sustainability

MDPI

Review

Legislation for the Reuse of Biosolids on Agricultural Land in Europe: Overview

Maria Cristina Collivignarelli [1], Alessandro Abbà [2], Andrea Frattarola [1], Marco Carnevale Miino [1], Sergio Padovani [3], Ioannis Katsoyiannis [4,*] and Vincenzo Torretta [5]

1 Department of Civil and Architectural Engineering, University of Pavia, via Ferrata 1, 27100 Pavia, Italy; mcristina.collivignarelli@unipv.it (M.C.C.); andrea.frattarola01@universitadipavia.it (A.F.); marco.carnevalemiino01@universitadipavia.it (M.C.M.)
2 Department of Civil, Environmental, Architectural Engineering and Mathematics, University of Brescia, via Branze 43, 25123 Brescia, Italy; alessandro.abba@unibs.it
3 ARPA Lombardia, Pavia Department, via Nino Bixio 13, 27100 Pavia, Italy; s.padovani@arpalombardia.it
4 Department of Chemistry, Laboratory of Chemical and Environmental Technology, Aristotle University of Thessaloniki, 54124 Thessaloniki, Greece
5 Department of Theoretical and Applied Sciences, University of Insubria, via G.B. Vico 46, 21100 Varese, Italy; vincenzo.torretta@uninsubria.it
* Correspondence: katsogia@chem.auth.gr

Received: 17 September 2019; Accepted: 25 October 2019; Published: 29 October 2019

check for updates

Abstract: The issues concerning the management of sewage sludge produced in wastewater treatment plants are becoming more important in Europe due to: (i) the modification of sludge quality (biological and chemical sludge are often mixed with negative impacts on sludge management, especially for land application); (ii) the evolution of legislation (landfill disposal is banned in many European countries); and (iii) the technologies for energy and material recovery from sludge not being fully applied in all European Member States. Furthermore, Directive 2018/851/EC introduced the waste hierarchy that involved a new strategy with the prevention in waste production and the minimization of landfill disposal. In this context, biological sewage sludge can be treated in order to produce more stabilized residues: the biosolids. In some European countries, the reuse of biosolids as soil improver/fertilizer in arable crops represents the most used option. In order to control the quality of biosolids used for land application, every Member State has issued a national regulation based on the European directive. The aim of this work is to compare the different approaches provided by European Member States for the reuse of biosolids in agricultural soils. A focus on the regulation of countries that reuse significant amount of biosolids for land application was performed. Finally, a detailed study on Italian legislation both at national and regional levels is reported.

Keywords: biosolids; agricultural reuse; European legislation; sludge management; Italian legislation; policy choice

1. Introduction

During the last 20 years in EU-15, the sewage sludge produced by urban wastewater treatment plants (WWTPs) has increased from 6.5 million t_{DM} (dry matter) up to 9.5 million t_{DM} [1]; in EU-28, more than 10 million of t_{DM} of sewage sludge have been produced [2]. The contemporary presence of a low wastewater quality [3–5] and more stringent requirements for WWTPs effluents quality [6,7] increases the production, and worsens the quality, of the sewage sludge [8]. Therefore, the issues related to sewage sludge management are increasing. The introduction of waste hierarchy, with Directive 2018/851/EC [9], and the opposition of citizens (who perceived sewage sludge as dangerous for human health and for the environment) [10] forced the technicians to re-think completely the sewage sludge management strategy: landfilling was banned, land application was limited, and other routes of recovery are underdeveloped [11–13].

Directive 2018/851/EC [9] provided a waste hierarchy that shall apply as a priority order in waste prevention and management legislation and policy: prevention (e.g., minimization techniques), preparing for reuse (e.g., chemical or biological stabilization), recycling (e.g., matter recovery), other recovery (e.g., energy recovery), and disposal (e.g., landfilling). Usually, the terms "sewage sludge" and "biosolids" are used interchangeably, but "sludge" refers to a liquid produced by WWTPs that is not submitted to further treatments, while "biosolids" indicates a sludge that had received one or more treatments [14–16], which can be: aerobic or anaerobic digestion, thermal drying, alkaline stabilization, composting, acid oxidation/disinfection, etc. [16]. This work focuses on the matter recovery, in particular on the reuse of biosolids in agricultural soils.

The biosolids application on agricultural land can represent an interesting strategy to improve crops productivity by increasing soil organic matter (SOM) content, fertility, and nutrient presence; moreover, biosolids can also improve soil physical properties, especially in cases of heavy textured and poorly structured soils [17–20]. Furthermore, the spreading of biosolids on agricultural land reduces the effect of organic matter loss in the soil, especially in southern Europe, where the depletion of SOM is one of the most serious processes of soil degradation [21,22].

In addition, developing a sustainable and integrated circular system to reuse biosolids in land application can be entirely inserted in the concept of circular economy [23–25]. In recent years, the European Commission adopted an ambitious Circular Economy Package to promote the reuse, recycling, and recovery of wastes [26]. The reuse of biosolids on land application enhanced the recovery of resources (e.g., nutrients [27,28]) and, therefore allows changing the classic view of WWTP in a more sustainable water resources recovery facility (WRRF) [16,29–31].

The main problems related with the reuse of biosolids concern the presence of heavy metals, organic contaminants, and/or pathogens in the sludge [32,33]. In the scientific literature, no agreement can be found about the adverse effects caused by the land application of biosolids. According to the literature [34–36], the following aspects can be reported: (i) raising of the levels of persistent toxins in soil, vegetation, and wild life, (ii) potentially slow and long-termed biodiversity reduction through the fertilizing nutrient pollution operating on the vegetation, (iii) greenhouse gas emissions (e.g., CH_4 and N_2O), and (iv) the release of odorous compounds.

In some European countries, the reuse of biosolids as soil improver/fertilizer in arable crops represents the most used disposal option [1]. This trend led to restricting the use of biosolids based on quality—Directive 86/278/EEC [37] (and subsequent amendments [38,39]) introduced limit values in order to protect human health by applying biosolids with good qualities. Based on the European directive, every Member State has issued a national regulation, which, in some cases, provided more stringent limit values and introduce more restrictions, such as limit values for pathogen and organic micropollutants.

This work reports the management options in every Member State; moreover, the regulations of European countries for the reuse of biosolids in agricultural land are compared considering the limit values in the biosolids and in the soils. Furthermore, this work focuses on the regulations provided in the Member States that spread on agricultural soil a significant amount of biosolids (more than 300,000 t_{DM} year^{-1} or more than 70% with respect the sludge produced). Moreover, the comparison between the different requirements in the European countries, based on political aspects, are carried out in order to investigate the choices and the prospective on the reuse of biosolids in agriculture. Finally, a focus on the Italian situation, with a comparison of national and regional legislation, is reported.

It is important to note that the land application of biosolids includes both agricultural use and compost production, but not the reuse for silviculture, forest, land reclamation, and green areas.

2. Sewage Sludge Production and Management Options: European Situation

In EU-28, the urban sewage sludge production is more than 10 million t_{DM} [2]. The amounts are very different in the European countries, due to the percentage of the resident population connected to WWTPs and the technologies used. Germany, the UK, Spain, France, and Italy account more than 55–65% of the total amount produced in EU-28 with a decrease in the last years [1,2] due to the implementation of procedures for evaluating the performance of the WWTPs (as already suggested for drinking water treatment plants [40,41]) and for minimizing sewage sludge production with integrated technologies, both in wastewater and sludge-handling units [8,42–47]. On other side, there are countries with a low sewage sludge production due to small population (e.g., Malta, Latvia, Estonia, and Luxembourg) or due to the low percentage of resident population connected to WWTPs. For instance, in 2017 in Bulgaria, almost 13% of population was not served by any treatment plant [48].

Nowadays, in EU-28, land application is the main route for sewage sludge recovery: 50% of sewage sludge is spread on agriculture soils, 28% is incinerated, and 18% are still disposed in landfills [2]. The remaining fraction is disposed through other methods such as pyrolysis, storage (e.g., Greece, Italy, and Poland), reuse in green areas and forestry (e.g., Ireland, Latvia, and Slovakia), and landfill cover (e.g., Sweden) [1]. Figure 1 shows the sewage sludge recovery route for every Member State. The political choices on the alternative routes of sludge recovery/disposal, in particular for the land spreading, are strongly influenced by the population density and the availability of agricultural lands. The low availability of soils for the spreading of biosolids led the northern European countries (such as Netherlands and Germany) to choose incineration as the principle recovery route. Furthermore, even though all of the sludge produced could be applied in less than 5% of the agricultural area (in most Member States), the limited use of biosolids in agriculture is due to the low level of acceptance by farmers and the public [49]. This aspect influences policy decisions on sludge management, too; therefore, every Member State has issued a national regulation.

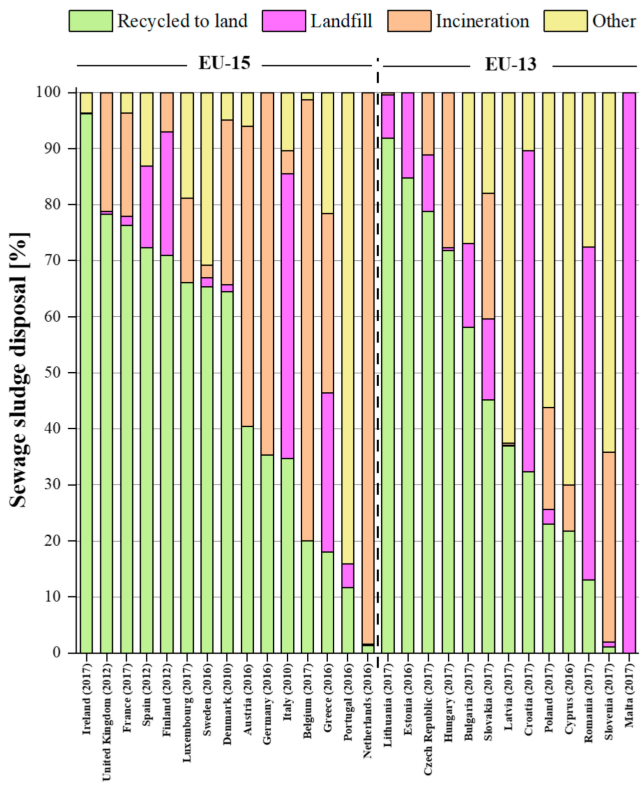

Figure 1. Sewage sludge recovery routes in Europe [2].

However, some inconsistencies in the EU terminology related to the management and recovery of sludge can be highlighted. For example, analyzing in more detail the trend of the data from 2012 to 2016 provided by Eurostat [2], the fraction reused in agriculture in some countries (e.g., Portugal) has decreased significantly unlike "other uses". However, it is legitimate to think that these "other uses" also include applications in agriculture. Moreover, sludge directed to grow plants for further compost production in Poland is classified in other countries as a sludge dewatering technique. Sometimes, in fact, the different national regulations lead to classifying the recoveries in different ways. This can sometimes create interpretation problems when the data is collected and made available by Eurostat.

3. The Reuse of Biosolids in Agriculture: Comparison Between Legislations in the European Countries

According to the data shown above, the yearly amount of biosolids recovered in agricultural soils is about 6 million t_{DM}. As reported in Figure 1, the scenarios (both in terms of quantities and percentages) are very different in the Member States: there are countries with high amounts of biosolids recovered in agriculture (e.g., France), and other states where percentage is close to zero (e.g., Malta).

