweighs industrial effluent, if any industrial wastewa-

https://doi.org/10.17221/36/2018-SWR

Table 5. Limits of Cd, Cr, Cu, Hg, Ni, Pb and Zn in sludge-treated soil (mg/kg dry soil)

State	Cd	Cr	Cu	Hg	Ni	Pb	Zn
Directive 86/278/EEC 6 < pH < 7	1–3		50–140	1–1.5	30–75	50–300	150–300
Austria (Burgenland)	2	100	100	1.5	60	100	300
Austria (Carinthia)							
5 < pH < 5.5	0.5	50	40	0.2	30	50	100
5.5 < pH < 6.5	1	75	50	0.5	50	70	150
pH > 6.5	1.5	100	100	1	70	100	200
Austria (Lower Austria)	1.5/1[a]	100	60	1	50	100	200
Austria (Salzburg)	application of sewage sludge and its mixtures is prohibited[b]						
Austria (Steiermark)	0.5	100	60	0.5	60	100	150
Austria (Tyrol)	application of sewage sludge and products with its content to farmland is prohibited						
Austria (Upper Austria)	0.5	100	60	0.5	60	100	150
Austria (Vienna)	application of sewage sludge is prohibited[c]						
Austria (Voralberg)	2	100	100	1	60	100	300
Belgium	1.5		50	1	20	50	200
Bulgaria							
6 < pH < 7.4	2	200	140	1	[d]	100	250
pH > 7.4	3	200	200	1	110	120	300
Croatia							
5 < pH < 5.5	0.5	50	40	0.2	30	50	100
5.5 < pH < 6.5	1	75	50	0.5	50	70	150
pH > 6.5	1.5	100	100	1	70	100	200
Cyprus, 6 < pH < 7	1–3	–	50–140	1–1.5	30–75	50–300	150–300
Czech Republic							
Common soil[e]	0.5	90	60	0.3	50	60	120
Light soil [f]	0.4	55	45	0.3	45	55	105
Denmark	0.5	30	40	0.5	15	40	100
Estonia	3	100	50	1.5	50	100	300
Finland	0.5	200	100	0.2	60	60	150
France	2	150	100	1	50	100	300
Germany[g]	1.5	100	60	1	50	100	200
Greece, 6 < pH < 7	1–3	–	50–140	1–1.5	30–75	50–300	150–300
Hungary	1	75 (1 mg/kg Cr^{VI})	75	0.5	40	100	200
Italy	1.5	–	100	1	75	100	300
Ireland, 5 < pH < 7[h]	1	–	50	1	30	50	150
Latvia							
5 < pH < 6, sand, sandy loam	0.5	40	15	0.1	15	20	50
5 < pH < 6, loamy soil, loam	0.6	50	25	0.2	25	25	65
6 < pH < 7, sand, sandy loam	0.6	60	35	0.25	35	25	70
6 < pH < 7, loamy soil, loam	0.7	70	50	0.35	50	30	80
pH > 7, sand, sandy loam	0.8	80	55	0.4	60	35	90
pH > 7, loamy soil, loam	0.9	90	70	0.5	70	40	100
Lithuania							
Sand, sandy loam	0.8	40	40	0.5	35	40	120
Clay, clay loam	1.1	60	60	0.8	45	60	200

