

Review

Land Application of Biosolids in Europe: Possibilities, Con-Straints and Future Perspectives

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Abstract: The agricultural use of good quality sludge represents a value-added route to ensure growth sustainability in Europe, where raw material availability, for example, for phosphorus, is insufficient to meet demand. However, the possible presence of pathogens, pharmaceuticals and heavy metals requires specific regulations to minimize sludge-related health issues and environmental risks. The current regulation on sludge agricultural use applied by many EU countries is here presented and compared, highlighting scarce harmonization of the legislative framework among Member States. Actual issues, such as the fate of emerging micropollutants and microplastics in sludge-amended soils, and public health concerns regarding sludge spreading during the COVID-19 epidemic, are considered, too.

Keywords: sewage sludge use; EU legislative framework; heavy metals; organic micropollutants; P-recovery; microplastics; pathogens



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1. Introduction

Sewage sludge chemical composition may vary depending on the wastewater source and treatment processes. Regarding nutrients, organic nitrogen and inorganic phosphorus represent the majority of total nutrient content in sludge [1]. The average content of total N, total P and total K in digested sludge is reported to be around 4.7% dw, 2.3% dw and 0.3% dw, respectively [2]. The agricultural benefit and environmental acceptability of sludge utilization in agriculture is well documented [2–4]. Therefore, sewage sludge could represent a renewable source of phosphorus, since white phosphorus (P₄) and phosphate rock are included among the 20 critical raw materials (CRM) for the EU, as reported in the “Report on critical raw materials for the EU” released by the European Commission in 2017 [5]. Recently, P₄ and phosphate rock were maintained on the 3 September 2020 update of the CRM list. However, Europe has no P₄ production and is 100% dependent on imports. This critical dependency could be addressed by up-cycling P₄ from wastes, in particular sewage sludge incineration ash.

Global food security for a growing population will depend on finding new sources of phosphorous to improve crop yields. In the framework of the new Green Deal, the EU’s “Farm-to-Fork” policy poses ambitious objectives for agro-food system sustainability, including a 50% nutrient losses reduction and a 20% fertilizer use reduction before 2030, to improve nutrient stewardship (revised Circular Economy Action Plan and European Integrated Nutrient Management Action Plan).

Hence, sludge utilization could be an opportunity for the industries across the chain, from water to agriculture, to minimize the reliance on chemicals. Nutrients are essential for plant growth. However, when applied excessively, they could be leached and transported to surface and/or ground water posing environmental risks [6,7]. The main risks related to agricultural use of sewage sludge are the potential presence of pathogens and pollutant enrichment in soils, plants and animal pastures and the subsequent entry into the food

chain [8,9]. Heavy metals, pathogens and organic pollutants can also affect soil functioning and biodiversity [10,11]. According to literature, low application doses of sludge did not cause a significant increase in heavy metal concentrations in soils [1]. On the contrary, low metal sludge has beneficial effects on microbial biomass, organic carbon and on soil microbial activity [12]. Excessive application of sewage sludge to soil has been found to increase the bioavailability of heavy metals that have a negative effect on soil [1,12].

Sewage sludge may contain a wide spectrum of harmful toxic micropollutants such as polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs), di(2-ethylhexyl) phthalate (DEHP), personal care products, detergent residues, endogenous hormones, pharmaceuticals, synthetic steroids, polychlorinated dibenzo-p-dioxins and dibenzo-p-furans (PCDD/Fs), and others [13]. Moreover, recent studies evidenced the accumulation of microplastics in agricultural soils after sludge [14]. One of the major concerns regarding sludge agricultural use is the possible health risks associated with this practice. It clearly appears therefore that the actions taken by the European Commission for sustaining the sludge agricultural use should be principally addressed to establish a set of appropriate standards that will guarantee against any possible risks to the humans, the soil and the environment.

The European Commission has released in December 2015 a Communication to the European Parliament and the Council called “closing the loop—An EU action plan for the circular economy” [15]. In this action plan, almost all economy sectors and related waste are included but not sewage sludge. In fact, the new Directives of 2018 included in the Circular Economy Package (Directives 849, 850, 851 and 852) do not plan a revision of the old sludge directive 86/278. According to the Commission, this directive does not require any update. In the document “ex post evaluation of certain waste stream directives” [16], the sludge directive was considered to be effective, efficient, relevant, and coherent with other EU legislation. However, looking more in-depth, this evaluation highlights that sludge is used in agriculture in nearly 50% of the cases.

Other recycling alternatives, which include production of fertilizing products (struvite, biochar, ashes) are to date still scarcely applied, although some processes are already running on full-scale in EU plants and new technologies specifically addressed to phosphorus recovery were patented and experienced on pilot scale. Many research projects were funded on Horizon 2020 including recovery of bio-plastics, (polyhydroxyalkanoates—PHA), from liquor after fermentation of sewage sludge. Some legislative barriers still exist regarding legal status of these materials, which are often included in the framework of wastes.

The European Commission revised the EU Fertilizer Regulation, expanding its scope to secondary-raw-material-based fertilizing products, and resulting in the publication of the new EU Fertilizing Products Regulation (EU 2019/1009). Such new regulation [17] sets the legal framework for the manufacturing and placing on the market of specific fertilizing products derived from bio-wastes and other secondary raw materials. Sewage sludge is excluded from the production of EU market organic fertilizers, with the exception of the sludge-derived material defined with the acronym STRUBIAS (Struvite, Biochar, Ashes). Sludge is also not considered among waste flows for fuel application at the EU level [18]. The European Commission has recently opened, from 25 August 2020, a public consultation to be completed between 20 November 2020 and 5 March 2021, for re-evaluation of the EU Sewage Sludge Directive (86/278). The roadmap underlines that the directive should encourage the safe use of sludge in agriculture, being that P recovery is a core objective and considering health risks due to the emerging concern about the contaminants (pharmaceuticals, PAH and PFAS, cosmetics and microplastics).