Moreover, the legislation of the Member States is different: the national regulations, which have been established based on EU Directive 86/278/EEC [37], have often introduced provisions that go beyond the requirements of the directive. Every Member State issued own normative, which could have different limit values for biosolids and soils.

Furthermore, in order to reduce possible health risks, the regulations of some countries (e.g., France and Italy) include limit values for pathogens and in a larger number of cases for organic compounds, both of which are not included in Directive 86/278/CEE [37,50]. A new directive that provided limits for organic micropollutants and pathogens was proposed in 2000, but it was withdrawn [51].

Directive 86/278/EEC [37] allows Member States to choose between limit values in the soil or in the sludge: every country has chosen to establish limit values for biosolids and soil except the UK, which imposed limit values only for the soils. The countries can be classified into two different categories in relation to their comparison with Directive 86/278/EEC [37]: (i) with national requirements (in some cases even much) more stringent than the European directive, and (ii) with national requirements similar to those in the European directive [50]. The countries are classified as follows:

i. Czech Republic, Denmark, Finland, Luxembourg, the Netherlands, Sweden, Austria, Belgium, Malta, Croatia, France, Germany, Hungary, Lithuania, Poland, Slovenia, and Romania
ii. Bulgaria, Cyprus, Estonia, Latvia, Greece, Ireland, Italy, Portugal, Slovakia, Spain, and the United Kingdom

3.1. Limit Values for Biosolids

The presence of heavy metals in the biosolids is an important parameter to check before its spreading on land. Their presence in biosolids can be due to the presence of industrial wastewater treated by WWTPs [52,53]. Some heavy metals are considered essential micronutrients for plant growth, but elevated concentrations of these compounds are toxic to food crops, domestic animals, and humans [54,55]. It is also known that heavy metals are not biodegradable, and their persistence in soil is much longer than any other reactive components of the terrestrial ecosystems [33].

Table 1 shows a comparison of limit values for heavy metals in the biosolids provided in Directive 86/278/EEC [37] and the legislations in different Member States.

It can be observed that the limit values provided for the European countries are very different; moreover, almost all countries provide limit values for total chromium, and in some cases for arsenic, too. Furthermore, Austria, Italy, Hungary, Romania, and the United Kingdom introduced limit values for additional heavy metals in the biosolids: Styria and Lower Austria (Austrian Landers) provide limits for molybdenum (20 mg kg_{DM}^{-1}) and cobalt (10–100 mg kg_{DM}^{-1}); Italy for beryllium (20 mg kg_{DM}^{-1}) and selenium (10 mg kg_{DM}^{-1}); Hungary for molybdenum (20 mg kg_{DM}^{-1}), cobalt (50 mg kg_{DM}^{-1}) and selenium (100 mg kg_{DM}^{-1}); Romania for cobalt (50 mg kg_{DM}^{-1}); and the United Kingdom for molybdenum (3 mg kg_{DM}^{-1}), selenium (2 mg kg_{DM}^{-1}) and fluoride (200 mg kg_{DM}^{-1}).

Table 1. Limit values for heavy metals and organic compounds in the biosolids—all values are reported in mg kg_{DM}^{-1} [13,50,56,57].

Legislation	Heavy Metals								Organic Compounds							References
	Cd	Cr	Cu	Hg	Ni	Pb	Zn	As	PCB	AOX	LAS	DEHP	NP/NPE	PAH	PCDD/F [g]	
Directive 86/278/EEC	20–40	-	1000–1750	16–25	300–400	750–1200	2500–4000	-	-	-	-	-	-	-	-	[37]
EU-15																
Germany	10	900	800	8	200	900	4000	-	0.1 [o]	400	-	-	-	-	100	[58,59]
UK	-	-	-	-	-	-	-	2	-	-	-	-	-	-	-	[60]
Spain [a]	20–40	1500	1000–1750	16–25	300–400	750–1200	2500–4000	-	-	-	-	-	-	-	-	[61]
France	20	1000	1000	10	200	800	3000	-	0.8 [l]	-	-	-	-	2–5 [p]	-	[62]
Italy	20	200	1000	10	300	750	2500	20	0.8	-	-	-	-	6	25	[63,64]
Netherlands	1.25	75	75	0.75	30	100	300	15	-	-	-	-	-	-	-	[65]
Austria [b]	2–10	50–500	70–500	0.4–10	25–100	45–500	200–2000	20	0.2–1	500	-	-	-	6	50–100	[66–69]
Sweden [c]	0.75	40	300	1.5	25	25	600	-	0.4 [l]	-	-	-	50	3 [f]	-	[70]
Portugal	20	1000	1000	16	300	750	2500	-	0.8	-	5000	-	450	6	100	[71]
Finland	1.5	300	600	1	100	100	1500	25	-	-	-	-	-	-	-	[72]
Denmark	0.8	100	1000	0.8	30	120	4000	25	0.2 [l]	-	1300	50	10	3 [f]	-	[73,74]
Ireland	20	-	1000	16	300	750	2500	-	-	-	-	-	-	-	-	[75]
Greece [d]	20–40	500	1000–1750	16–25	300–400	750–1200	2500–4000	-	-	-	-	-	-	-	-	[76]
Belgium [b]	6–10	100–150	600–800	1–1.6	100	300–500	1500–2000	20–150	0.6–0.8 [l]	-	-	-	-	3–20	20	[77,78]
Luxembourg [d]	2.5	100	700	1.6	80	200	3000	-	0.2 [m]	-	-	-	-	20 [i]	20	[79]
EU-13																
Poland	20	500	1000	16	300	750	2500	-	-	-	-	-	-	-	-	[80]
Hungary	10	1000	1000	10	200	750	2500	75	1 [l]	-	-	-	-	10 [i]	-	[81]
Czech Republic	5	200	500	4	100	200	2500	30	0.6 [l]	500	-	-	-	10 [j]	-	[82,83]
Romania	10	500	500	5	100	300	2000	10	0.8 [l]	500	-	-	-	5 [k]	-	[84]
Lithuania [e]	1.5–20	140–400	75–1000	1–8	50–300	140–750	300–2500	-	-	-	-	-	-	-	-	[85]
Slovakia	10	1000	1000	10	300	750	2500	20	0.8 [l]	500	-	-	-	6 [h]	-	[86]
Bulgaria	30	500	1600	16	350	800	3000	25	-	-	-	-	-	-	-	[87]
Estonia	20	1000	1000	16	300	750	2500	-	-	-	-	-	-	-	-	[88]
Cyprus [d]	20–40	-	1000–1750	16–25	300–400	750–1200	2500–4000	-	-	-	-	-	-	-	-	[89]
Latvia [f]	2–10	100–600	400–800	3–10	50–200	150–500	800–2500	-	-	-	-	-	-	-	-	[90]
Slovenia	1.5	200	300	1.5	75	250	1200	-	-	-	-	-	-	-	-	[91]
Malta	5	800	800	5	200	500	2000	-	-	-	-	-	-	-	-	[92]
Croatia	5	500	600	5	80	500	2000	-	0.2 [n]	-	-	-	-	-	100	[93]

[a] Different values for different soil pH; [b] Different values for different regions/Landers; [c] Value expressed as g ha^{-1} $year^{-1}$; [d] Lower values = recommended values, higher values = maximum limits (the same limits of Directive 86/278/EEC); [e] Different values for different sludge categories; [f] Different values for capacity of wastewater treatment plants (WWTPs) (expressed in P.E.—population equivalent); [g] PCDD/F are expressed in ng TE (Toxic Equivalency) kg_{DM}^{-1}; [h] 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; [i] Sum of 16 United States Environmental Protection Agency (US EPA) PAH (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); [j] 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, and pyrene; [k] Sum of anthracene, benzoanthracene, benzofluoranthene, benzoperylene, benzopyrene, chrysene, fluoranthene, indeno (1,2,3)pyrene, naphthalene, fenanthrene, and pyrene; [l] Sum of seven congeners: PCB 28, 52, 101, 118, 138, 153, and 180; [m] Sum of six congeners: PCB 28, 52, 101, 138, 153, and 180; [n] For each of these congeners: PCB 28, 52, 101, 141, and 180; [o] For each congener; [p] Different values for different compounds (fluoranthene-5, benzo(b)fluoranthene-2.5, benzo(a)pyrene-2); PCB: polychlorinated biphenyls; AOX: absorbable organic halogens; LAS: linear alkylbenzene sulfonates; DEHP: di(2-ethylhexyl) phthalates; NP: nonylphenols; NPE: nonylphenol ethoxylates; PAH: polycyclic aromatic hydrocarbons; PCDD/F: polychlorinated dibenzo-p-dioxins and furan; UK: United Kingdom.

Despite Directive 86/278/EEC [37] not providing any limit values or requirements for organic micropollutants in biosolids, several national regulations on the use of biosolids have added specifications, as shown in Table 1. It can be noted that the organic compounds most checked by different Member States are PCB (polychlorinated biphenyls), AOX (absorbable organic halogens), and PAH (polycyclic aromatic hydrocarbons).

In order to reduce possible health risks related to pathogens, different Member States (e.g., Austria and Bulgaria) have added specific requirements for biosolids spread on land. Despite Directive 86/278/EEC [37] not including limit values for the pathogens content in biosolids, most countries' national legislation checks the presence of salmonella (with the exception of Lithuania, Luxembourg, and Slovakia) and, in many cases, other pathogens are included. Types of pathogens and limit values are quite different (see Table 2).

Table 2. Limit values for pathogens in the biosolids [13,50,56,57].

Legislation	Types of Pathogens	Limit	Units of Measure (u.m.)	References
Directive 86/278/EEC	-	-	-	[37]
Austria [1]				
	Enterococci	<10^3	CFU g_{DM}^{-1}	[66–69]
	Escherichia Coli	100	CFU g_{DM}^{-1}	
	Helminths eggs	Absent	eggs kg_{DM}^{-1}	
	Salmonella	No occurrence in 1 g		
Bulgaria				
	Salmonella	Absent	MPN 20^{-1} g_{WW}^{-1}	[87]
	Escherichia Coli	100	MPN g_{WW}^{-1}	
	Clostridium perfringens	300	MPN g_{WW}^{-1}	
	Viable eggs of helminths	1	eggs kg_{DM}^{-1}	
Czech Republic				
	Salmonella	Absent	CFU g_{DM}^{-1}	[82,83]
	Thermotolerant coliforms	<10^3	CFU g_{DM}^{-1}	
	Enterococci	<10^3	CFU g_{DM}^{-1}	
Denmark [2]				
	Salmonella	No occurrence		[73,74]
	Faecal streptococci	<100 g^{-1}		
Finland				
	Salmonella	Not detected in 25 g		[72]
	Escherichia Coli	1000	CFU g^{-1}	
France				
	Salmonella	8	MPN $10^{-1}g_{DM}^{-1}$	[62]
	Enterovirus	3	MPCN $10^{-1}g_{DM}^{-1}$	
	Helminths eggs	3	eggs $10^{-1}g_{DM}^{-1}$	
Italy				
	Salmonella	1000	MPN g_{DM}^{-1}	[63,64]
Lithuania				
	Escherichia Coli	1000	CFU g^{-1}	[85]
	Helminths eggs	0	Units kg^{-1}	
	Enterobacteria	0	CFU g^{-1}	
	Clostridium perfringens	100,000	CFU g^{-1}	
Luxembourg				
	Enterobacteria	<100 g^{-1}		[79]
	Helminths eggs	No eggs of worm likely to be contagious		
Malta				
	Salmonella	Absent	CFU 50^{-1} g_{WW}^{-1}	[92]
Poland				
	Salmonella	Biosolids cannot be used in agriculture if it contains salmonella in 100 g_{DM}		[80]
	Helminths eggs	0	eggs kg_{DM}^{-1}	
Portugal				
	Salmonella	No occurrence in 50 g		[71]
	Escherichia Coli	1000	CFU g^{-1}	
Slovakia				
	Thermotolerant coliforms	2×10^6	CFU g_{DM}^{-1}	[86]
	Fecal streptococci	2×10^6	CFU g_{DM}^{-1}	

[1] Only for three Lander: Carinthia, Lower Austria, and Styria; [2] For advanced treated sludge only; u.m.: units of measure; MPN: most probable number; MPCN: most probable cytopathic number; CFU: colony-forming unit; WW: wet weight; DM: dry matter.