https://doi.org/10.17221/36/2018-SWR

Table 5 to be continued

State	Cd	Cr	Cu	Hg	Ni	Pb	Zn
Luxembourg	1–3	100–200	50–140	1–1.5	30–75	50–300	150–300
Malta							
5 < pH < 6	0.5	30	20	0.1	15	70	60
6 < pH < 7	1	60	50	0.5	50	70	150
pH > 7	1.5	100	100	1	70	100	200
Netherlands	0.8	100	36	0.3	35	85	140
Poland							
Light soil	1	50	25	0.8	20	40	80
Medium soil	2	75	50	1.2	35	60	120
Heavy soil	3	100	75	1.5	50	80	180
Portugal							
pH < 5.5	1	50	50	1	30	50	150
5.5 < pH < 7	3	200	100	1.5	75	300	300
pH > 7	4	300	200	2	110	450	450
Romania, pH > 6.5	3	100	100	1	50	50	300
Slovakia	1	60	50	0.5	50	70	150
Slovenia	1	100	60	0.8	50	85	200
Spain							
pH < 7	1	100	50	1	30	50	150
pH > 7	3	150	210	1.5	112	300	450
Sweden	0.4	60	40	0.3	30	40	100
United Kingdom							
5 < pH < 5.5	3	–	80	1	50	300	200
5.5 < pH < 6.0	3	–	100	1	60	300	250
6 < pH < 7	3	–	135	1	75	300	300
pH > 7	3	–	200	1	110	300	450

[a]At pH < 6
[b]This restriction is not applicable to agricultural wastewater based mixtures, three-month matured sanitized sludge from septic tanks and sludge of the wastewater treatment plants of structures that are located in extreme positions if there is no sludge application restriction; compost with an admixture of sludge can be used only on soil that is not intended for food production
[c]Only hygienically safe products containing the treated sludge marketed as fertilizers, compost and soil can be applied
[d]pH 6–7 = 75 mg/kg; pH 7–7.4 = 80 mg/kg
[e]Common soil – sandy loam, loam, clay loam and clay soil, which make up the bulk of agricultural land; this is a soil with normal variability of elements, with normal soil development in different geomorphological conditions, in this concept including the soil on carbonate rocks
[f]Light soil – soil formed on very light and poor parent rocks such as sand and gravel; defining these types of soil begins with the presence of fine particles (0.01 mm), forming a maximum of 20%; this soil is characterized by a very low absorption capacity
[g]In the case of soil that is classified as light soil with a clay content below 5% at 5 < pH < 6, the maximum allowable concentrations of Cd and Zn in sludge are Cd – 1 mg/kg DM and Zn – 150 mg/kg DM
[h]If the soil pH is consistently higher than 7, the given values must not exceed more than 50%, assuming that there is no threat to human health, environment and particularly to groundwater

ter ever occurs. Therefore, heavy metal content in the sludge of the given WWTP tends to be lower than in a larger urban WWTP, however, the potential danger of single contamination that is not offset by dilution cannot be overlooked.

On the basis of Council Directive 91/271/EEC (CEC 1991) municipalities with more than 2000 PE should have mechanical-biological (two-stage) WWTPs. The Directive emphasizes the reuse of produced sludge and prohibits the disposal of sludge in surface water

https://doi.org/10.17221/36/2018-SWR

as of Dec 31, 1998. As stated by the Commission Decision 2001/118/EC (CEC 2001) sludge from urban wastewater treatment is considered non-hazardous waste. In accordance with the Directive of the European Parliament and Council Directive 2008/98/EC (EPOC 2008) on waste, the following hierarchy of waste management is requested: (a) prevention, (b) preparing for reuse, (c) recycling, (d) other recovery, e.g. energy recovery, (e) disposal.

The use of sewage sludge in agriculture is regulated by Council Directive 86/278/EEC (CEC 1986). Sludge must be used with regard to the nutritional needs of plants and its use must not impair the quality of the soil or surface water and groundwater. Member States must regulate the use of sludge in such a way as to avoid exceeding the limit values specified in Table 5 by the accumulation of heavy metals in soil. The required sludge application rates also depend on the sludge fertilizer potential and properties of soil.

A wide range of sludge treatment technologies is used in EU countries. The most common stabilization methods seem to be anaerobic (24 EU countries) and aerobic digestion (20 EU countries). Mechanical sludge dewatering is preferred compared to the use of drying beds. Regarding the EU-15 countries (old Member States), in particular Germany, Italy, France and the UK mainly apply thermal drying (Kelessidis & Stasinakis 2012).