On the other hand, problems and issues related to the quality of sludge used in agriculture have been raised since the publication of the old sludge directive. New organic pollutants have been put on the market and their release into the wastewaters, for microplastics, perfluorinated compounds, and flame-retardants, requires attention. To fill the European legislative gap, some Member States (MS) have provided national regulations

fixing stringent limits for sludge use. This results in a fragmented legislative framework in which sludge is still qualified as waste and coherently managed.

In this paper, different MS regulations on sludge management, with specific regard to agricultural use, are presented and compared. Limits on heavy metals in sludge and soil, new limits on organic micropollutants, and additional requirements on pathogens and pathogens indicators will be discussed. Emerging issues regarding P-recovery technologies, the fate of microplastics (MPs) in sludge-amended soils, and the current health concerns related to sludge application during the COVID-19 pandemic will be described too.

2. Limits for Pollutants and Pathogens in Sludge

Land application of sludge poses a source of concern about the long-term accumulation of toxic elements in soil and their potential uptake by crops. However, sludge utilization in agriculture is currently regulated only by the limits of heavy metals specified in Council Directive 86/278/EEC [19], which therefore appears not to address the new risks on vegetation, animals and humans, posed by the presence of new contaminants in domestic wastewater and consequently in sewage sludge. The directive [19] sets the limits for concentrations of heavy metals both in sludge and in sludge-treated soil (Table 1).

Table 1. Limits of heavy metals for sludge use in agriculture.

Directive 86/278/EEC	Cd	Cu	Hg	Ni	Pb	Zn
Sludge (mg/kg dw)	20–40	1000–1750	16–25	300–400	750–1200	2500–4000
Sludge-treated soil (mg/kg dw of soil) ($6 < \text{pH}_{\text{soil}} < 7$)	1–3	50–140	1–1.5	30–75	50–300	150–300

According to [19], sludge must be applied on land with specific regard to the nutritional needs of crops and its use must not impair the soil, surface water, and groundwater quality. MS must therefore manage the sludge utilization in order to comply with the limit values specified in Table 1, regarding heavy metals in sludge and soil. The required sludge application rates also depend on the sludge fertilizer potential and soil properties. Nevertheless, the legislation framework resulted in too much fragmented legislation without any effective harmonization among the Member States. This may be considered as a prejudice to the principle of competitiveness and free competition.

All MS have transposed different limits into their own legislation causing high fragmentation of the legislation framework. The limits for Cd, Cu, Ni, Pb and the limits for Hg, Cr, Zn, and As adopted by the European Countries, are presented in Figures 1 and 2. As a result, most of the MS adopted more stringent limits with respect to European Directive 86/278/EEC. Nevertheless, in Austria the regulation differs among the nine Regional States (“Länder”) [20]. The Burgenland region based the limits on the application rate of the maximum allowable annual pollutant load: Class I from 4.17 t dw/(ha × year) (at limited concentrations of Cu or Ni in sludge) to 12.5 t dw/(ha × year) (at limited concentrations of Cr, Pb, Cd and Hg in sludge), Class II 2.5 t dw/(ha × year). Carinthia subdivided the sludge into classes: Class I: 10 t dw/(ha × 2 year), Class A: 8 t dw/(ha × 2 year), Class AB: 6 t dw/(ha × 2 year), Class B: 4.8 t dw/(ha × 2 year) (exceeding the limit values for one parameter at a maximum of 25% is permissible).

In Lower Austria, Class I sludge must not exceed the regional average value in sludge for Zn, Cu, Cr, Pb, Cd, Hg and AOX in the upper soil layer (depth of 25 cm for arable soil, 10 cm for pastures). At the same time, they must not exceed the value of Class II for these indicators. In the Vienna region, the application of sludge is prohibited with the exception of hygienically safe fertilizers containing treated sludge, compost and soil. In the Salzburg region, the application of sludge is prohibited or restricted to designated soils. In the Tyrol region, the application of sludge and other sludge-derived products is prohibited.