The regulatory requirements on pathogens content in biosolids still remains quite limited in most countries' national legislation. This can be partially explained by the fact that national codes of practice are considered to sufficiently cover this issue, by providing recommendations on sludge treatment (mainly with biological, chemical, and heat treatments) and biosolids land spreading (i.e., the UK).

Finally, in order to have a good fertilizer, biosolids must contain a significant amount of agronomic parameters such as organic matter, nitrogen, and phosphorus. Most of the Member States (e.g., France and Spain) prescribe the analyses of agronomic parameters, but only Italy sets minimum values for organic matter (>20% of dry matter (DM)), total nitrogen (>1.5% of DM), and total phosphorus (>0.4% of DM). Instead, other countries (such as Sweden and Latvia) provide a maximum annual rate of nitrogen and/or phosphorus that can be spread on land.

3.2. Limit Values for Soil

In addition to restriction for heavy metals in biosolids, the European legislation provides limit values for heavy metals in soils. In all Member States, regulations on the use of biosolids specify limit values, which are in most cases similar or lower than the requirements set in Directive 86/278/EEC [37] (Table 3). The limit values have been defined by three different criteria as follows:

i. *Soil pH*: Bulgaria, Slovenia, Romania, Croatia, Latvia, Malta, Portugal, the United Kingdom, and Spain defined limit values according to the soil pH, which vary between 5 and 7.5. In some cases, these values can change in relation to the different region/lander (e.g., in Austria and in Belgium).
ii. *Granulometric content*: Czech Republic, Lithuania, and Latvia based their prescription on soil granulometric content (sand or clay). Poland adopted another classification (heavy, medium, or light soils) which is not yet defined by the decree [94].
iii. Italy uses cation exchange capacity (CEC) in order to characterize soil for land spreading of biosolids.

Table 3. Limit values for heavy metals in the soil. All values are reported in mg kg_{DM}^{-1} [13,50,56,57].

Legislation	Cd	Cr	Cu	Hg	Ni	Pb	Zn	As	References
Directive 86/278/EEC	1–3	-	50–140	1–1.5	30–75	50–300	150–300	-	[37]
EU-15									
Germany	0.4–1.5	30–100	20–60	0.1–1	15–70	40–100	60–200	-	[58,59]
United Kingdom	3	400	80–200	1	50–110	300	200–300	50	[60]
Spain [1]	1–3	100–150	50–210	1–1.5	30–112	50–300	150–450	-	[61]
France	2	150	100	1	50	100	300	-	[62]
Italy	1.5	-	100	1	75	100	300	-	[63,64]
Netherlands	0.8	10	36	0.3	30	35	140	-	[65]
Austria [2]	0.5–2	50–100	40–100	0.2–1.5	30–70	50–100	100–300	-	[66–69]
Sweden	0.4	60	40	0.3	30	40	100	-	[70]
Portugal [1]	1–4	50–300	50–200	1–2	30–110	50–450	150–450	-	[71]
Finland	0.5	200	100	0.2	60	60	150	-	[72]
Denmark	0.5	30	40	0.5	15	40	100	-	[73,74]
Ireland	1	-	50	1	30	50	150	-	[75]
Greece [3]	1–3	-	50–140	1–1.5	30–75	50–300	150–300	-	[76]
Belgium [2]	1.2–1.5	91–100	50–72	1–1.5	20–56	50–120	200	22	[77,78]
Luxembourg [3]	2	150	100	1.5	75	200	300	-	[79]
EU-13									
Poland [4]	1–3	50–100	25–75	0.8–1.5	20–50	40–80	80–180	-	[80]
Hungary	1	75 (Cr^{VI})	75	0.5	40	100	200	15	[81]
Czech Republic [4]	0.4–0.5	55–90	45–60	0.3	45–50	55–60	105–120	15–20	[82,83]
Romania	3	100	100	1	50	50	300	-	[84]
Lithuania [4]	1–1.5	50–80	50–80	0.6–1	50–60	50–80	160–260	-	[85]
Slovakia	1	60	50	0.5	50	70	150	25	[86]
Bulgaria [1]	1.5–3	200	80–200	1.5	75–110	60–120	200–300	25	[87]
Estonia	3	100	50	1.5	50	100	300	-	[88]
Cyprus [3]	1–3	-	50–140	1–1.5	30–75	50–300	150–300	-	[89]
Latvia [1,4]	0.5–0.9	40–90	15–70	0.1–0.5	15–70	20–40	50–100	-	[90]
Slovenia	1	100	60	0.8	50	85	200	-	[91]
Malta [1]	0.5–1.5	30–100	20–100	0.1–1	15–70	70–100	60–200	-	[92]
Croatia [1]	0.5–1.5	50–100	40–100	0.2–1	30–70	50–100	100–200	-	[93]

[1] Different values for different soil pH values; [2] Different values for different regions/Lander; [3] Lower values = recommended values, Higher values = maximum limits (the same limits as those of Directive 86/278/EEC); [4] Different values for different types of soil.

Greece and Cyprus have minimum values, which are recommended, and threshold values. In addition, the legislation in several Member States (Austria, Belgium, Cyprus, Estonia, Greece, Finland, France, Hungary, Latvia, Lithuania, Luxembourg, Netherlands, Portugal, Slovakia, Slovenia, Spain, Sweden, Romania, and the UK) includes restrictions in terms of the maximum annual load of heavy metals on agricultural land. Moreover, some countries (Austria, Hungary, and the UK) introduce limit values for additional heavy metals in the soil—Styria (Austria Lander) provides limits for molybdenum (10 mg $kg_{DM}{}^{-1}$) and cobalt (50 mg $kg_{DM}{}^{-1}$); Hungary for molybdenum (7 mg $kg_{DM}{}^{-1}$), cobalt (30 mg $kg_{DM}{}^{-1}$), and selenium (1 mg $kg_{DM}{}^{-1}$); and the United Kingdom for molybdenum (4 mg $kg_{DM}{}^{-1}$), selenium (3 mg $kg_{DM}{}^{-1}$) and fluoride (500 mg $kg_{DM}{}^{-1}$).

Studies on the heavy metals content in the soils due to the application of biosolids were carried out, but in some cases, the results are conflicting. For instance, Collivignarelli et al. [53] analyzed the heavy metal presence in the soils where the spreading of biosolids occurs during 2012 to 2016 in Lombardy (Italy). In the analyzed area, the soils presented mainly silty–sandy components [95]. They found that heavy metal average concentrations were largely below the national and regional normative limits in the whole period of the survey [53]. Only sporadic overruns of the Zn, Ni, and Cd have been reported [53]. However, in a study conducted in the USA, Islam et al. [33] studied the effects of the application of biosolids on well-drained silt soil, and they found that the heavy metal concentration increased significantly. Moreover, they highlighted that the extractable fractions of Pb, As, Zn, and Cu were significantly higher at soil depth from 0 to 15 cm. Consequently, they noted that accumulated heavy metals may mobilize from the soils to groundwater and surface water bodies [33]. Several studies try to understand the effect of heavy metals content on soil, due to biosolids application, on crops [96,97].

4. Land Spreading as Main Route: Detailed Survey of European Legislations

4.1. Comparison Between Regulations in EU-15 Member States with Land Spreading as Main Route

This section concerns the detailed study (and comparison) of regulations in EU-15 Member States where the recovery on land represents the main routes for sludge management. The countries considered in this section are the Member States where the percentage of biosolids recycled to land is more than 70%, or the amount of biosolids spread on soil is higher than 300,000 t_{DM} year^{-1} [2]—France, Finland, Germany, Ireland, Italy, Spain, and the United Kingdom. In addition, Denmark has been considered due to its very detailed regulation on the reuse of biosolids [73,74]. At a first analysis, in 2016, Portugal seemed to show a very low value of reuse in agriculture. However, as already discussed in Section 2, analyzing in greater detail the trend of data from 2012 to 2016 provided by Eurostat [2], the fraction reused in agriculture decreased significantly (from 90% to 12%) in spite of "other uses" (from 0% to 84%). However, it is legitimate to think that these other uses also include applications in agriculture, such as compost. For this reason, Portugal was still included in the analysis. The selected countries, the percentage of biosolids recycled to land, and the annual amount of biosolids spread on soil are reported in Figure 2.

Considering that the selected countries belong to different climatic areas (Mediterranean, continental, oceanic, and subarctic), with different types of soils, this allows developing a complete view of the situation in Europe concerning the spreading of biosolids on the soil.

The legislation of the countries analyzed presents significant differences, in addition to differences among the heavy metals, pathogen, and organic micropollutants (see Tables 1–3), as concerns, there is also: (i) the maximum amount of biosolids spread on land; (ii) the soils on which the use of biosolids is prohibited; and (iii) the treatment requirements.

The maximum amount of biosolids that can be spread on land is not prescribed in Directive 86/278/EEC, but the regulation states that it is necessary to limit the amount of heavy metals added to cultivated soil [37]. Thus, five countries have set the maximum amount of biosolids that can be spread on land per year (Table 4). The maximum amount of biosolids differs as well as their quality—for example, Denmark allows spreading 10 t_{DM} ha^{-1} year^{-1} on land, but it prescribes requirements more stringent than the European directive. In contrast, Ireland set a maximum amount of biosolids that is significantly lower than that of Denmark (2 t_{DM} ha^{-1} $year^{-1}$), but in this case, the limit values for pollutants in biosolids are similar to Directive 86/278/EEC [37]. Therefore, the maximum load of pollutants (heavy metals, pathogens, organic micropollutants) permitted on agricultural land in different Member States is comparable. Moreover, different amounts of biosolids are associated with SOM declines in different countries: the content of SOM tends to decrease when moving from a warmer (e.g., Italy, Spain) to cooler climate (e.g., Denmark, Sweden). This is due to the overall trend in the decomposition of SOM that is accelerated in warm climates, while a lower rate of decomposition is the case for cool regions [21].