The most common method of final sludge disposal in the EU-15 in 2014 and 2015 was incineration (47.3%, 61.5%), followed by sludge reuse including direct agricultural application and composting (48.2%, 38.2%). The national legislation of some Member States has set very strict limits for organic matter or total organic carbon (TOC) contained in sludge (Austria, Germany, Netherlands), practically prohibiting sludge landfilling (Kelessidis & Stasinakis 2012). In some of the EU-13 countries (new Member States that joined the EU after 2004) the most frequently used disposal method was still landfilling in 2014 and 2015 (Table 6): in Malta (100% in both years), Croatia (94.5%, 94.7%) and Romania (75.5%, 66.9%).

Total sludge production and disposal in the EU and division by the method of sludge disposal in 2014 and 2015 according to Eurostat (Eurostat 2016), and in the Czech Republic according to the Czech Statistical Office (CZSO 2016) are shown in Table 6.

Only a few countries have allowed the use of untreated sludge under certain authorized conditions (e.g. France, Sweden and Estonia) and some have to meet given obligations for treatment such as biological stabilization, lime stabilization, chemical treatment, heat treatment, composting, pasteurization or digestion before use (Austria, Denmark, Finland and Poland). In several countries the use of sludge in forests (Austria, Belgium, Germany and Netherlands) and green areas has also been prohibited (Kelessidis & Stasinakis 2012).

Percentages of total sludge used in agriculture in 18 EU countries, reported for the year 2014, are shown in Figure 1. Other EU countries did not publish those values for that year. Only in Ireland more than 70% of total sludge was disposed in agriculture, in 7 countries it was between 70% and 20% (Bulgaria over 50%), in 10 countries less than 20% (Slovenia 0.7%, Netherlands, Slovakia and Malta 0%). Malta, Netherlands and Slovenia belong to the group of countries with the most strict limit values for the basic group of heavy metals in sludge (Table 1). For the year 2015, only 14 EU countries reported values of the amount of sludge disposal. Ireland used 80% of total sludge disposal, 5 countries used between 70% and 20%. Slovakia, Slovenia and Malta used 0%.

For several years the European Union made efforts to re-unify the approach to sludge treatment, culminating with the document of the Joint Research Centre (JRC) "End-of-waste criteria for biodegradable waste subjected to biological treatment (compost & digestate): Technical Proposals", where sludge was excluded from organic waste admitted for producing an "end-of-waste" compost (Saveyn & Eder 2014; Mininni *et al.* 2015).

Although sludge minimization, full stabilization and hygienization by thermal hydrolysis processes before anaerobic digestion, on-site incineration in a fluidized bed furnace and other methods have come to the forefront of interest, classifying sewage sludge fulfilling strict output quality criteria as potential input material for producing "end-of-waste" compost has been proposed. This decision was also based on the evidence that many Member States already produce large quantities of compost with sludge, i.e. Estonia, Finland, France, Germany, Italy, Lithuania and Spain. The largest sludge producers in Europe are Spain, Germany, Great Britain, France and Italy (Table 6). In Italy (the data were not published by Eurostat – see Table 6), total compost production is currently estimated at 1.0 Mt/year + 0.3 Mt/year of compost with sludge. Approximately 450 000 t per year of dewatered sludge (10–15% of total sludge production) are composted (Mininni *et al.* 2015).

https://doi.org/10.17221/36/2018-SWR

Table 6. Total sludge production and disposal in the EU and division by the method of sludge disposal in 2014 and 2015 according to Eurostat (Eurostat 2016), in the Czech Republic according to the Czech Statistical Office (CZSO 2016); values are given in thousand tonnes of DM