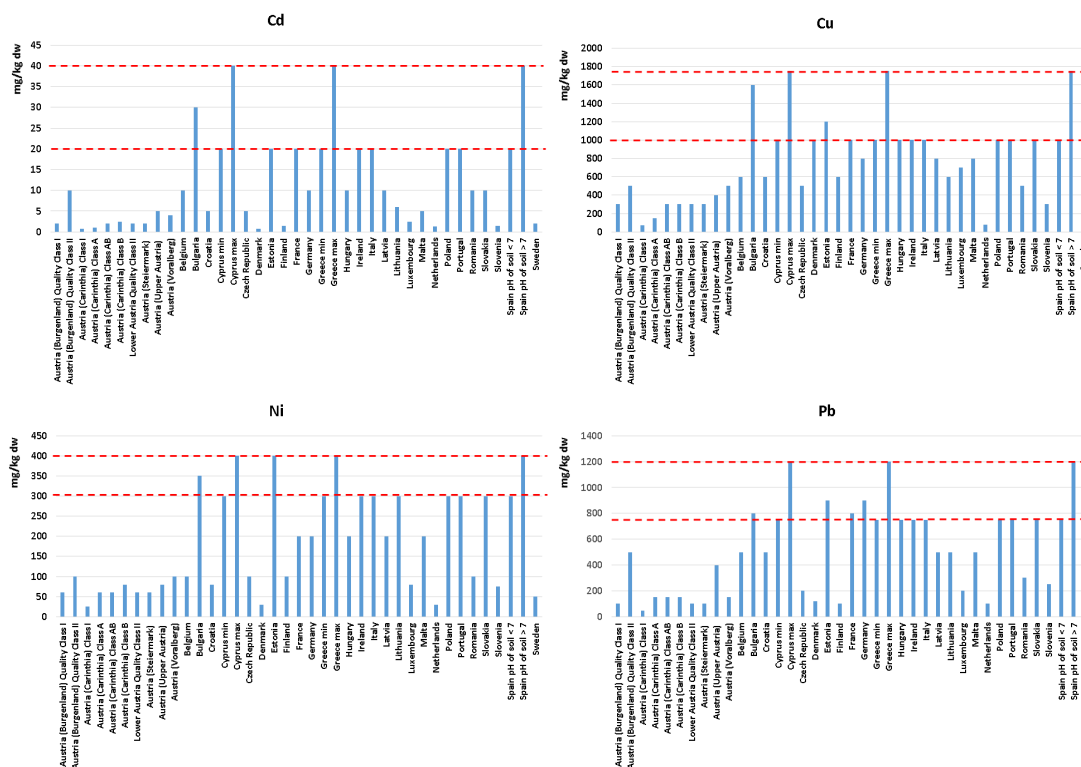


Figure 1. Limits of Cd, Cu, Ni, and Pb for sludge utilization in agriculture (mg/kg dw). Dotted lines: limits proposed by Directive 86/278/EEC.

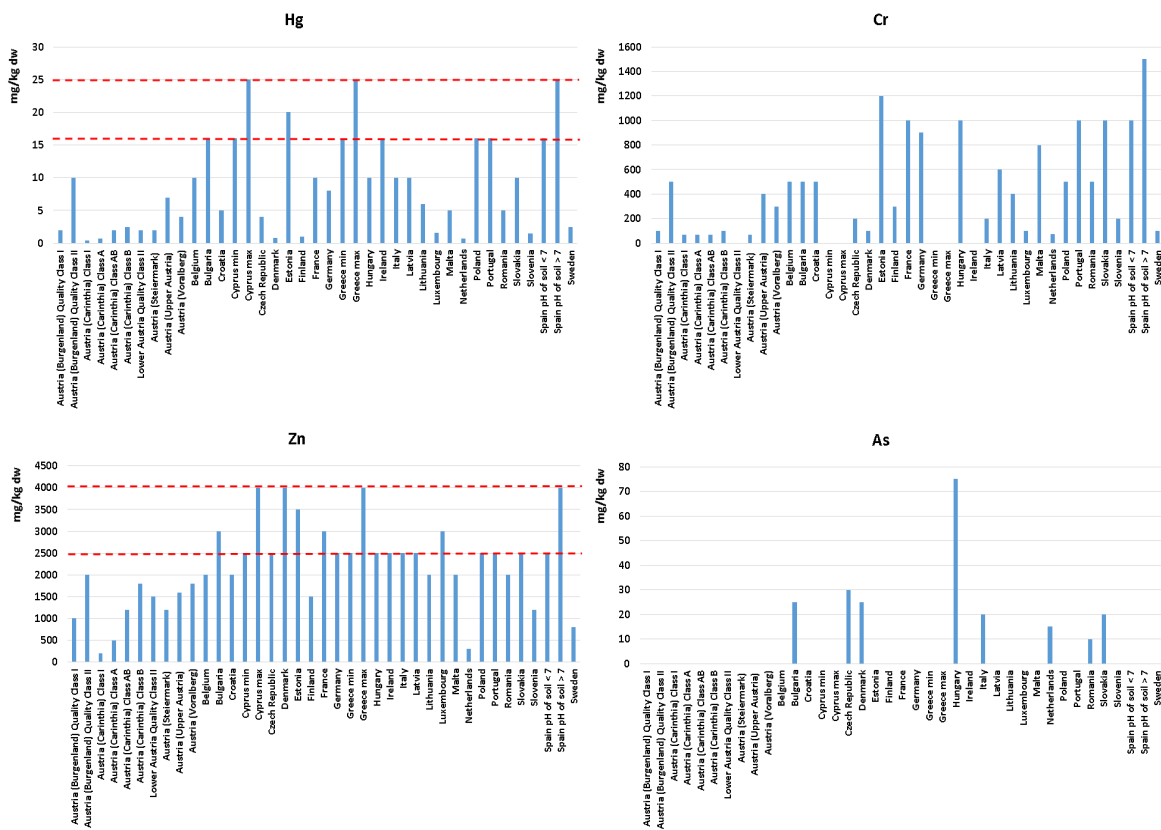


Figure 2. Limits of Hg, Cr, Zn, and As for sludge utilization in agriculture (mg/kg dw). Dotted lines: limits proposed by directive 86/278/EEC.

Figures 1 and 2 show that 18 out of 27 states fixed more stringent limits for cadmium, 14 out of 27 for both copper and lead, 19 out of 27 for mercury, 16 out of 27 for nickel, 10 out of 27 for zinc. Therefore, the majority of EU countries adopted more stringent limits for mercury and cadmium than identified in the Directive 86/278/EEC.

The United Kingdom was not included in the assessment, since it adopted a different approach. The use of sewage sludge in the UK, in fact, is regulated by the maximum allowable contents of Cd, Cu, Hg, Ni, Pb, and Zn in soil intended for sludge application [20].

Moreover, several MS introduced limit values for other elements. Limits for arsenic (up to 75 mg/kg dw) and chromium (up to 1500 mg/kg dw) were adopted by 8 and 23 countries, respectively (Figure 2). Only Romania and Hungary adopted a limit for molybdenum (50 mg/kg dw), while Hungary was the only MS with limits for cobalt and selenium (20 and 100 mg/kg dw, respectively). Italy recently introduced (law No. 130/2018) the limit for selenium (10 mg/kg dw) and for beryllium (2 mg/kg dw). With the exception of Hungary and Italy, that set a limit value of 1 and 2 mg/kg dw, respectively, for hexavalent chromium (Cr VI); the other EU countries have not set any limit for heavy metal ions, although more reactive and toxic to plants [20].