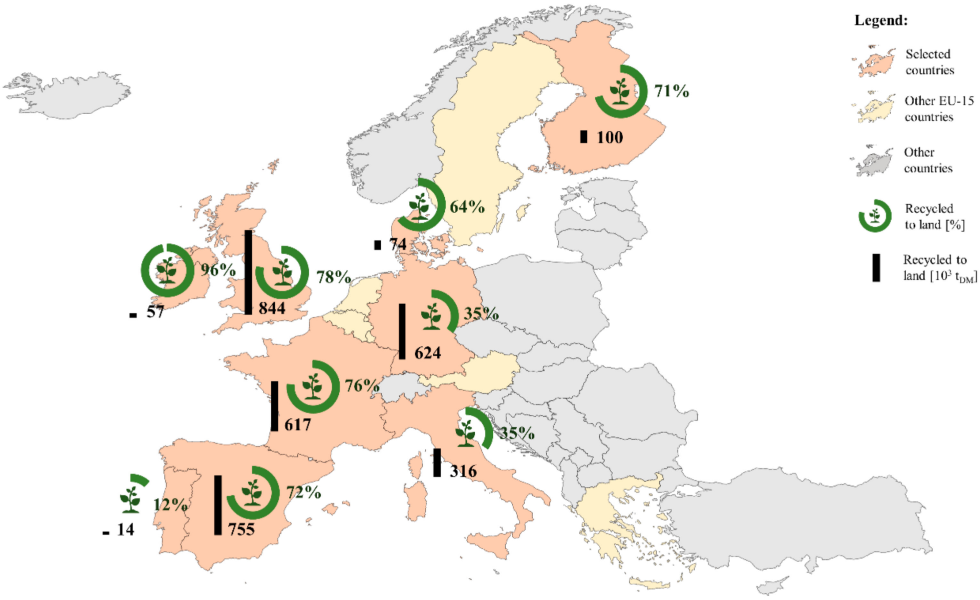

Figure 2. The selected countries, the percentage of biosolids recycled to land, and the total annual amount of biosolids spread on soil [2].

Nitrates Directive 91/676/EEC [98] (and subsequent amendments [99,100]) set out the maximum permissible addition of total nitrogen which for most purposes is 250 kg_N ha^{-1} $year^{-1}$ and, generally, it represents the limiting factor determining the rate of application of biosolids to the land. This value will be reduced in Nitrate Vulnerable Zones to 175 kg_N ha^{-1} $year^{-1}$. In some cases, it may be permissible to apply 500 kg_N ha^{-1} every two years if the nitrogen availability of the material is low (e.g., sludge compost, dewatered sludge cake) [49]. Currently, soils in different areas of Europe, particularly in the Mediterranean area, suffer from nutrient depletion [101,102]. Therefore, the application of biosolids can represent a solution to this problem [16,103].

Concerning the soils on which the use of biosolids is prohibited, Directive 86/278/EEC [37] (article 7) provides restrictions concerning the spreading of biosolids on grazing and pastureland, and on land on which vegetables and fruits are grown. These dispositions have been transposed by Member States, which have introduced additional requirements (Table 4) for land spreading.

Concerning the requirement for sludge treatment, Directive 86/278/EEC specifies that sludge need to be treated before being spread on land in order to reduce its fermentability and the health hazards [37]. However, the European directive allows the use of untreated sludge in the case of injection or if it is worked into the soil [37]. Most countries prohibit the use of untreated sludge, while some Member States (France, Ireland, and the UK) permit the use of untreated sludge [50].

Table 4. Maximum amount and soils on which the use of biosolids is prohibited [50,56].

	Directive 86/278/EEC	Denmark	Finland	France	Germany	Ireland	Italy	Portugal	Spain	UK
Maximum quantitative (t_{DM} ha^{-1} $year^{-1}$)	-	10	-	-	1.7	2	5	6	-	-
Obligations for treatment	Permit use of untreated sludge under certain conditions	Stabilization, composting or pasteurization	Digestion or lime stabilization	Permit use of untreated sludge under certain conditions	Prohibition of use of untreated sludge	Permit use of untreated sludge under certain conditions	Prohibition of use of untreated sludge	Prohibition of use of untreated sludge	Prohibition of use of untreated sludge	Permit use of untreated sludge under certain conditions
Soil										
Grazing and pastureland	P	P	P	P	P	P	P	P	P	P
Vegetables and fruits crops	P	P	P	P	P	P	P	P	P	P
Frozen or snow-covered ground	NP	NP	NP	P	NP	NP	NP	NP	NP	NP
Sloping land	NP	NP	NP	P	NP	NP	P	NP	NP	NP
Wet land, or after heavy rain	NP	NP	NP	P	NP	NP	P	P	NP	NP
GW protection areas	NP	P	NP	P	P	NP	NP	P	NP	NP
Near surface water	NP	P	NP	P	P	NP	NP	P	NP	NP
Forest Soil	NP	P	NP	P	P	NP	NP	NP	NP	NP
Additional restriction		On soil where sludge is likely to cause significant nuisances or unsanitary conditions		Not regularly worked out land in areas close to human settlements and public buildings			Soils of pH <5, and CEC <8 meq 100^{-1} g^{-1}	In areas close to individual houses and human settlements		Soils of pH <5

P: Prohibited; NP: Not prohibited; GW: Groundwater; UK: United Kingdom; meq: milliequivalents.

4.2. Political Choices in Land Spreading Regulations

As reported in the Section 3, the countries mentioned above could be classified into two different categories depending on the severity of existing legislation. The differences are connected to different aspects (e.g., the economic impact, the public perception, etc.), but in particular to the extension of agricultural land and the stakeholders' (farmers, landowners, communities, etc.) positions that influence the policy decisions.

In the Member States with national requirements (even much) more stringent than the European directive, farmers and landowners' associations have expressed their growing hostility toward the agricultural use of biosolids [56]. In Germany and France, the normative is more stringent than the European directive, and the debate is open: the stakeholders have divergent opinions [56,104]. These conflicting positions in France and Germany have led to changing the regulation despite different stakeholders highlighting the problem connected with high limit values [56]. In Denmark and Finland, instead, in addition to the hostility of stakeholders, there is less land suitable for the land spreading of urban biosolids [56]. For this reason, the government introduced new legislation (e.g., in Denmark [74]) but, despite the improvements in biosolids quality, the perception on land spreading remains negative [105].

Finally, for the Member States that have requirements similar to the European directive (Ireland, Italy, Portugal, Spain, and the United Kingdom), the scenarios vary widely. In most cases (e.g., Ireland, Italy, Portugal, and Spain), there is not a real debate on the agricultural recycling of biosolids [105], while in other cases (e.g., the UK), the debate is over [56]. Despite the absence of a real debate leading these countries to follow the values imposed by the European directive, stakeholders' opinions change in different countries [56]. For instance, in Ireland, the majority of farmers and national authorities are reportedly very positive about land, although some concerns remain regarding environmental problems [105,106]. In Portugal, someone highlighted the worse quality of the biosolids [56,107].

5. Italian Scenario: Comparison of Legislation at National and Regional Level

In Italy, the reuse of biosolids in agriculture in 2012 was about 30% [53]. Agricultural reuse is the most prevalent route in the regions of northern Italy. In fact, it can be observed that the total amounts of biosolids reused in agricultural land was more than 50% in Lombardy, Emilia Romagna, and Veneto [108].

This context, in which the reuse of biosolids on soil represents a significant route, has led to issuing regional legislations in order to regulate the reuse operation in these territories; in particular, Lombardy, Veneto, and Emilia Romagna have their own regional regulation derived from the Italian legislation.

These regions prescribe the same limit values of Legislative Decree n.99/1992 [64] for heavy metals in soil (Table 3). Instead, for the biosolids that can be spread on agricultural land, the regional regulations, especially for Lombardy, initially show significantly different issues (Table 5). As a consequence of a new Italian regulation [63], Italy, Lombardy, Veneto, and Emilia Romagna imposed the same limits on heavy metals and organic micropollutants [109–111]. Furthermore, legislation at the regional level has maintained the same limit of Italy regulation for salmonella and for agronomic parameters (organic carbon, total nitrogen, and phosphorus).

An interesting approach was introduced in the Lombardy regulation that concerns the classification of biosolids in two different classes [112]:

- the "biosolids suitable for spreading", which must respect the limit values imposed by the current Italian normative;
- the "high quality biosolids", which requires more stringent limit values (similar to the limits prescribed in Denmark).

Moreover, for both kinds of biosolids, the volatile suspended solids (VSS) to total suspended solids $(TSS)^{-1}$ ratio and organic micropollutants (also for the regulation of Emilia Romagna and Veneto) are prescribed. In particular, the VSS TSS^{-1} are provided in order to minimize the problems concerning the odor emissions (one of the main critical issues of biosolids land spreading).

Table 5. Limit values, maximum quantitative values, and soil on which the use of biosolids is prohibited in Italy and in some regions [63,64,109–111,113–115].

Parameter.		Unit of Measure (u.m.)	Italy	Lombardy		Veneto	Emilia Romagna
			Legislative Decree n.99/1992 and n. 130/2018	DGR n. X/2031/2014 and d.d.u.o. 6665/2019		DGR n. 2241/2005 and n. 130/2018	DGR n. 285/2005 and n. 326/2019
				Suitable	High Quality		
Heavy metals	As	mg kg_{DM}^{-1}	<20	≤10	≤20	<20	<20
	Be	mg kg_{DM}^{-1}	≤2	≤2		≤2	≤2
	Cd	mg kg_{DM}^{-1}	<20	≤20	≤5	<20	≤20
	Cr	mg kg_{DM}^{-1}	<200	≤200	≤150	<200	≤200
	Cr (VI)	mg kg_{DM}^{-1}	<2	<2		<2	<2
	Cu	mg kg_{DM}^{-1}	<1000	≤1000	≤400	<1000	≤1000
	Hg	mg kg_{DM}^{-1}	<10	≤10	≤5	<10	≤10
	Ni	mg kg_{DM}^{-1}	<300	≤300	≤50	<300	≤300
	Pb	mg kg_{DM}^{-1}	<750	≤750	≤250	<750	≤750
	Se	mg kg_{DM}^{-1}	≤10	≤10		≤10	≤10
	Zn	mg kg_{DM}^{-1}	<2500	≤2500	≤600	<2500	≤2500
Chemical/physical parameters	VSS TSS^{-1}	%	-	<65	<60	-	-
	pH	-	-	5.5 < pH ≤ 11		-	>5.5
Agronomic parameters	Organic carbon	% DM	>20	>20		>20	>20
	Total nitrogen	% DM	>1.5	>1.5		>1.5	>1.5
	Total phosphorus	% DM	>0.4	>0.4		>0.4	>0.4
Organic micropollutants	PAH	mg kg_{DM}^{-1}	≤6	<6		≤6	≤6
	PCB	mg kg_{DM}^{-1}	≤0.8	<0.8		≤0.8	≤0.8
	AOX	mg kg_{DM}^{-1}	-	<500		-	<500
	LAS	mg kg_{DM}^{-1}	-	-		-	<2600
	Toluene	mg kg_{DM}^{-1}	≤100	≤100		≤100	≤100
	DEHP	mg kg_{DM}^{-1}	-	<100		-	≤100
	NPE	mg kg_{DM}^{-1}	-	<50		-	≤50
	PCDD/F	$ng_{TE}\ kg_{DM}^{-1}$	≤25	≤25		≤25	≤25
	Hydrocarbon (C10-C40)	mg kg_{DM}^{-1}	-	<10,000		-	-
	Hydrocarbon (C10-C40)	mg kg_{WW}^{-1}	<1000	<1000		≤1000	≤1000
Microbiological parameters	Salmonella	MPN g_{DM}^{-1}	<1000	<100		<1000	≤1000
	Faecal Coliforms	MPN g_{DM}^{-1}	-	<10,000		-	-
Maximum quantitative (based on pH and CEC values)		t_{DM} ha^{-1} $year^{-1}$ for maximum quantitative meg 100^{-1} g^{-1} for CEC	2.5 pH < 6; CEC < 15	2.5	pH < 6; CEC ≤ 15	2.5 5 < pH < 7.5; CEC < 15	2.5 5 < pH < 6; CEC ≤ 15
				3.7	5 < pH < 6; CEC > 15	2.5 5 < pH < 6; CEC > 15	3.7 5 < pH < 6; CEC > 15
			5 6 < pH < 7.5; CEC > 15	3.7	6 < pH < 7.5; CEC ≤ 15	5 6 < pH < 7.5; CEC > 15	3.7 6 < pH < 7; CEC ≤ 15
				5	6 < pH < 7.5; CEC > 15	5 pH > 7.5; CEC < 15	5 6 < pH < 7.5; CEC > 15
							5 pH > 7.5; CEC ≤ 15
			7.5 pH > 7.5	7.5	pH > 7.5	7.5 pH > 7.5; CEC > 15	7.5 pH > 7.5; CEC > 15