State	Total sludge production		Total sludge disposal		Method of sludge disposal									
					agricultural use		composting and other applications		landfilling		incineration		other	
	2014	2015	2014	2015	2014	2015	2014	2015	2014	2015	2014	2015	2014	2015
Austria	239.0	–	239.0	–	39.6	–	77.7	–	3.2	–	118.5	–	0	–
Belgium	–	–	–	–	–	–	–	–	–	–	–	–	–	–
Bulgaria	54.9	57.4	32.6	47.2	16.4	30.4	0.8	3.4	8.5	8.5	0	0	7.0	4.8
Croatia	16.3[b]	17.9	16.3	17.1	0.8	1.0	0.008	0	15.4	16.2	0	0	0.083	0.001
Cyprus	6.2	6.7	6.2	6.7	1.4	0.9	0	0	0	0	0	0	4.8	5.8
Czech Republic	159.2	173.0	159.2	173.0	47.8	63.1	60.5	67.1	5.2	6.5	3.4	2.2	42.2	34.2
Denmark	–	–	–	–	–	–	–	–	–	–	–	–	–	–
Estonia	–	–	–	–	–	–	–	–	–	–	–	–	–	–
Finland	–	–	–	–	–	–	–	–	–	–	–	–	–	–
France	961.5	–	937.1	–	421.3	–	305.1	–	31.1	–	170.6	–	8.9	–
Germany	1837.1	1820.6	1803.0	1803.0	470.9	427.7	251.5	223.7	0	0	1077.9	1148.7	2.6	3.0
Greece	116.1	–	116.1	–	22.8	–	9.0	–	39.0	–	38.5	–	6.8	–
Hungary	166.5	156.9	112.4	111.0	4.7	9.4	74.7	83.7	5.3	5.1	22.8	12.7	0	0
Ireland	53.5	58.4	53.5	58.4	42.5	46.7	9.3	10.9	0.4	0.1	0	0	1.4	0.7
Italy	–	–	–	–	–	–	–	–	–	–	–	–	–	–
Latvia	–	–	–	–	–	–	–	–	–	–	–	–	–	–
Lithuania	40.7	42.9	34.4	37.3	8.5	11.2	14.6	15.6	0	0	0	0	11.2	10.4
Luxembourg	–	9.2	–	9.2	–	3.1	–	2.2	–	–	–	0.8	–	3.0
Malta	8.5	8.4	8.5	8.4	0	0	0	0	8.5	8.4	0	0	0	0
Netherlands	344.2	–	319.7	–	0	–	0	–	0	–	319.7	–	0	–
Poland	556.0	568.0	556.0	568.0	107.2	107.5	46.3	47.1	31.5	40.5	84.2	79.3	286.7	293.6
Portugal	–	–	–	–	–	–	–	–	–	–	–	–	–	–
Romania	192.3	210.5	192.3	155.8	13.1	10.6	0.2	–	145.1	104.2	1.2	0.5	32.7	40.9
Slovenia	28.3	29.1	28.0	29.0	0.2	0	1.5	0.6	0.3	0.2	15.0	15.1	11.0	13.0
Slovakia	56.9	56.2	56.9	56.2	0	0	26.1	24.9	4.3	4.6	16.0	16.9	10.5	9.8
Spain	–	–	–	–	–	–	–	–	–	–	–	–	–	–
Sweden	200.5	197.5	183.9	–	51.0	59.5	59.1	–	3.6	–	2.2	–	68.1	–
United Kingdom	–	–	–	–	–	–	–	–	–	–	–	–	–	–

– values are not available; [b]break in time series

https://doi.org/10.17221/36/2018-SWR

Based on Eurostat data, Germany and France are the largest producers of compost containing sludge (Table 6). Compost containing sludge consists of one to two-thirds of sewage sludge; other inputs are green waste and biowaste (Saveyn & Eder 2014).

According to the European Federation for Agricultural Recycling (EFAR), about 25% of sewage sludge production is co-composted with green waste in France. The NF U44-095 standard applicable since May 2002 has defined product criteria for sewage sludge, allowing the development of a well-structured market for this type of soil improver (Saveyn & Eder 2014).

Despite the fact that Germany is one of the largest producers of sludge and creates large quantities of compost containing sludge, a new Decree on sewage sludge was passed by the German government on January 18, 2017. The new Decree significantly reduces the conventional land use associated with sewage sludge to further reduce pollutant inputs into the soil. After the transition periods (12 and 15 years), the application of sludge to agricultural land is possible only from WWTPs up to 50 000 PE and under stricter conditions (BMUB 2017; Stránský 2017). The main aim of the draft Decree is to return valuable components of sewage sludge (such as phosphorus) to the economic cycle, more comprehensively than before. Within 12 years after the commencement of the Decree all German WWTPs up to 100 000 PE must recover phosphorus from sewage sludge and within 15 years after the commencement of the Decree, also WWTPs up to 50 000 PE. An exception to this is sewage sludge with a phosphorus content of less than 2%. The regulation does not impose any specific technology for the recovery of phosphorus, but leaves plenty of room for using or developing innovative recovery methods (BMUB 2017; Stránský 2017).