Limits of Cd, Cr, Cu, Hg, Ni, Pb and Zn in sludge-treated soil (mg/kg dw of soil) in EU member states are presented in Figure S1. Minimum and maximum limits of heavy metals in sludge-treated soils are dependent by differences in regulations among regional areas, or by the composition and the pH value of the soil.

The European Commission released in 2000 a draft-working document on sludge [21], including some regulations to manage sludge use on land. In particular, the working document also indicated, for the first time, limits for different classes of organic micropollutants, together with hygienization requirements and appropriate sludge treatments. In particular, the document indicated that—in order to be used without restrictions—sludge should undergo hygienization by an advanced treatment able to achieve at least a 6 log reduction in *Escherichia coli*, and produce sludge complying with the following limits: absence of salmonella in 50 g ww, and *E. coli* < 500 cfu/g dw. It is also proposed that the use of sludge produced by conventional treatments, able to reduce 2 log of *E. coli*, is allowed with restrictions on their application time, site and modality. In any case, the document was not legally binding.

The limits adopted by national legislations of the EU countries regarding pathogens or indicators (i.e., *Clostridium perfringens*, Enterococci, Helminth eggs and Thermotolerant Coliform Bacteria) in sludge to be applied on land are reported in Table 2.

Table 2. Limits for pathogens in sludge utilization in agriculture [20].

State	Salmonella sp.	Other Pathogens ¹
Bulgaria	no occurrence in 20 g	<i>Escherichia coli</i> < 100 MPN/g Helminths eggs and larvae, 1 unit/kg dw <i>Clostridium perfringens</i> < 300 MPN/g
Czech Republic	no occurrence in 50 g	<i>Escherichia coli</i> or Enterococci < 10 ³ cfu/g (4 samples from 5) < 5 × 10 ³ cfu/g (1 sample from 5)
Denmark	no occurrence	Faecal streptococci < 100/g
Finland	no occurrence in 25 g	<i>Escherichia coli</i> < 1000 cfu, < 100 cfu in greenhouse cultivation
France	8 MPN/10 g dw	Enterovirus < 3 MPCN/10 g dw Helminths eggs < 3/10 g dw

Table 2. Cont.

State	Salmonella sp.	Other Pathogens ¹
Italy	1000 MPN/g dw	
Lithuania	-	<i>Escherichia coli</i> ≤ 1000 cfu/g <i>Clostridium perfringens</i> ≤ 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	<i>Escherichia coli</i> < 1000 cfu/g
Austria (Carinthia)	no occurrence in 1 g	Enterococci < 10 ³ /g
Austria (Lower Austria)	no occurrence in 1 g	<i>Escherichia coli</i> < 100 cfu no Helminths eggs
Austria (Steiermark)	no occurrence in 1 g	Enterococci < 10 ³ /g
Slovakia	-	Thermotolerant coliforms < 2 × 10 ⁶ cfu/g dw Faecal streptococci < 2 × 10 ⁶ cfu/g dw

¹ CFU = colony forming unit; MPN = most probable number; MPCN = most probable cytopathic number.

Moreover, limits for selected organic micropollutants, such as halogenated organic compounds, linear alkylbenzenesulphonates (LAS), phthalates, nonylphenol, PAHs, PCBs and PCDD/Fs, were included in national legislations of MS. Each country adopted different limits (Table 3). For example, Italy recently also introduced (law No. 130/2018) the limit for Toluene at 100 mg/kg dw.

As shown in Table 3, PAHs and PCBs are the most frequently regulated pollutants, followed by AOX (absorbable organic halogen) with the common limit of 500 mg/kg dw in all the countries that introduced this restriction, namely Austria, Czech Republic, Germany and Romania. As regards AOX, it is worth noting that it is still unclear which compound class of AOX, absorbed onto an activated carbon cartridge, is expected to be predominantly present in SS. Surely not the volatile chlorinated or brominated aliphatic compounds suspected to be carcinogenic.

Table 3. Limits (mg/kg dw) of organic micropollutants for sludge use in agriculture, updated from [20] (polychlorinated dibenzo-p-dioxins and dibenzo-p-furans (PCDD/F) expressed in ng toxic equivalency, ng TEQ/kg dw).

State	AOX	DEHP	LAS	NP/NPE	PAH	PCB	PCDD/F (ngTEQ/kgdw)	C ₅ –C ₄₀
EC [19]	500	100	2600	50	6 ²	0.8 ⁶	100	-
Austria (Carinthia)	500	-	-	-	6 ²	1	50	-
Austria (Lower Austria)	500	-	-	-	-	-	-	-
Austria (Steiermark) ¹	500	-	-	-	6 ³	-	-	-
Austria (Vorarlberg)	-	-	-	-	-	0.2 ¹⁰	100	-
Austria (Upper Austria)	500	-	-	-	-	-	-	-
Belgium	-	-	-	-	-	0.8 ⁶	-	-
Bulgaria	-	-	-	-	6.5	1	-	-
Croatia	-	-	-	-	-	0.2 ⁸	100	-
Czech Republic	500	-	-	-	10 ⁴	0.6 ⁶	-	-
Denmark	-	50	1300	10	3 ²	-	-	-
France	-	-	-	-	Fluoranthene 5 benzo(b)fluoranthene 2.5 benzo(a)pyrene 2	0.8 ⁶	-	-
Germany	500	-	-	-	-	0.2 ⁹	100	-
Hungary	-	-	-	-	10 ³	1 ⁶	-	4000
Italy ¹³	-	-	-	-	6	0.8 ¹²	25 ¹¹	1000 ¹⁰
Luxembourg	-	-	-	-	20 ³	0.2 ⁷	20	-
Portugal	-	-	5000	450	6	0.8	100	-
Romania	500	-	-	-	5 ⁵	0.8 ⁶	-	-
Sweden	-	-	-	50	3 ²	0.4 ⁶	-	-