Table 5. *Cont.*

Parameter.	Unit of Measure (u.m.)	Italy	Lombardy		Veneto	Emilia Romagna
		Legislative Decree n.99/1992 and n. 130/2018	DGR n. X/2031/2014 and d.d.u.o. 6665/2019		DGR n. 2241/2005 and n. 130/2018	DGR n. 285/2005 and n. 326/2019
			Suitable	High Quality		
Soil	Grazing and pastureland	P	P		P	P
	Vegetables and fruits crops	P	P		P	P
	Frozen or snow-covered ground	NP	P		P	P
	Sloping land	P	P		P	P
	Wet land, or after heavy rain	P	P		P	P
	Groundwater protection areas	NP	P		P	P
	Near surface water	NP	P		P	P
	Forest soil	NP	P		P	NP

u.m.: unit of measure; meq: milliequivalents; TE: toxic equivalency; PCB: polychlorinated biphenyls; AOX: absorbable organic halogens; LAS: linear alkylbenzene sulfonates; DEHP: di(2-ethylhexyl) phthalates; NP: nonylphenols; NPE: nonylphenol ethoxylates; PAH: polycyclic aromatic hydrocarbons; PCDD/F: polychlorinated dibenzo-p-dioxins and furan; CEC: cation exchange capacity; DM: dry matter; VSS: volatile suspended solids; TSS: total suspended solids; WW: wet weight; P: prohibited; NP: not prohibited.

It should be noted that exceeding the limit values for "biosolids suitable for spreading" involves choosing another route (such as incineration).

Concerning the maximum amount of biosolids that can be spread on land, the regulations at the regional level are different from those at the national one. The Italian legislation provides three soil categories based on pH and CEC, while Emilia Romagna, Veneto, and Lombardy provide five to six categories with different maximum amounts of biosolids.

Moreover, the regional regulations provide a more detailed list of soil on which the use of biosolids is prohibited, with respect to Legislative Decree n.99/1992 [64]. The use of biosolids on grazing, pastureland, and land on which vegetables and fruits are grown (prescribed by Directive 86/278/EEC [37]) is prohibited both at national and regional levels. As shown in Table 5, while Italian regulation prohibits only the use of biosolids on sloping land and wetland, for Lombardy, Veneto, and Emilia Romagna, the application of biosolids on other soil is prohibited.

Concerning the treatment of sludge, Legislative Decree n.99/1992 [64] provides the requirement for treatment (e.g., stabilization) for biosolids before the use on agricultural soil. This aspect is also prescribed in regional regulations.

Finally, an important aspect introduced in Italian and regional regulation is the limit for hydrocarbon C10–C40—the concentration must be equal or less than 1000 mg kg_{WW}^{-1}, and only for the Lombardy Region, it must be less than 10,000 mg kg_{DM}^{-1}. The introduction of a hydrocarbon limit is becoming important in Europe, too, and some countries have imposed limits in their regulations such as Hungary (4000 mg kg_{DM}^{-1} for C5–C40) and Belgium (5600 mg kg_{DM}^{-1} for C20–C40 in the Flemish region) [57].

Another possibility for the reuse of biosolids is through the production and spreading of defecation gypsum on land. In this way, a fertilizer is produced that does not fall under the legislation of Legislative Decree 99/1992 [64] and which, on suitable land, can replace the biosolids regulated by the aforementioned decree. It should be noted that in more than one case, the spreading of defecation gypsums can rise to a significant malodor, without it being clarified whether it is due to the material or to the spreading methods. The defecation gypsums are classified by the Legislative Decree 75/2010 [116] (and subsequent amendments [117]). The qualitative characteristics of defecation gypsums must comply with the limits shown in Table 6. As can be seen, the concentration of heavy metals allowed in sludge defecation plasters is significantly lower than that imposed for the spreading of biosolids in agriculture.

Table 6. Limiting concentrations of heavy metals and microbiological parameters for defecation gypsum from sludge and biosolids [109,113,116,117].

Parameter		Unit of Measure (u.m.)	D.Lgs. 75/2010 and M.D. 28 June 2016	DGR n. X/2031/2014 and d.d.u.o. 6665/2019	
				Suitable	High Quality
Heavy metals	Pb	mg kg_{DM}^{-1}	140	750	250
	Cd	mg kg_{DM}^{-1}	1.5	20	5
	Ni	mg kg_{DM}^{-1}	100	300	50
	Zn	mg kg_{DM}^{-1}	500	2500	600
	Cu	mg kg_{DM}^{-1}	230	1000	400
	Hg	mg kg_{DM}^{-1}	1.5	10	5
	Cr^{VI}	mg kg_{DM}^{-1}	0.5	<2	<2
Microbiological parameters	Salmonella	MPN g_{DM}^{-1}	Absence in 25 g of sample as it is; *n* (1) = 5; c (2) = 0; m (3) = 0; m (4) = 0	100	100
	E. Coli	MPN g_{DM}^{-1}	In 1 g of sample as it is.; *n* (1) = 5; c (2) = 1; m (3) = 1000 CFU g^{-1}; m (4) = 5000 CFU g^{-1}	-	-
	Total Coliforms	MPN g_{DM}^{-1}	-	<10,000	<10,000
Minimum titulus	CaO	-	15% on DM	-	-
	SO_3	-	10% on DM	-	-

u.m.: unit of measure; MPN: most probable number; CFU: colony-forming unit; DM: dry matter.

6. Discussion

Analysing and comparing the different regulations of EU countries, there is a strong heterogeneity in the monitored parameters and in the limits imposed. While some countries have imposed only the limits reported in EU Directive 86/278/EEC, others have imposed more stringent limits and/or additional parameters. As regards the limits of heavy metals in soils, this difference is partially justifiable by the marked pedoclimatic differences that involve the European continent, and which have therefore led countries to practice different choices. It would be interesting to foresee in the European directive different ranges of limits according to the prevailing pedoclimatic zone in each nation. Although not easy to implement, this method would allow countries to better adapt their legislation to the different needs of the territories.

Different types of soil react differently to the same contribution of pollutants. To date, this aspect is little considered by the legislation. However, in some countries such as in Italy, national legislation already differentiates the maximum amounts of biosolids to be spread according to the pH and the CEC of soils. One of the key points derived from the analysis of the current regulatory overview could be to differentiate soils, even in countries where this approach is not currently followed, by distinguishing the maximum amounts of biosolids spread on the basis of the maximum value of the pollutant load tolerable by soils. In fact, providing a limit to the pollutant load and not to the concentration would make it possible to operate more efficiently considering the effective response capacity of the soils.

As for the actions on the quality of the reused biosolids in the agricultural land, the introduction of specific legislation that regulates the acceptability limits for the sludge inlet to sludge treatment plants (STPs) would improve the selection of sludge and the quality of biosolids recovered. In fact, STPs are often not equipped to remove specific pollutants such as heavy metals, organic contaminants, etc.

It is considered of fundamental importance that biosolids reused for agricultural purposes must be those of better quality. Denmark has already legislated in this direction by placing a greater maximum amount for recovery in agriculture when the quality of biosolids is better. In addition, in Italy, more precisely in Lombardy, the legislation clearly distinguishes between "suitable" biosolids recovered in agriculture (which must comply with the limits of the current Italian law) and "high quality" biosolids that instead must meet more stringent limits. Furthermore, the adoption of regulations concerning the biosolids spreading methodology would make it possible to legislate in a sector that to date is not completely regulated and controlled.

7. Conclusions

In the last years, the main route for the reuse of biosolids is the application on agricultural land. This practice is controlled in different ways at the European level, due to the implementation in the Member States of Directive 86/278/CEE, which allows the reuse on land only for biosolids with a good quality. However, every country provides different requirements (with respect to Directive 86/278/CEE) for heavy metals, pathogens, and organic micropollutants both in biosolids and soils. Significant differences are also clear in other aspects of regulation (the maximum amount of biosolids spread on land, the soil where the use of biosolids is prohibited, the treatment requirements) in Member States where the reuse on land represents the main route of sludge management.

In order to comply with the European trend shown in this report, some actions, addressed to the stakeholders of biosolids land spreading activity (the WWTPs managers that produce biosolids and the farmers that recover the treated sludge), could improve the policy decisions making:

i. In order to follow the steps of Directive 2008/98/EC, the *minimization* from the producers (WWTPs managers) is the first step to control the quantitative of sludge.
ii. In addition to the enhancement of minimization, the *reuse of biosolids* must be improved in order to cope with nutrient depletion in soils (especially phosphorous).
iii. *Monitoring* all discharges is necessary. In fact, the lack of knowledge about the industrial discharges into the public sewer can significantly affect the wastewater treated and the sludge quality.

iv. Suitable *characterization*, especially in terms of heavy metals content and stabilization degree, of sludge deriving from WWTPs must be carried out, with the aim of sending the best quality sludge to agricultural land and reducing the environmental impacts.

v. Correct *planning* is very useful to give structure to the policy decisions at different levels (national, regional, etc.) in order to guarantee proper sludge management that provides for the safeguard of environment and public health.

Author Contributions: A.F. contributed to the drafting of the Section 1, Section 2, Section 3, while M.C.M. and S.P. contributed to the drafting of Sections 4 and 5, respectively. A.A. contributed to the drafting of the discussion and the conclusions. M.C.C. and V.T. and I.K. supervised the study and the paper drafting. All authors have read and approved the final manuscript.

Funding: This research received no external funding.

Acknowledgments: This work is dedicated to the Memory of Vincenzo Riganti.

Conflicts of Interest: The authors declare no conflict of interest.

