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Review

Reuse of Sludge as Organic Soil Amendment: Insights into the Current Situation and Potential Challenges

Oumaima Mabrouk [1,2], Helmi Hamdi [1,2,*], Sami Sayadi [3], Mohammad A. Al-Ghouti [1], Mohammed H. Abu-Dieyeh [1] and Nabil Zouari [1,2,*]

[1] Environmental Sciences Program, Department of Biological and Environmental Sciences, College of Arts and Sciences, Qatar University, Doha P.O. Box 2713, Qatar
[2] FWWS Program, Center for Sustainable Development, College of Arts and Sciences, Qatar University, Doha P.O. Box 2713, Qatar
[3] Biotechnology Program, Center for Sustainable Development, College of Arts and Sciences, Qatar University, Doha P.O. Box 2713, Qatar
* Correspondence: hhamdi@qu.edu.qa (H.H.); nabil.zouari@qu.edu.qa (N.Z.)

Abstract: Sludge generation as an organic by-product of wastewater treatment has seen a consistent increase worldwide due to population growth and industrial activities. This poses a chronic challenge regarding management options and environmental concerns. The agricultural valorization of unconventional organic materials has become inevitable, especially in semi-arid and arid countries that suffer from depleted soils and shortages in farm manure supply. High-income countries have also been interested in this recycling practice to mitigate landfilling or incineration issues. Sewage and some industrial sludges contain a complex mixture of beneficial and harmful substances, which varies with the origin of effluents. Therefore, sludge land application should be well managed in order to achieve sustainable agro-environmental goals. This review paper focuses on different aspects related to sludge reuse in agriculture, starting by investigating the diversity of sludge types and composition. In addition to the preponderant urban sewage sludge, the less-studied industrial sludges, such as those generated from pulp and paper mills or gas-to-liquid industries, are hereby addressed as well. Then, post-land application effects are discussed in relation to sludge quality, dose, and reuse conditions. The present paper also examines the disparities between guidelines that determine sludge conformity for land application in various countries or regions. Accordingly, special attention is given to increasing risks related to emerging pollutants in sludge such as pharmaceuticals, which have been overused since the outbreak of COVID-19 pandemic. This exhaustive investigation will assist the establishment of sustainable strategies for the safe agricultural reuse of biosolids.

Keywords: sewage sludge; industrial sludge; land application; guidelines; emerging pollutants

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1. Introduction

Agriculture is a vital sector that plays a strategic role in all economies. Its scope is not restricted to food production and security but goes well beyond the production of many raw materials for other industries [1]. As the world population grows, the demand and consumption of food increases and the use of agricultural inputs for crop production follows a similar trend. Among the major inputs needed to grow crops are water and nutrients for irrigation and fertilization purposes [2]. While water remains a vital resource to sustain life on earth, global food security will also depend on plant nutrients to improve crop production and quality [3]. In this regard, chemical fertilizers have one of the greatest consumption rates in the agricultural sector, with significant amounts of energy required for manufacturing [4]. However, excessive chemical fertilization can cause soil salinity, heavy metal accumulation, the eutrophication of water bodies, and the contamination of surface and groundwater with nitrates [5]. In the framework of the new Green Deal, the EU's "Farm-to-Fork" policy poses potential goals for more sustainable agricultural systems, including a 50% reduction in nutrient loss and a 20% reduction in fertilizer use before 2030, to improve nutrient management [3]. In this regard, research on alternatives to alleviate the excessive use of chemical fertilizers has been underway for a long time [6].

Multiple studies have shown that the land application of organic wastes can enrich soil with organic matter, provide essential nutrients to crops, and improve soil's physico-chemical and microbiological properties [7–9]. Historically, on-farm-generated wastes such as animal manures and plant residues have been the most reused "conventional" fertilizing materials in place of chemical fertilizers [10]. Most recently, these agricultural wastes have gradually been substituted by "unconventional" biowastes for intensification purposes and due to availability issues [6,11,12]. To reinforce this concept, hundreds of studies have addressed this practice in depth to illustrate the advantages and disadvantages of using different types of biowastes as substitute organic amendments [13,14]. For instance, current debates have included strong references to the many essential properties of wastewater sludge or biosolids, such as their high contents of organic carbon and mineral nutrients and their textural properties, which ensure a slow release of nutrients. All together, these parameters play a vital role in structuring the soil components and increasing the water-holding capacity. The motivation for the agricultural valorization of sludge resides also in the lower environmental impact and management cost as compared to landfilling or incineration, and the consistent availability of wastewater biosolids as compared to other organic materials [6,15]. Considerable research has been accomplished worldwide on the effect of sludge on soil and crops. In many investigations under different climatic and soil conditions, there have been reports of substantial improvements in soil quality, biomass production, and crop yield after sludge application that are comparable with or even higher than chemical fertilizers [14–18].