¹ These limits apply to sewage sludge that comes from Wastewater Treatment Plants (WWTPs) for more than 30 000 PE; ² 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; ³ 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); ⁴ 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-c,d) pyrene, naphthalene, pyrene; ⁵ Sum of anthracene, benzoanthracene, benzofluoranthene, benzoperylene, benzopyrene, chrysene, fluoranthene, indeno (1–3) pyrene, naphthalene, fenanthrene, pyrene; ⁶ Sum of 7 congeners: PCB 28, 52, 101, 118, 138, 153, 180; ⁷ Sum of 6 congeners: PCB 28, 52, 101, 138, 153, 180; ⁸ For each of these congeners: PCB 28, 52, 101, 141, 180; ⁹ For each congener; ¹⁰ This limit refers to mg(C10-C40)/kg ww; ¹¹ This limit refers to ngWHO-TEQ/kg dw of PCDD/F + the sum of 12 PCB (No. 77, 81, 105, 114, 118, 123, 126, 156, 157, 167, 169, 189); ¹² Sum of all the 209 congeners; ¹³ Italy sets a limit for Toluene at 100 mg/kg dw.

3. Discussion

Among EU-15 countries, over 76% of total sludge production belongs to Germany, UK, Italy, France and Spain [22]. Sewage sludge treatment solutions vary considerably across Europe. The most common stabilization methods resulted from anaerobic digestion (predominant in 24 EU countries), followed by the aerobic one (predominant in 20 EU countries) Mechanical sludge dewatering is preferred compared to the use of drying beds. Nevertheless, thermal drying is mainly applied in Germany, Italy, France and UK [23]. In the framework of the circular economy, in recent years the most favored route for sludge management in Europe (in particular Germany and France) is sludge reuse on land. Thanks to the Landfill Directive, in fact, the sludge sent to landfill has remarkably decreased both in EU15 and in EU12, while the energy recovery by incineration and co-incineration is a diffuse route just in some EU-15 countries (Belgium, Germany, Netherland, Slovakia, Slovenia); those countries encounter high investment costs and diffuse social unacceptance [24]. The observed decrease in the amount of unknown disposal of sewage sludge (still remarkable in EU-12), underlines how the improvement of the data management system is already an ongoing process, but at an insufficient rate of implementation to bring the system to conditions of high reliability [25]. Many public institutions and research centers have studied the main risks to be evaluated for the safe use of sludge on the land. Among others, we should mention risks as supply of potentially toxic elements (i.e., heavy metals) and organic pollutants, pathogens dwelling in the sludge, associated odors and massive release of nitrogen to the groundwater. Consequently, from an environmental and a sanitary viewpoint, strict control of the potentially toxic elements, a full hygienization assessment and the evaluation of nitrogen release as compared to the root uptake and the capability of soil to keep it in place, are mandatory. Different authors [26,27] reviewed the research focused on organic contaminants concentrations in treated sewage sludge, and they concluded that there is a growing body of evidence demonstrating that the majority of the studied compounds do not endanger human health when they are recycled to farmland [28]. However, source control and responsible management of the pollutant inputs into WWTP must be a priority to underpin the long-term sustainability of land application practices into the future. The reuse of sewage sludge for agricultural purposes also faces additional constraints such as climate, seasons, and harvest, due to the fact that sludge is being produced all year round, whereas its application on land takes place once or twice a year during the good season [24]. One solution could be the construction of storage silos for the “unapplied” sludge, to be ideally composted with other organic wastes, namely agricultural wastes.

3.1. Emerging Pollutants

Threats arising in the past from excessive amounts of heavy metals are slowly replaced by threats from specific forms of selected trace elements-nanoparticles. Households are increasingly replacing industries as main source of contaminants, like pharmaceuticals and personal care products [13]. It is interesting to highlight that the concentrations of persistent organic pollutants (POPs) which were banned/restricted more than three decades ago (PCBs, PCDD/Fs) are still declining with apparent half-lives of ca. 10 years. For recently banned/restricted POPs (PBDEs, PFOS, and PFOA) no general trend can be seen over the past twenty years [29].

Emerging POPs such as triclosan and triclocarban are the most abundant hormone-like personal care products in biosolids, with concentrations in the range of 0.4 to 30 mg/kg dw [30]. Because of their persistence in soils, they have recently been restricted within the EU [31]. However, Clarke and Smith [26] found that the primary route of human exposure to these antimicrobials is due to the domestic environment and not to land applying biosolids.

Perfluorooctane sulfonic acid (PFOS) and perfluorooctanoic acid (PFOA), members of a large family of toxic perfluorinated chemicals frequently detected in drinking water [32], were included 10 years ago in the Annex B of the Stockholm Convention on

Persistent Organic Pollutants (POPs). The inclusion into the Stockholm convention fosters the worldwide phasing-out of the production and usage of PFOS and a faster replacement by substitutes [33]. Application of sewage sludge to land, in fact, could be a potential exposure route for PFOS to agricultural soil and subsequent contamination of groundwater or surface water [34]. A field study on PFAS accumulation in wheat plants grown in biosolid-amended soils confirmed that sewage sludge was an important source of perfluorinated chemicals, although in a less than-proportional manner with respect to amendment rate [35]. Nevertheless, to our knowledge no statutory limits for PFOS and related substances in SS have been introduced, except in Austria where the limit for Perfluorooctanesulfonate is of 100 µg PFOS/kg dw (0.1 ppm) for sewage sludge used on agricultural soils [36]. The German Fertilizer Ordinance of 2009, last amended in 2017 [37], admitting sewage sludge as a main component for fertilizers, introduced a new limit value of 0.1 mg/kg dry matter as the sum of PFOA and PFOS.