Nomenclature

AOX	Absorbable organic halogens
CEC	Cation exchange capacity
CFU	Colony-forming unit
DEHP	Di(2-ethylhexyl) phthalates
DM	Dry matter
GW	Groundwater
LAS	Linear alkylbenzene sulfonates
NP	Nonylphenols
NPE	Nonylphenol ethoxylates
MPCN	Most probable cytopathic number
MPN	Most probable number
PAH	Polycyclic aromatic hydrocarbons
PCB	Polychlorinated biphenyls
PCDD	Polychlorinated dibenzo-p-dioxins
PCDF	Polychlorinated dibenzofuran
P.E.	Population equivalent
SOM	Soil organic matter
STP	Sludge treatment plant
TE	Toxic equivalency
TSS	Total suspended solids
UK	United Kingdom
VSS	Volatile suspended solids
WW	Wet weight
WRRF	Water resources recovery facility
WWTP	Wastewater treatment plant

References

1. Kelessidis, A.; Stasinakis, A.S. Comparative study of the methods used for treatment and final disposal of sewage sludge in European countries. *Waste Manag.* **2012**, *32*, 1186–1195. [CrossRef] [PubMed]
2. Eurostat. Sewage Sludge Production and Disposal from Urban Wastewater. Available online: https://ec.europa.eu/eurostat/web/environment/water (accessed on 6 September 2019).
3. Sun, Y.; Chen, Z.; Wu, G.; Wu, Q.; Zhang, F.; Niu, Z.; Hu, H.-Y. Characteristics of water quality of municipal wastewater treatment plants in China: Implications for resources utilization and management. *J. Clean. Prod.* **2016**, *131*, 1–9. [CrossRef]
4. Collivignarelli, M.C.; Abbà, A.; Carnevale Miino, M.; Damiani, S. Treatments for color removal from wastewater: State of the art. *J. Environ. Manag.* **2019**, *236*, 727–745. [CrossRef] [PubMed]

5. Collivignarelli, M.C.; Carnevale Miino, M.; Baldi, M.; Manzi, S.; Abbà, A.; Bertanza, G. Removal of non-ionic and anionic surfactants from real laundry wastewater by means of a full-scale treatment system. *Proc. Saf. Environ. Prot.* **2019**, *132*, 105–115. [CrossRef]
6. EUR-Lex Council Directive 91/271/EEC of 21 May 1991 concerning urban waste-water treatment. *Off. J. Eur. Communities* **1991**, *135*, 40–52.
7. EUR-Lex Commission Directive 98/15/EC of 27 February 1998 amending Council Directive 91/271/EEC with respect to certain requirements established in Annex I thereof. *Off. J. Eur. Communities* **1998**, *67*, 29–30.
8. Collivignarelli, M.C.; Abbà, A.; Carnevale Miino, M.; Torretta, V. What Advanced Treatments can be Used to Minimize the Production of Sewage Sludge in WWTPs? *Appl. Sci.* **2019**, *9*, 2650. [CrossRef]
9. EUR-Lex Directive EU/2018/851 of the European Parliament and of the Council of 30 May 2018 Amending Directive 2008/98/EC on Waste. *Off. J. Eur. Communities* **2018**, *150*, 109–140.
10. Liu, M.; Wang, C.; Bai, Y.; Xu, G. Effects of sintering temperature on the characteristics of lightweight aggregate made from sewage sludge and river sediment. *J. Alloys Compd.* **2018**, *748*, 522–527. [CrossRef]
11. Fytili, D.; Zabaniotou, A. Utilization of sewage sludge in EU application of old and new methods—A review. *Renew. Sustain. Energy Rev.* **2008**, *12*, 116–140. [CrossRef]
12. Inglezakis, V.J.; Zorpas, A.A.; Karagiannidis, A.; Samaras, P.; Voukkali, I.; Sklari, S. European Union legislation on sewage sludge management. *Fresenius Environ. Bull.* **2014**, *23*, 635–639.
13. Mininni, G.; Blanch, A.R.; Lucena, F.; Berselli, S. EU policy on sewage sludge utilization and perspectives on new approaches of sludge management. *Environ. Sci. Pollut. Res.* **2015**, *22*, 7361–7374. [CrossRef] [PubMed]
14. EPA. *Biennial Review of 40 CFR Part 503 As Required Under the Clean Water Act Section 405(d)(2)(C)—Reporting Period Biosolids Biennial Review 2015*; EPA: Washington, DC, USA, 2015.
15. EPA Basic Information about Biosolids. Available online: https://www.epa.gov/biosolids/basic-information-about-biosolids (accessed on 20 May 2019).
16. Collivignarelli, M.C.; Canato, M.; Abbà, A.; Carnevale Miino, M. Biosolids: What are the different types of reuse? *J. Clean. Prod.* **2019**, *238*, 117844. [CrossRef]
17. Alvarenga, P.; Mourinha, C.; Farto, M.; Santos, T.; Palma, P.; Sengo, J.; Morais, M.-C.; Cunha-Queda, C. Sewage sludge, compost and other representative organic wastes as agricultural soil amendments: Benefits versus limiting factors. *Waste Manag.* **2015**, *40*, 44–52. [CrossRef]
18. Aranda, V.; Macci, C.; Peruzzi, E.; Masciandaro, G. Biochemical activity and chemical-structural properties of soil organic matter after 17 years of amendments with olive-mill pomace co-compost. *J. Environ. Manag.* **2015**, *147*, 278–285. [CrossRef]
19. Castán, E.; Satti, P.; González-Polo, M.; Iglesias, M.C.; Mazzarino, M.J. Managing the value of composts as organic amendments and fertilizers in sandy soils. *Agric. Ecosyst. Environ.* **2016**, *224*, 29–38. [CrossRef]
20. Neczaj, E.; Grosser, A. Circular Economy in Wastewater Treatment Plant–Challenges and Barriers. *Proceedings* **2018**, *2*, 614. [CrossRef]
21. Jones, R.J.A.; Hiederer, R.; Rusco, E.; Loveland, P.J.; Montanarella, L. *The Map of Organic Carbon in Topsoils in Europe: Explanation of Special Publication Ispra 2004 No.72 (S.P.I.04.72)*; Office for Official Publications of the European Communities: Luxembourg, Luxembourg, 2004.
22. Lal, R. Restoring Soil Quality to Mitigate Soil Degradation. *Sustainability* **2015**, *7*, 5875–5895. [CrossRef]
23. European Commission. *Communication from the Commission to the European Parliament, the Council, the European Economic and Social Committee and the Committee of the Regions. A Policy Framework for Climate and Energy in the Period from 2020 to 2030*; EU Commission: Brussels, Belgium, 2014.
24. Ashekuzzaman, S.M.; Forrestal, P.; Richards, K.; Fenton, O. Dairy industry derived wastewater treatment sludge: Generation, type and characterization of nutrients and metals for agricultural reuse. *J. Clean. Prod.* **2019**, *230*, 1266–1275. [CrossRef]
25. Kacprzak, M.; Neczaj, E.; Fijałkowski, K.; Grobelak, A.; Grosser, A.; Worwag, M.; Rorat, A.; Brattebo, H.; Almås, Å.; Singh, B.R. Sewage sludge disposal strategies for sustainable development. *Environ. Res.* **2017**, *156*, 39–46. [CrossRef]
26. European Commission. Circular Economy-Implementation of the Circular Economy Action Plan. Available online: https://ec.europa.eu/environment/circular-economy/index_en.htm (accessed on 13 September 2019).
27. Sgroi, M.; Vagliasindi, F.G.A.; Roccaro, P. Feasibility, sustainability and circular economy concepts in water reuse. *Curr. Opin. Environ. Sci. Health* **2018**, *2*, 20–25. [CrossRef]

28. Case, S.D.C.; Jensen, L.S. Nitrogen and phosphorus release from organic wastes and suitability as bio-based fertilizers in a circular economy. *Environ. Technol.* **2019**, *40*, 701–715. [CrossRef]
29. Cornejo, P.K.; Becker, J.; Pagilla, K.; Mo, W.; Zhang, Q.; Mihelcic, J.R.; Chandran, K.; Sturm, B.; Yeh, D.; Rosso, D. Sustainability metrics for assessing water resource recovery facilities of the future. *Water Environ. Res.* **2019**, *91*, 45–53. [CrossRef] [PubMed]
30. Goh, C.H.; Short, M.D.; Bolan, N.S.; Saint, C.P. Biosolids: The Growing Potential for Use. In *Unmaking Waste in Production and Consumption: Towards the Circular Economy*; Emerald Publishing Limited: Bingley, UK, 2018; Chapter 4; pp. 67–88.
31. Peng, S.; Cui, H.; Ji, M. Research on the Sustainable Water Recycling System at Tianjin University's New Campus. In *Unmaking Waste in Production and Consumption: Towards the Circular Economy*; Emerald Publishing Limited: Bingley, UK, 2018; Chapter 17; pp. 295–307.
32. Clarke, B.O.; Smith, S.R. Review of 'emerging' organic contaminants in biosolids and assessment of international research priorities for the agricultural use of biosolids. *Environ. Int.* **2011**, *37*, 226–247. [CrossRef] [PubMed]
33. Islam, K.R.; Ahsan, S.; Barik, K.; Aksakal, E.L. Biosolid Impact on Heavy Metal Accumulation and Lability in Soiln Under Alternate-Year No-Till Corn–Soybean Rotation. *Water Air Soil Pollut.* **2013**, *224*, 1451. [CrossRef]
34. Manzetti, S.; van der Spoel, D. Impact of sludge deposition on biodiversity. *Ecotoxicology* **2015**, *24*, 1799–1814. [CrossRef] [PubMed]
35. Ashekuzzaman, S.M.; Kwapinska, M.; Leahy, J.J.; Richards, K.; Fenton, O. Novel Use of Dairy Processing Sludge Derived Pyrogenic Char (DPS-PC) to Remove Phosphorus in Discharge Effluents. In *Waste and Biomass Valorization*; Springer: Berlin, Germany, 2019. [CrossRef]
36. Lleó, T.; Albacete, E.; Barrena, R.; Font, X.; Artola, A.; Sánchez, A. Home and vermicomposting as sustainable options for biowaste management. *J. Clean. Prod.* **2013**, *47*, 70–76. [CrossRef]
37. EUR-Lex Council Directive 86/278/EEC of 12 June 1986 on the Protection of the Environment, and in Particular of the Soil, when Sewage Sludge is Used in Agriculture. *Off. J. Eur. Communities* **1986**, *181*, 6–12.
38. EUR-Lex Regulation (EC) No 219/2009 of the European Parliament and of the Council of 11 March 2009 adapting a number of instruments subject to the procedure referred to in Article 251 of the Treaty to Council Decision 1999/468/EC with regard to the regulatory pro. *Off. J. Eur. Communities* **2009**, *87*, 109–154.
39. EUR-Lex Decision (EU) 2018/853 of the European Parliament and of the Council of 30 May 2018 amending Regulation (EU) No 1257/2013 and Directives 94/63/EC and 2009/31/EC of the European Parliament and of the Council and Council Directives 86/278/EEC and 87/217/EEC. *Off. J. Eur. Communities* **2018**, *150*, 155–161.
40. Sorlini, S.; Collivignarelli, M.C.; Castagnola, F.; Crotti, B.M.; Raboni, M. Methodological approach for the optimization of drinking water treatment plants' operation: A case study. *Water Sci. Technol.* **2015**, *71*, 597–604. [CrossRef] [PubMed]
41. Torretta, V. Environmental and economic aspects of water kiosks: Case study of a medium-sized Italian town. *Waste Manag.* **2013**, *33*, 1057–1063. [CrossRef] [PubMed]
42. Foladori, P.; Andreottola, G.; Ziglio, G. *Sludge Reduction Technologies in Wastewater Treatment plants*; IWA Publishing: London, UK, 2010.
43. Coma, M.; Rovira, S.; Canals, J.; Colprim, J. Integrated side-stream reactor for biological nutrient removal and minimization of sludge production. *Water Sci. Technol.* **2015**, *71*, 1056–1064. [CrossRef] [PubMed]
44. Collivignarelli, M.C.; Castagnola, F.; Sordi, M.; Bertanza, G. Treatment of sewage sludge in a thermophilic membrane reactor (TMR) with alternate aeration cycles. *J. Environ. Manag.* **2015**, *162*, 132–138. [CrossRef] [PubMed]
45. Collivignarelli, M.C.; Abbà, A.; Bertanza, G. Why use a thermophilic aerobic membrane reactor for the treatment of industrial wastewater/liquid waste? *Environ. Technol.* **2015**, *36*, 2115–2124. [CrossRef] [PubMed]
46. Collivignarelli, M.C.; Abbà, A.; Castagnola, F.; Bertanza, G. Minimization of municipal sewage sludge by means of a thermophilic membrane bioreactor with intermittent aeration. *J. Clean. Prod.* **2017**, *143*, 369–376. [CrossRef]
47. Collivignarelli, M.C.; Castagnola, F.; Sordi, M.; Bertanza, G. Sewage sludge treatment in a thermophilic membrane reactor (TMR): Factors affecting foam formation. *Environ. Sci. Pollut. Res.* **2017**, *24*, 2316–2325. [CrossRef]
48. Eurostat. Population Connected to Urban Wastewater Collecting and Treatment Systems, by Treatment Level. Available online: https://ec.europa.eu/eurostat/web/environment/water (accessed on 20 October 2019).
49. European Commission. *Environmental, Economic and Social Impacts of the Use of Sewage Sludge on Land*; Final Report, Part III: Project Interim Reports; Milieu Ltd.: Brussels, Belgium, 2010.