There are different types of sludge that could be used as organic fertilizers. The most abundant one is sewage sludge, generated during urban wastewater treatment, followed by industrial sludges from different activities [19]. For instance, the pulp and paper mill (PPM) industry plays an integral role in the global economy and each year, produces around 400 million tons of paper and substantial amounts of sludge [20]. The chemical and petroleum industries have been actively growing, and as a result, there has been an increase in wastewater generation during gas-to-liquid (GTL) processes and subsequent sludge production. Recycling GTL sludge as soil conditioner can potentially provide nutrients for plants and improve soil properties and crop yield, restore arable lands, and create symbioses across sectors [18,21]. As sludges are complex biowastes originating from urban or industrial wastewater treatment, the challenges related to sludge reuse in agriculture include odor nuisance if it is not stabilized and the presence of toxic chemicals and potentially harmful pathogens [18,22]. Consequently, sludge reuse in agriculture is regulated by guidelines that vary from one country or region to another depending on local pedo-climatic conditions, technological advances, and socio-economic situation [23]. Thus, this review summarizes recent findings on the agronomic effect of different sludges as well as the current and emerging environmental issues that restrict land application. A special focus is also given to best practices and regulatory frameworks that promote sludge reuse as sustainable and eco-friendly biofertilizers.

2. Wastewater Generation and Treatment

In recent decades, urbanization and industrialization have led to the global generation of huge amounts of waste, which can be solid, liquid, gaseous, or radioactive. Liquid wastes are mostly composed of effluents or raw wastewaters that are generated during household, industrial, and agricultural activities [24,25]. In general, wastewater from households is referred to as municipal or urban wastewater, while the term 'industrial' also applies to agricultural activities. In the literature, data on wastewater volumes are generally scarce and scattered; comprehensive reviews and assessments at a global level are missing, with only a few partial exceptions [24]. However, recent efforts from international organizations such as FAO/IWMI through AQUASTAT and the Global Water Intelligence (GWI) permit updating these assessments and providing a more comprehensive review [26]. In a recent study, Jones et al. [25] estimate global annual wastewater generation to be 359.4×10^9 m^3, of which 63% is collected and 52% is treated. Moreover, an estimated volume of 40.7×10^9 m^3 of treated wastewater is intentionally reused each year in various applications, most prominently in irrigation.

As long as human activities prevail on earth, wastewater generation will increase with the growing world population rate (estimated to be +6.5% between 2023 and 2030) and the diversification of industrial activities. This poses a serious challenge in terms of managing huge volumes of effluents daily. Wastewater, which is actually "used water", has a much more complex composition as compared to its original state before usage. As such, it may contain various contaminants of chemical and biological origins [22]. If released untreated into the environment, the raw wastewater can affect the biota and degrade natural resources. Therefore, wastewater treatment is necessary to preserve the environment by reducing or ultimately eliminating contaminants. Other benefits include the reuse of wastewater treatment output for various purposes and applications, should treatment procedures be appropriate [27]. On average, rich countries treat about 70% of their generated wastewater using advanced technologies. Treated volumes decrease to only 38% and 28% for upper and lower middle-income countries, respectively. In low-income countries, only 8% undergoes treatment of any kind. These estimates support the often cited approximation that over 80% of global wastewater is discharged untreated [28]. In high-income countries, the objective of improving wastewater treatment technologies is to maintain environmental quality and/or to provide an alternative water source when natural freshwater is scarce. Nevertheless, the direct discharge of untreated wastewater remains common in most developing countries due to several factors, including the lack of awareness, technical and institutional capacity, and infrastructure [28].

Depending on the origin and composition of the collected wastewater, specific treatments are applied at wastewater treatment plants (WWTPs), ultimately resulting in: (i) a treated sludge effluent (TSE) also referred to as TWW (treated wastewater) or RWW (reclaimed wastewater), and (ii) a residual semi-solid "by-product" called sludge. Obviously, the quality of TSE and sludge varies largely with effluent properties and treatment technologies. In general, most WWTPs are equipped for preliminary treatment (screening system), primary and secondary treatment (generally biological) while a tertiary (complementary) treatment unit can be mounted in-line to improve TSE quality in advanced WWTPs [29].

3. Sludge Composition and Pretreatment

Sludge (also known as biosolid) is defined as the residual, semi-solid organic material that is generated during the treatment of industrial or municipal wastewater [22,30]. More specifically, sludge is the settlement and accumulation of solid particles during different stages of wastewater treatment. While industrial wastewater generally requires specific treatment technologies and, in most cases, is treated on-site, urban sewer systems may concomitantly collect effluents from household, industrial, and commercial activities [31]. In most developing countries, urban wastewater collection systems also receive stormwater runoff and effluents from various origins, including small industries, restaurants, hospitals, hotels, research laboratories, service stations, etc. These daily activities do not generally adhere to internal guidelines of wastewater management and directly release various types of contaminants into the public (urban) collection system. Therefore, sludge volume and composition vary largely with the geographic area, local conditions, economic activities, and even the season [22]. Sludge quality variation concerns urban and industrial effluents equally, as both activities may undergo changes over time. In addition, treatment processes at a given WWTP could fluctuate between stabilization, improvement, and malfunction. This will pose more management challenges in the cases of landfilling, on-site storage, and reuse [22].