3.2. Fate and Effects of Microplastics in Sludge-Amended Soils

Plastic debris is ubiquitous in all ecosystems on earth. Through chemical, physico-chemical, and biological processes, plastic debris disintegrates into smaller particles in the environment [38]. When those particles reach a size below five millimeters, they are called microplastics (MP). In addition, MPs are specifically produced for a variety of applications, like cosmetic products or household cleaners. Many organisms are known to ingest MP particles, with reported adverse effects from the molecular level up to the behavioral level; yet unknown consequences for environmental and human health [38].

Wastewater is a main source of MP contamination in freshwater environments, since it is capable of transporting plastics from many different sources [14]. Horton et al. [39] observed that storm drains carry considerable amounts of synthetic fibers, the major source of MPs in sewage.

WWTPs effectively remove nearly 99% of microplastics from wastewater, accumulating them in sewage sludge [40]. For example, Mohajerani et al. [41] reported that approximately 0.5–3% by weight of biosolids applied to Australian farmlands consist of MPs, whose degradation over time creates nanoplastics absorbable by plants. Therefore, land application of sludge could create a pathway for MPs to enter agricultural soils [42]. Nevertheless, little is known regarding contents of MPs in the sludge-based fertilizers by digesting and composting treatments. Sludge applications on soils resulted in increased MP counts in soil samples [14]. However, studies on the fate and spread of MPs in soils after being amended with sludge-based products are lacking [43], thus the true scale of the problem has yet to be assessed. Whether the presence of MPs in agricultural soils has implications for the economy and environmental health is already under discussion [38].

Research on plastic weathering and transport processes within the soil profile are needed to understand the fate of the pollutants in the overall environment [14]. In addition, MPs are often mixed with heavy metals, dioxins, and polycyclic aromatic hydrocarbons [44]. Deliberate addition of flame-retardants and plasticizers to the plastic products, which subsequently become MPs, can cause hazards to soil flora and fauna [45].

Overall research on MPs pollution in agricultural soils is still in an embryonic stage. There are many knowledge gaps regarding: (a) plant ability to accumulate MPs from soil through uptake; (b) how absorbed MPs affect the plant growth and subsequently reach the food chain; (c) how soil microorganisms (bacteria and fungi) can be affected by the exposure to MPs; (d) thresholds for plastic particles to bring positive or negative impacts to the microbial activity; (e) transfer of MPs from one trophic level to another; (f) the effects of MPs on the growth, survival and metabolism of soil animals (invertebrates); (g) the bioaccumulation and biomagnification tendencies of MPs in the soil environment [45].

Furthermore, there is no generalized protocol to isolate, quantify, characterize, and report MP content in sludge or in soil environment [46]. MPs are being reported per surface area, per weight, per volume or as a weight ratio [14,39], hampering comparison between studies.

Depending on the origin, shape, size, and composition of MPs, it is necessary to standardize specific methods for sample collection, identification, and analysis of MPs in the organic matter-rich agricultural soils [45].

In addition, no validation method could be performed due to the lack of natural blank samples in soils. Even soils without application of sewage sludge could contain plastic particles, making them unsuitable as blank. Van den Berg et al. [46] reported an average light density plastic load of 930 ± 740 MPs/kg and a heavy density plastic load of 1100 ± 570 MPs/kg in soils without sludge addition.

Most countries allow a certain amount of foreign matter such as plastics in fertilizers. For example, Germany, which has some of the strictest regulations on fertilizer quality worldwide, allows up to 0.1% weight (wt %) of plastics [47]. In this regulation, particles smaller than 2 mm are not even considered. Thus, organic fertilizers may be a source of environmental MPs that should not be overlooked. Nevertheless, several questions such as ecological threats, dispersion mechanisms, and development in analytical and quantification technologies still require further studies in view of the high heterogeneity of the soil environment [45].

3.3. P-recovery from Sludge

More than 80% of mined phosphate rock is used for fertilizer, providing about half of the fertilizer consumed globally. The remaining P comes from recycled organic waste, as animal manures and sewage sludge [48].

The use of sewage sludge, compost and other different organic waste as organic soil amendments is a very important strategy to comply with the Landfill Directive (Directive 2018/850 of the European Parliament and of the Council), and with the “end-of-waste” policy in Europe [49]. At the same time, the land application of sludge “as a waste” is contributing to the increase of soil organic matter content, which can be very low in countries under the Mediterranean influence [28]. The EU made efforts towards achieving a unified approach to sludge management, 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”, in which sludge was excluded from the list of waste admitted for producing an “end-of-waste” compost [49,50]. Nevertheless, many MS produce large quantities of compost with sludge, like Estonia, Finland, France, Germany, Italy, Lithuania and Spain. ORBIT/ECN [51], estimated a total compost production of about 13 Mt in the EU in 2005. In Italy, the total compost production is currently estimated at 1.0 Mt/year + 0.3 Mt/year of compost with sludge, which is generally commercialized at a very low price. About 450,000 t/year of dewatered sludge (10–15% of total sludge production) are composted [50]. More or less, these figures are in line with the medium ones of the 14 MS in 2008–2010 and included in the IPTS report [49] where it results that a total production of 1.65 Mt of compost with sludge is produced in the EU27 with potential expansion to 5–10 Mt (Italy not included).