50. European Commission. Disposal and Recycling Routes for Sewage Sludge Part 2—Regulatory Report. Available online: http://ec.europa.eu/environment/archives/waste/sludge/pdf/sludge_disposal2.pdf (accessed on 5 September 2019).
51. European Commission. *Working Document on Sludge, 3*; European Water Association: Brussels, Belgium, 2000.
52. Collivignarelli, M.C.; Abbà, A.; Bestetti, M.; Crotti, B.M.; Carnevale Miino, M. Electrolytic Recovery of Nickel and Copper from Acid Pickling Solutions Used to Treat Metal Surfaces. *Water Air Soil Pollut.* **2019**, *230*, 101. [CrossRef]
53. Collivignarelli, M.C.; Abbà, A.; Benigna, I. The reuse of biosolids on agricultural land: Critical issues and perspective. *Water Environ. Res.* **2019**. [CrossRef]
54. Walter, I.; Martínez, F.; Cala, V. Heavy metal speciation and phytotoxic effects of three representative sewage sludges for agricultural uses. *Environ. Pollut.* **2006**, *139*, 507–514. [CrossRef]
55. Singh, R.P.; Agrawal, M. Potential benefits and risks of land application of sewage sludge. *Waste Manag.* **2008**, *28*, 347–358. [CrossRef] [PubMed]
56. European Commission. Disposal and Recycling Routes for Sewage Sludge Part 1—Sludge Use Acceptance Report. Available online: https://ec.europa.eu/environment/archives/waste/sludge/pdf/sludge_disposal1.pdf (accessed on 5 September 2019).
57. Hudcová, H.; Vymazal, J.; Rozkošný, M. Present restrictions of sewage sludge application in agriculture within the European Union. *Soil Water Res.* **2019**, *14*, 104–120. [CrossRef]
58. Government of Germany. Sewage Sludge Regulation of 15 April 1992. Available online: https://www.bafg.de/Baggergut/DE/04_Richtlinien/klaerschlammverordnung.pdf?__blob=publicationFile (accessed on 5 September 2019). (In German)
59. Government of Germany. Ordinance on the Reorganization of Sewage Sludge Utilization. Available online: https://www.bgbl.de/xaver/bgbl/start.xav?startbk=Bundesanzeiger_BGBl&start=%2F%2F%2A%5B%40attr_id=%27bgbl117s3465.pdf%27%5D#__bgbl__%2F%2F*%5B%40attr_id%3D%27bgbl117s3465.pdf%27%5D__1562866673861 (accessed on 5 September 2019). (In German)
60. Government of UK. Guidance of 23 May 2018: "Sewage Sludge on Farmland: Code of Practice for England, Wales and Northern Ireland". Available online: https://www.gov.uk/government/publications/sewage-sludge-in-agriculture-code-of-practice/sewage-sludge-in-agriculture-code-of-practice-for-england-wales-and-northern-ireland (accessed on 5 September 2019).
61. Government of Spain. Royal Decree 1310/1990, of 29 October, Regulating the Use of Sludge from Depuration in the Agricultural Sector. Available online: https://www.boe.es/diario_boe/txt.php?id=BOE-A-1990-26490 (accessed on 6 September 2019). (In Spanish)
62. Government of France. Order of 8 January 1998 Laying Down the Technical Requirements for Sludge Spreading on Agricultural Soils under Decree No. 97-1133 of 8 December 1997 on the Spreading of Sludge from Waste Water Treatment. Available online: https://www.legifrance.gouv.fr/affichTexte.do?cidTexte=JORFTEXT000000570287 (accessed on 6 September 2019). (In French)
63. Government of Italy. Conversion into Law, with Amendments, of the Decree-Law 28 September 2018, n. 109, Containing Urgent Provisions for the City of Genoa, the Security of the National Infrastructure and Transport Network, the 2016 and 2017 Seismic Events, Work and other Emer. Available online: https://www.gazzettaufficiale.it/eli/id/2018/11/19/18G00157/sg (accessed on 5 September 2019).
64. Government of Italy. Implementation of Directive 86/278/EEC on the Protection of the Environment, and in Particular of the Soil, when Sewage Sludge is Used in Agriculture. Available online: https://www.gazzettaufficiale.it/eli/id/1992/02/15/092G0139/sg (accessed on 4 September 2019). (In Italian)
65. Government of Netherlands. Decision of 30 January 1998 Laying Down Rules on the Quality and the Placing on the Market of other Organic Fertilizers (Decision Quality and Use of other Organic Fertilizers). Available online: https://wetten.overheid.nl/BWBR0009360/2006-01-01 (accessed on 6 September 2019). (In Dutch)
66. Lander of Carinthia Ordination n.74 of 15/12/2000 on the Application of Treated Sewage Sludge, Biowaste and Green Waste to Land-Based Soil—Carinthia Sewage Sludge and Compost Regulation. Available online: https://www.ris.bka.gv.at/GeltendeFassung.wxe?Abfrage=LrK&Gesetzesnummer=20000166 (accessed on 4 September 2019). (In German)
67. Lander of Lower Austria. Consolidated Landrecht Lower Austria: Entire legislation for Sewage Sludge Ordinance, Version of 11.07.2019. Available online: https://www.ris.bka.gv.at/GeltendeFassung.wxe?Abfrage=LrNO&Gesetzesnummer=20001009 (accessed on 3 September 2019). (In German)

68. Lander of Styria. Ordinance of the Steiemark Provincial Government of 8 October 2007, on the Application of Sewage Sludge to Agricultural Soils. Available online: https://www.ris.bka.gv.at/GeltendeFassung.wxe?Abfrage=LrStmk&Gesetzesnummer=20000867 (accessed on 2 September 2019). (In German)
69. Lander of Voralberg. Ordinance of the National Government on the Application of Sewage Sludge Published on 9 October 1997. Available online: https://www.ris.bka.gv.at/Dokumente/Lgbl/LGBL_VO_19971009_75/LGBL_VO_19971009_75.pdf (accessed on 7 September 2019). (In German)
70. Government of Sweden. Notice of Protection of the Environment, in Particular the Soil, When Sewage Sludge Is Used in Agriculture; Decided on 30 May 1994. Available online: https://www.naturvardsverket.se/Documents/foreskrifter/nfs1994/snfs1994-02k.pdf (accessed on 5 September 2019). (In Swedish)
71. Government of Portugal. Law Decree No. 276/2009 of 2 October on the Agricultural Use of Sewage Sludge. Available online: https://dre.pt/pesquisa/-/search/490974/details/maximized (accessed on 6 September 2019). (In Portuguese)
72. Government of Finland. Council of State Decision on Use of Sewage Sludge in Agriculture of December 5, 1994. 1994. Available online: https://www.finlex.fi/fi/laki/alkup/1994/19940282 (accessed on 7 September 2019). (In Finnish)
73. Government of Denmark. Executive Order no. 49 of 20 January 2000 on Supervision of Waste Water Sludge for Agricultural Purposes. Available online: https://www.retsinformation.dk/Forms/R0710.aspx?id=6919 (accessed on 11 September 2019). (In Danish)
74. Government of Denmark. Order no. 1001 of 27/06/2018 on the Use of Waste for Agricultural Purposes. Available online: https://www.retsinformation.dk/Forms/R0710.aspx?id=202047 (accessed on 11 September 2019). (In Danish)
75. Government of Ireland. S.I. No. 148/1998—Waste Management (Use of Sewage Sludge in Agriculture) Regulations. 1998. Available online: http://www.irishstatutebook.ie/eli/1998/si/148/made/en/print (accessed on 11 September 2019).
76. Government of Greece. Methods, Conditions and Restrictions for the Use in Agriculture of Sludge from Domestic and Urban Effluent Treatment. Ministerial Decision: 80568/4225/91. Available online: http://www.elinyae.gr/el/lib_file_upload/641b_91.1149837816400.pdf (accessed on 11 September 2019). (In Greek)
77. Walloon Region. Order of the Walloon Government of 12 January 1995 Regulating the Use of Sewage Sludge or Sludge from Sludge Treatment Centers in Septic Tanks. Available online: http://environnement.wallonie.be/legis/solsoussol/sol002.htm (accessed on 11 September 2019). (In French).
78. Flemish Region. Decision of the Flemish Government Establishing the Flemish Regulation Concerning the Sustainable Management of Material Cycles and Waste. Available online: https://navigator.emis.vito.be/mijn-navigator?woId=43992 (accessed on 11 September 2019). (In Dutch)
79. Government of Luxembourg. Grand-Ducal Regulation of 23 December 2014 Relating to Sewage Sludge. Available online: http://data.legilux.public.lu/file/eli-etat-leg-memorial-2015-2-fr-pdf.pdf (accessed on 3 September 2019). (In French)
80. Government of Poland. Ordinance of the Minister of the Environment of 25 February 2015 on Municipal Sewage Sludge. Available online: http://prawo.sejm.gov.pl/isap.nsf/download.xsp/WDU20150000257/O/D20150257.pdf (accessed on 3 September 2019). (In Polish)
81. Government of Hungary. Government Decree on the Rules of Agricultural Use and Treatment of Sewage and Sewage Sludge. 50/2001 (IV.3). Available online: https://net.jogtar.hu/jogszabaly?docid=A0100050.KOR (accessed on 2 September 2019). (In Hungarian)
82. Government of the Czech Republic. Decree of the Ministry of the Environment No. 382/2001, on the Conditions of Use of Treated Sludge on Agricultural Land. Available online: https://www.zakonyprolidi.cz/cs/2001-382 (accessed on 2 September 2019). (In Czech)
83. Government of the Czech Republic. Declaration 437/2016 on the Conditions of Use of Treated sludge on Agricultural Land and Amending Decree No. 383/2001. Available online: https://www.zakonyprolidi.cz/cs/2016-437 (accessed on 2 September 2019). (In Czech)
84. Government of Romania. Ordinance No. 344/708 of 16 August 2004 for Approval of Technical Norms on Environmental Protection and Especially of Soils, when Using Sewage Sludge in Agriculture. Available online: http://www.adideseuribn.ro/legislatie/Ordin344_2004.pdf (accessed on 3 September 2019). (In Romanian)