Similarly, Austria published the draft of the "Federal Waste Management Plan 2017" on January 11, 2017 and launched the process of public participation (BMLFUW 2017; Stránský 2017).

In terms of strategy for the future management of sewage sludge, which is part of the plan, significantly reduced agricultural use of sewage sludge is proposed. During a transition period of 10 years the direct application of sludge to soil and composting of sewage sludge from WWTPs with a designed capacity greater than or equal to 20 000 PE should be discontinued (BMLFUW 2017; Stránský 2017).

The strategy in Austria also includes the obligation to recycle phosphorus from sewage sludge from WWTPs with a designed capacity greater than or equal to 20 000 PE, which should be achieved on the spot (WWTPs over 50 000 PE), if the phosphorus content is higher than 2% or by sewage sludge monocombustion and subsequent recovery of phosphorus from the ashes of incinerated sludge (BMLFUW 2017; Stránský 2017).

Toxicity tests as a quick verification tool of sludge applicability

Sewage sludge can be contaminated with a wide range of xenobiotics and other pollutants resulting in a financially expensive analysis. It is often difficult to assess the impact of such pollutants on the environment and organisms. Methods of complex assessment of possible negative effects are being sought.

One group of methods is toxicity tests that allow assessing the complex effect of the studied matrix on the target organisms. In the case of small sources of sludge, where commercial use of sludge is not expected, the main concern of WWTP operators and farmers is to reduce the contamination of the resulting sludge and to ensure that it does not represent a health hazard to the environment in terms of toxicity. The potential toxicity can be assessed for example by phytotoxicity tests and the earthworm avoidance test (Moreira *et al.* 2008).

Phytotoxicity tests exist in the form of Directives issued by major environmental agencies such as United States Environmental Protection Agency (US EPA), Organisation for Economic Co-operation and Development (OECD), International Standards Organization (ISO), American Society for Testing and Materials (ASTM) and others. A summary of phytotoxicity tests is given in Table 7.

Phytotoxic data provide information about the impact of various hazardous substances including xenobiotics on ecosystems and allow assessing the effect of interaction between external and internal factors and sensitivity of individual plant species. They are also necessary for the criteria formation of the adopted regulatory measures and, ultimately, to evaluate the effectiveness of bioremediation technologies. The advantage of phytotoxicity tests is the ability to provide an estimation of the short-term and long-term effects of contaminants on the structure and function of the exposed plant communities and ecosystems (Bakalová 2006). An even more sensitive indicator of contamination may be seen in the avoidance test (Yeardley 1996). Organisms used in these assays have chemoreceptors sensitive to the

https://doi.org/10.17221/36/2018-SWR

Table 7. Overview of phytotoxicity tests (Pfleeger *et al*. 1991; US EPA 1996, 2012a, b, c; European Commission 2000b; ISO 2005a, b; 2012a, b; OECD 2006a, b; Jozífková 2011; ASTM 2014)