The new EU Fertilizing Products Regulation [17] states that an EU fertilizing product must not contain compost (or digestate) containing sludge as input material. Under such Regulation, the European Commission announced a public consultation on the new criteria for use of STRUBIAS materials as components for CE-Mark fertilizers (CMCs). These criteria are expected to be adopted in the first quarter of 2021. According to the JRC proposal [52], sewage sludge will be allowed as input material only for struvite and other precipitated phosphate salts, ashes, and thermal oxidation materials. The addition of STRUBIAS materials as possible component materials for EU fertilizing products is currently stimulating industry innovation to develop new processes and new fertilizer production chains. Therefore, most P-recovery from sewage sludge, in the form of STRUBIAS materials, is likely to take place in Western Europe due to increasing concerns associated with sludge land application.

The P-recovery technologies developed in the last decade have been directed to both wet sludge and ashes from sludge incineration, since the phosphorus entering the WWTP is

mainly concentrated in these two matrices. Theoretically, 90% of the phosphorus arriving at the WWTP can be recovered from sludge or ashes, while at optimal performance only about 50% can be recovered by crystallization technologies with struvite and hydroxyapatite recovery [48].

Struvite crystallization technologies have been successfully commercialized by several companies, offering multiple benefits to WWTP such as: (i) preventing operational problems associated with unwanted struvite precipitation, (ii) increasing sludge dewatering and (iii) reducing phosphorus load on the water line. In addition, the final product proved to be an excellent slow release fertilizer. The precipitation processes of phosphorus from wet streams can be applied in the sludge line on the digestates upstream from mechanical dewatering, on the return liquid streams after thickening, or other liquid streams (i.e., effluents from wet oxidation). The case of the pilot plant (Struvia™ process, Veolia group) at the Brussels North treatment plant is a typical example of the application of crystallization technology on various return streams within the WWTP, with global P-recovery yields $\geq 80\%$. Among the wet processes successfully implemented on an industrial scale in EU, AirPrex® and Ostara Pearl® showed the best operational performances in P-recovery as struvite from sludge upstream of mechanical dewatering. The AirPrex® process owns several full-scale installations in Europe and is capable of producing a product compliant with fertilizer regulations, with official compliance approval and REACH registration [53]. The first installation of Ostara Pearl® technology dates back to 2013 at the Slough treatment plant (London); to date 14 full-scale plants have been installed [54] in North America and Europe.

The P-recovery technologies from the ashes deriving from sludge incineration are recently gaining increasing popularity. Despite the fact that only a few full-scale installations are currently in operation in EU, several processes are already applied in pre-industrial or industrial pilot-scale [52]. The AshDec® process has only pilot plants currently in operation. The Zurich Werdhölzli full-scale centralized plant, completed in 2015, treats sewage sludge from over 70 WWTPs and offers the future perspective of retrofitting the AshDec® process, in order to obtain a phosphate fertilizer from the separated ashes.

As regards the full-scale installations of the EuPhoRe® process, there are two plants already built, one under construction and 4 under contract [55]. Remondis® full-scale technology is currently in the commissioning stage at Hamburg municipal WWTP and will be able to produce around 7000 tons of phosphoric acid from 20,000 tons of sludge ash every year. However, the industrial pilot plant Tetraphos (Remondis®) boasts over two years of operation. The EcoPhos technology is currently applied only at one industrial-scale pilot installation for ashes from sludge. Moreover, the Ash2Phos process has already 2 pre-industrial scale pilot installations and the first full-scale installation in Sweden is under contract [56].

Regarding sludge-derived products (bioactive compounds, biopolymers, etc.), the uncertainty is even greater [57]. There can be other sustainable solutions for efficient and cost-effective disposal of sludge with simultaneous energy and materials recovery (i.e., recovery of embedded energy, metals, and nutrients). An integrated bio-refinery system could be designed focusing on reuse options rather than disposal pathways to generate energy and recover all the possible resources [22]. It will not only valorize the sludge but will also aid in reducing adverse environmental impact to a significant extent (i.e., anaerobic bio-leaching of metals, thermal pre-treatments to enhance organics solubilization and pathogen removal). Overall, application of advanced technologies for sludge management must transform from responsible treatment toward a promising economic opportunity, while enduring to protect the environment and public health.

3.4. Pathogens

Pathogen reduction in sewage sludge is a function of temperature, time and moisture content. The deactivation rate increases with increasing temperature and decreasing humidity. Therefore, to achieve the same level of pathogen reduction, with the same dry

content, conventional mesophilic anaerobic digestion treatment takes a longer time than the thermophilic one.

The typical concentration ranges of pathogens for raw and treated sludge are shown in Table 4.

Table 4. Typical concentration ranges of pathogen presence in raw and conventionally treated sludge.

Type of Sludge	E. coli	Salmonella	Fecal Coliforms
Non treated	1×10^6 – 1×10^7 CFU/g dw	100–1000	1×10^7 – 1×10^9 MPN/g dw
Conventionally treated	1×10^4 – 1×10^5 CFU/g dw	3–100	3×10^4 – 6×10^6 MPN/g dw

But their effectiveness when pH decreases due to carbonization should still be assessed. In Germany [58], depending on the treatment with quick lime or calcium hydrate different residence time are required. With quick lime 3 + 3 h (double stage) is required with temperature of at least 55 °C, with calcium hydrate a dosage of at least 20% of lime on solid basis is required reaching a pH of 12.8. After treatment, the sludge should be stored for 3 months before use.

A big debate has recently arisen about possible health concerns of sludge spreading during the COVID-19 pandemic. Since the first publications reporting the detection of SARS-CoV-2 virus in faeces [59], it became clear that human wastewater might contain the novel coronavirus. Randazzo et al. [60] reported the first secondary treated water samples that tested positive for at least one SARS-CoV-2 RT-qPCR target. Lodder et al. [61] collected 24 h 10 L samples once a week from Amsterdam Airport Schiphol wastewater for virus analyses. Samples tested positive for virus RNA by RT-qPCR methodology, confirming that potentially symptomatic or asymptomatic individuals passed through the airport since 24 February 2020. Furthermore, other Dutch wastewater samples tested positive for the presence of viral RNA within a week of the first day of disease.