85. Government of Lithuania. Declaration n.349 Concerning the Approval of the Normative Document Land 20-2005 "Requirements for the Use of Waste Dumping and Recycling". Available online: https://e-seimas.lrs.lt/portal/legalAct/lt/TAD/TAIS.143603/WdiYOWmTAy (accessed on 3 September 2019). (In Lithuanian)
86. Government of Slovakia. Act no. 188/2003 on Application of Sludge and Bottom Sediments to Soil and on Amendment of Act no. 223/2001 on Waste and Amendments to Certain Acts as Amended. Available online: https://www.zakonypreludi.sk/zz/2003-188 (accessed on 10 September 2019). (In Slovak)
87. Government of Bulgaria. Ordinance on the Procedure and the Way of Utilization of Sludge from the Treatment of Waste Waters through Their Use in Agriculture. Adopted by Decree of the Council of Ministers No 201 of 4.08.2016, Promulgated, SG, no. 63 of 12.08.2016. Available online: http://eea.government.bg/bg/legislation/waste/Naredba_utaiki_26.10.16.pdf. (accessed on 10 September 2019). (In Bulgarian)
88. Government of Estonia. Regulations No. 78 of 30 December 2002, Entry into Force on 01/01/2016, on Requirements for Use of Sewage Sludge in Agriculture, Greenery and Re-cultivation. Available online: https://www.riigiteataja.ee/akt/761407 (accessed on 10 September 2019). (In Estonian)
89. Government of Cyprus. Control of Water Pollution (Use of Sludge) Regulations of 2002, PI. 517/2002. Available online: http://www.cylaw.org/KDP/data/2002_1_517.pdf (accessed on 2 September 2019). (In Greek)
90. Government of Latvia. Cabinet of Ministers Regulation No. 362 of 2 May 2006 on Regulations on the Use of Sewage Sludge and Its Compost, Monitoring and Control. Available online: https://likumi.lv/doc.php?id=134653 (accessed on 8 September 2019). (In Latvian)
91. Government of Slovenia. Decree No. 62 of 20 June 2008 on the Use of Sludge from Municipal Wastewater Treatment Plants in Agriculture. Available online: http://www.pisrs.si/Pis.web/pregledPredpisa?id=URED4880 (accessed on 7 September 2019). (In Slovenian)
92. Government of Malta. Legal Notice 212 of 28 June 2002 on Sludge (Use in Agriculture) Regulations. Available online: http://www.justiceservices.gov.mt/DownloadDocument.aspx?app=lom&itemid=11496&l=1 (accessed on 8 September 2019).
93. Government of Croatia. Ordinance No. 38 of 2 April 2008 on Sludge Management from Sewage Treatment Plants when Sludge is Used in Agriculture. Available online: http://www.poslovni-savjetnik.com/propisi/pravilnik-o-gospodarenju-muljem-iz-uredaja-za-prociscavanje-otpadnih-voda-kada-se-mulj (accessed on 2 September 2019). (In Croatian)
94. PURE. Good Practices in Sludge Management. Available online: http://www.purebalticsea.eu/index.php/gpsm:good_practices (accessed on 9 September 2019).
95. Province of Pavia. The Soil of the Province of Pavia. Available online: https://www.provincia.pv.it/attachments/article/1690/Il%20suolo%20della%20Provincia%20di%20Pavia.pdf (accessed on 17 October 2019). (In Italian)
96. Chang, A.C.; Hinesly, T.D.; Bates, T.E.; Poner, H.E.; Dowdy, R.H.; Ryan, J.A. Effects of Long-Term Sludge Application on Accumulation of Trace Elements By Crops. In *Land Application of Sludge*; CRC Press: Boca Raton, FL, USA, 2018; pp. 53–66.
97. Somtners, L.; Van Volk, V.; Giordano, P.M.; Sopper, W.E.; Bastían, R. Effects of Soil Properties on Accumulation of Trace Elements By Crops. In *Land Application of Sludge*; CRC Press: Boca Raton, FL, USA, 2018; pp. 5–24.
98. EUR-Lex Council Directive 91/676/EEC of 12 December 1991 concerning the protection of waters against pollution caused by nitrates from agricultural sources. *Off. J. Eur. Communities* **1991**, *375*, 1–8.
99. EUR-Lex Regulation (EC) No 1882/2003 of the European Parliament and of the Council of 29 September 2003 adapting to Council Decision 1999/468/EC the provisions relating to committees which assist the Commission in the exercise of its implementing powers laid down. *Off. J. Eur. Communities* **2003**, *284*, 1–53.
100. EUR-Lex Regulation (EC) No 1137/2008 of the European Parliament and of the Council of 22 October 2008 adapting a number of instruments subject to the procedure laid down in Article 251 of the Treaty to Council Decision 1999/468/EC, with regard to the regulatory p. *Off. J. Eur. Communities* **2008**, *311*, 1–54.
101. Napoli, M.; Marta, A.D.; Zanchi, C.A.; Orlandini, S. Assessment of soil and nutrient losses by runoff under different soil management practices in an Italian hilly vineyard. *Soil Tillage Res.* **2017**, *168*, 71–80. [CrossRef]
102. Pardini, G.; Gispert, M.; Dunjo, G. Runoff erosion and nutrient depletion in five Mediterranean soils of NE Spain under different land use. *Sci. Total Environ.* **2003**, *309*, 213–224. [CrossRef]
103. Torri, S.I.; CorrÊa, R.S.; Renella, G. Biosolid Application to Agricultural Land—A Contribution to Global Phosphorus Recycle: A Review. *Pedosphere* **2017**, *27*, 1–16. [CrossRef]

104. Evans, T.D. Biosolids in Europe. In Proceedings of the 26th WEF Residuals & Biosolids Conference, Raleigh, NC, USA, 25–28 March 2012; pp. 1–10.
105. Rashid, M.M.; Kattou'a, M.G.; Al-Khatib, I.A.; Sato, C. Farmers' attitude toward treated sludge use in the villages of West Bank, Palestine. *Environ. Monit. Assess.* **2017**, *189*, 353. [CrossRef]
106. Amajirionwu, M.; Connaughton, N.; McCann, B.; Moles, R.; Bartlett, J.; O'Regan, B. Indicators for managing biosolids in Ireland. *J. Environ. Manag.* **2008**, *88*, 1361–1372. [CrossRef]
107. Schulte, R.P.O.; Creamer, R.E.; Donnellan, T.; Farrelly, N.; Fealy, R.; O'Donoghue, C.; O'hUallachain, D. Functional land management: A framework for managing soil-based ecosystem services for the sustainable intensification of agriculture. *Environ. Sci. Policy* **2014**, *38*, 45–58. [CrossRef]
108. Mininni, G.; Sagnotti, G. Sludge production and utilization in Italy. In Proceedings of the Wastewater and Biosolids Treatment and Reuse: Bridging Modeling and Experimental Studies, Otranto, Italy, 8–14 June 2014.
109. Lombardy Region. Regional Decree n. 6665 of 14 May 2019. Available online: https://www.regione.lombardia.it/wps/wcm/connect/19df87f4-0ac9-4fac-b171-084e37a5bc51/D.d.u.o.+6665_2019.pdf?MOD=AJPERES&CACHEID=ROOTWORKSPACE-19df87f4-0ac9-4fac-b171-084e37a5bc51-mH4MG8K (accessed on 9 September 2019). (In Italian)
110. Veneto Region. Law n. 130 of 11/15/2018, Article 41: Urgent Provisions on the Management of Sewage Sludge that Integrate the Characteristics of the Sludge that can be Used in Agriculture. Available online: http://www.arpa.veneto.it/temi-ambientali/suolo/file-e-allegati/normativa/fanghi-di-depurazione-utilizzo-in-agricoltura/Legge130-2018art_41.pdf/view (accessed on 4 September 2019). (In Italian)
111. Emilia-Romagna Region. Deliberation of the Regional Council 4 March 2019, n. 326: Urgent Provisions on the Agronomic Use of Sewage Sludge. Available online: http://bur.regione.emilia-romagna.it/dettaglio-inserzione?i=b5f9fa6aa4d146618e11832370e056e7 (accessed on 5 September 2019). (In Italian)
112. Collivignarelli, M.C.; Abbà, A.; Padovani, S.; Frascarolo, M.; Sciunnach, D.; Turconi, M.; Orlando, M. Recovery of sewage sludge on agricultural land in Lombardy: Current issues and regulatory scenarios. *Environ. Eng. Manag. J.* **2015**, *14*, 1477–1486.
113. Lombardy Region. Regional Decree n. X/2031/2014, Regional Regulations for the Treatment and Use, for the Benefits of Agriculture, of Sewage Sludge from Civil and Industrial Wastewater Treatment Plants. Available online: https://www.regione.lombardia.it/wps/wcm/connect/e81d0f54-bedf-4918-886f-ad4db4c04911/D.g.r.+2031_2014.pdf?MOD=AJPERES&CACHEID=ROOTWORKSPACE-e81d0f54-bedf-4918-886f-ad4db4c04911-mH4K-bC (accessed on 9 September 2019). (In Italian)
114. Veneto Region. Deliberation of the Regional n. 2241/2005, Directive B—Technical Regulations Regarding the Use in Agriculture of Sewage Sludge and other Sludge Non-toxic and Harmful, with Agronomic Usefulness. Available online: http://www.arpa.veneto.it/temi-ambientali/suolo/file-e-allegati/normativa/fanghi-di-depurazione-utilizzo-in-agricoltura/DGRV2241_05_D49.pdf (accessed on 4 September 2019). (In Italian)
115. Emilia-Romagna Region. Deliberation of the Regional n. 285/2005, Regional Regulations Concerning the Management and the Authorization for Sewage Sludge Use in Agriculture. Available online: http://ambiente.regione.emilia-romagna.it/it/acque/approfondimenti/normativa/DGR_285_2005.pdf (accessed on 5 September 2019). (In Italian)
116. Government of Italy. Legislative Decree n. 75 of April 29, 2010—Reorganization and Revision of the Regulation on Fertilizers, Pursuant to Article 13 of Law no. 88. of 7 July 2009. Available online: https://www.gazzettaufficiale.it/eli/id/2010/05/26/010G0096/sg (accessed on 13 September 2019). (In Italian)
117. Government of Italy. Ministerial Decree of June 28, 2016—Amendments to Annexes 1, 2, 3, 6 and 7 of Legislative Decree No. 75 of April 29, 2010 Containing: Reorganization and Revision of the Discipline on Fertilizers, in Accordance with Article 13 of the Law n. 88 of 7 July. Available online: https://www.gazzettaufficiale.it/atto/serie_generale/caricaDettaglioAtto/originario?atto.dataPubblicazioneGazzetta=2016-08-12&atto.codiceRedazionale=16A05930&elenco30giorni=false (accessed on 13 September 2019).