Test	Evaluated parameters	Guideline	Source
Terrestrial plant growth test	germination, growth (EC_{50}, LC_{50})	OECD 208	OECD (2006a)
Terrestrial plant growth test	vegetative vigour of growth	OECD 227	OECD (2006b)
Test of inhibition of root growth	root elongation (EC_x)	ISO 11269-1:2012	ISO (2012a)
Test of the emergence and early growth of higher plants	germination, growth (NOEC, LOEC)	ISO 11269-2:2012	ISO (2012b)
Chronic growth test	germination, growth, sprout length, number of plant buds	ISO 22030:2005	ISO (2005a)
Screening test for emergence of lettuce seedlings	germination, root elongation	ISO 17126:2005	ISO (2005b)
Seed germination/root elongation toxicity test	germination, root elongation (EC_{10}, EC_{50})	OPPTS 850.4200	US EPA (1996)
Plant growth test	germination, growth	OCSPP 850.4100	US EPA (2012a)
Early seedling growth toxicity test	growth of root, sprout and whole plant	OCSPP 850.4230	US EPA (2012b)
Plant uptake and translocation test	plant intake, translocation	OCSPP 850.4800	US EPA (2012c)
A short-term bioassay for whole plant toxicity	growth of root, sprout and whole plant (EC_{50})	ASTM STP1115	Pfleeger *et al.* (1991)
Standard guide for conducting terrestrial plant toxicity tests		ASTM E1963-09	ASTM E1963-09 (2014)

effect of some environmental pollutants (Edwards & Bohlen 1996; Römbke & Schmidt 1999). The earthworm avoidance test is set out in ISO 17512-1 (ISO 2008).

CONCLUSIONS

The use of sludge in agriculture within the EU is currently regulated mainly by limits on heavy metals (Cd, Cu, Hg, Ni, Pb and Zn) listed in Council Directive 86/278/EEC, which is outdated and does not reflect the current needs of ensuring the safety of sludge application in agriculture. The aim of the Directive was to encourage the safe use of sewage sludge in agriculture in order to prevent harmful effects on soil and transference to plants, animals and humans. Several European countries additionally imposed more stringent Directives and laws and set limits for concentrations of other heavy metals. The most common additional limit is set for chromium.

The legislation limits among individual EU countries vary widely by up to several orders of magnitude. For cadmium the limits range from 0.7 to 40 mg per kg DM, for copper from 75 to 1750 mg/kg DM, for mercury from 0.4 to 25 mg/kg DM, for nickel from 25 to 400 mg/kg DM, for lead from 45 to 1200 mg per kg DM, for zinc from 200 to 4000 mg/kg DM and for chromium from 70 to 1500 mg/kg DM. Furthermore, limit values for certain synthetic organic compounds were defined both in the Working Document on Sludge from 2000 and in national regulations. The most common is the limit for absorbable organic halogen (AOX), which is the same in all countries that have introduced this limit. The value is 500 mg per kg DM of sludge. The other most frequently used limits are for PAHs and PCBs.

The last group of pollutants for which limits have been defined is microbial contamination. Limits for the content of *Salmonella* and *Escherichia coli* are also proposed in the Working Document on Sludge. These indicators or even others (enterococci, thermotolerant coliform bacteria, *Clostridium perfringens*, and helminth eggs) are provided in the national legislation of just 11 countries.

Besides limits for sludge itself, the limits of heavy metals in soil intended for sludge application are also defined by legislative regulations. The differences in national limits are much smaller as compared to limits for sludge itself and range up to one order of magnitude.

The use of sewage sludge in agriculture in 2014 and 2015 in 13 EU countries which provided data amounted to 22.6% (2014) and 22.1% (2015) of produced sludge and 23.3% (2014) and 23.1% (2015) of sludge disposed. EU countries use a wide range of technologies for sludge processing, however, the

https://doi.org/10.17221/36/2018-SWR

predominant technologies are incineration (39.9% in 2014 and 41.5% in 2015 of sludge disposed) and direct application in agriculture and composting (39.2% in 2014 and 38.6% in 2015 of sludge disposed). The use of sewage sludge in agriculture is highly variable among EU countries, ranging between zero (Malta, Slovenia, Slovakia) and 80% (Ireland).

Phytotoxicity tests are valuable tools for assessing the applicability of sludge to agricultural land. Toxicity tests are included in the Directives of relevant agencies (e.g. US EPA, OECD, ISO, etc.).

The issue of sludge use in agriculture is very complex and carries many risks. The development of common European legislation, as well as national regulations, is important in order to avoid these risks. It is necessary to have the correct safety measures facilitating the prevention of possible leakage of contaminants into surface and ground water and to avoid toxic effects on soil, plants, animals and humans.

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