Surprisingly, in Italy, SARS-CoV-2 RNA was detected in river water and influent wastewater samples but not in treated water. This could be explained as being due to some uncontrolled discharges from isolated houses into the river [62]. Moreover, the rate of positivity in raw wastewater samples decreased after eight days, according with the epidemiological trend estimated for the interested areas.

Even though these findings indicate that wastewater could be a sensitive early warning tool to evaluate if the virus is circulating in the human population, the related risk for human health is still under debate as infectivity of viral particles in sewage and faeces remain to be confirmed. Rimoldi et al. [62] claimed the absence of significant risk of infection from wastewaters, being virus infectiveness always not significant in wastewater effluents, thus indicating the effectiveness of wastewater treatments or the natural decay of viral vitality.

Balboa et al. [63] hypothesized that the sludge line could act as a concentrator of SARS-CoV-2 genetic material. The study carried out at the Ourense WWTP (Spain), showed that most of SARS-CoV-2 particles cannot be detected in the water effluent as they are retained in sludge, confirming that the affinity of virus particles for biosolids would divert the genetic material of SARS-CoV-2 towards the sludge line. The primary settlers and the sludge thickeners were identified as suitable spots for detecting SARS-CoV-2 particles, thanks to their higher solids concentration (more virus particles).

However, the search for SARS-CoV-2 RNA alone is not sufficient to determine any risk situations due to the spreading of the sludge on the soil: the mere discovery of the nucleotide sequences of the virus is not in itself indicative of the virulence of the virus [64]. Since transmission occurs through the respiratory tract, the concerns mainly relate to exposure to droplets and dust that could be emitted during sludge application. However, there are several aspects that the literature is not yet able to clarify: (i) the impact of conventional or advanced sludge treatments on the presence of SARS-CoV-2, (ii) the possible presence in the sludge spread in agriculture (to date not demonstrated),

and (iii) the virulence of SARS-CoV-2 in these contexts [64]. The French Agency for Food, Environmental and Occupational Health & Safety (ANSES) was unable to define a level of contamination for sewage sludge due to the scarcity of available data. Nevertheless, on the basis of knowledge of other viruses such as enteroviruses, phages and animal coronaviruses, it reported that the risk of SARS-CoV-2 contamination could be considered negligible if sludge has undergone a disinfection process according with the regulations. Therefore, sewage sludge produced during the COVID-19 pandemic should be spread only after proper treatments as composting, thermal drying, thermophilic anaerobic digestion and liming.

However, ANSES recommends reinforcing controls to verify the proper implementation of treatment processes and compliance with the protective measures that must normally be adopted by wastewater treatment plant workers and professionals carrying out the spreading. Lastly, ANSES believes it necessary to carry out specific scientific studies on the monitoring of bacteriophages infecting intestinal bacteria, which are proposed as indicators of faecal or viral pollution.

4. Conclusions

The direct use of biosolids or of their derived products was always a critical point of the urban wastewater treatment plant management in Europe since at least 40 years when the European Commission started the preparatory work which took to the Directive 86/278 (Cost Action 68 and 68 bis). The attention progressively was diverted from heavy metals to organic micropollutants but the European Commission was unable to find a compromise solution between the so-called precautionary approach and the so-called risk-based approach. Some important countries, like Spain, France, Italy and UK certainly played a role to impair decisions, which considerably might restrict sludge use in agriculture with the strong argument that there is still not evidence that environmental and health problems arise within the well-established rules of the Directive 86/278.

Currently, many stringent new problems are approaching also in view of the circular economy pacall sewage sludge as a waste stream to be considered in order to recover substances. In the Draft STRUBIAS Technical Proposal, the potential contamination of soils by the organic micropollutants embedded in sewage sludge and in the sludge-derived phosphate fertilizers, is addressed. Therefore, while phosphorus is fully recognized as a critical material, the possibilities of its direct recovery by land application of sewage sludge are restricted by the prejudice that sludge remain waste and could be recovered only in the strict framework of the waste legislation.

The recent EU Fertilizing Products Regulation and the JRC proposal, established clear end-of-waste criteria for sludge-derived products; sewage sludge will be allowed as input material only for the production of (i) struvite and other precipitated phosphate salts, (ii) ashes, and (iii) thermal oxidation materials. Hence, an increasing number of pilot/full scale plants for P-recovery from sludge are currently under operation or under construction in Europe.

The sludge legislation does not yet consider the microplastic issue, despite the risk that land application could create a pathway for MPs to enter and accumulate in agricultural soils. Nevertheless, little is known regarding contents of MPs in the sludge-based fertilizers and fate of MPs in sludge-amended soils. The application of eco-design principles to products such as pharmaceuticals, cosmetics, the constituents of which (e.g., microplastics, clothing fibers, nanoparticles etc.) can be found in sludge, may help to ensure the sewage sludge quality.

Based on current knowledge, the risk of SARS-CoV-2 contamination could be considered negligible if sludge has undergone a disinfection process according with the regulations. Therefore, the sewage sludge produced during the COVID-19 pandemic should be spread only after strict controls to verify the implementation of proper treatments and compliance with the protective measures against pathogens adopted by wastewater treatment plant workers carrying out the spreading.

Supplementary Materials: The following are available online at <https://www.mdpi.com/2073-444/1/13/1/103/s1>, Figure S1: Limits of Heavy Metals for sludge-treated soils (mg/kg dw of soil; Dotted lines: limits proposed by Directive 86/278/EEC).

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