In general, most of the available literature has focused on the composition and recycling pathways of urban sewage sludge, including mainly agricultural valorization [15,32].

Generally, sewage sludge is composed of 20% fats, 50% carbohydrates (sugar, starch, and fiber), 30% to 40% organic matter, 3% total nitrogen, 1.5% total phosphorus, and 0.7% total potassium and has a C:N ratio of 10–20 [17]. Sludge pH is generally in the neutral range (7.0–8.5). The heat value of fully dry sludge is fairly low (~12 MJ/kg). Contamination levels depend largely on wastewater origin and treatment efficiency. On the other hand, sludges of industrial origin are less abundant than urban sewage sludge but contain a wider range of inorganic and organic pollutants [33]. As compared to agricultural soils, sewage sludge shows generally higher levels of heavy metals, salinity, and pathogens. Table 1 compares the properties of an aerobically digested urban sewage sludge to two typical south Mediterranean semi-arid agricultural soils meant to be amended by the same sludge [6,22].

Table 1. Properties of semi-arid agricultural soils and sewage sludge [6,22].

	Sandy Soil	Sandy Loam Soil	Sewage Sludge
Sand (%)	83.3	70.9	-
Clay (%)	5.2	11.9	-
Loam (%)	11.5	16.2	-
pH (1:2.5)	7.24	7.72	7.7
EC (μS/cm) (1:5)	119	155	1702
Organic matter (%)	1.15	1.31	31.8
TOC (%)	0.67	0.76	18.5
N (%)	0.1	0.071	1.18
C:N	7	10.7	15.7
P Olsen (mg kg^{-1})	17.5	14.1	220
K (mg kg^{-1})	8.44	58.8	9.54
Ca (mg kg^{-1})	1540	9560	11,354
Na (mg kg^{-1})	80	196	1231
Cd (mg kg^{-1})	0.36	0.74	4.04
Pb (mg kg^{-1})	16.2	16.5	35
Ni (mg kg^{-1})	0.58	0.44	22.2
Zn (mg kg^{-1})	5.88	2.48	342
Cu (mg kg^{-1})	1.37	0.1	174.4
Bacteria (CFU g^{-1})	103×10^5	122×10^5	125×10^7
Fungi (CFU g^{-1})	52×10^2	58×10^2	3×10^7
Fecal coliforms (CFU g^{-1})	nd	nd	9×10^2
F. streptococci (CFU g^{-1})	nd	nd	20×10^5

If properly treated and processed, sewage sludge becomes biosolids, which are stabilized, nutrient-rich, organic materials [17]. Management practices for sludges in general increase their stability and usability and reduce the environmental nuisance by limiting the non-controlled discharge of contaminants into the terrestrial ecosystems [34,35]. The treatment and post-treatment management of various sludge types are decisive factors in the design and operation of all WWTPs. The main objectives of treating sludge are to reduce its volume and stabilize the organic fraction. Properly stabilized sludge is generally odorless and can be handled with fewer risks. Reduced sludge volume also decreases the costs of pumping, transportation, and storage [36]. Untreated sludge may contain significant amounts of pathogens and hazardous contaminants that affect the soil and groundwater after disposal or reuse. Therefore, proper treatment and post-treatment management are necessary to protect the environment from adverse effects [37]. For these reasons, more effective treatment methods and new infrastructure are needed to cope with the consistently increasing sludge volumes [38]. As for treated wastewater, most environmental agencies require that raw sludge should be processed to meet quality guidelines at the outlet of WWTPs. Sludge treatment include several techniques that aim primarily to reduce sludge volume and odor nuisance following microbial degradation.

3.1. Drying and Dewatering

Settled raw sludge has a semi-solid consistency, with its water content varying between 90% and 98%, which needs to be reduced for better handling and reuse [22,39]. Drying significantly decreases sludge volume and environmental impact, producing a final stabilized, dry, granular product that is more easily transportable (Figure 1). In addition, dry sludge is better suited for agricultural use since it concentrates organic carbon and nutrients, limits biodegradation, and ensures better mixing with soil particles [40]. Open-air drying in concrete drying beds has been the most simple and widespread technique to reduce sludge moisture worldwide [22,41]. Faster dewatering techniques include plate and frame filter press, centrifugation, and belt press. At present, sludge deep dewatering is being carried out in advanced WWTPs using chemical preconditioning with high-pressure filtration and electro-mechanical dewatering [42]. For instance, Bougrier et al. [43] investigated the treatment of sludge using ultrasonic, ozone, and heat treatments. They found that the heat treatment reduced sludge viscosity and increased filtration capacity, which made it an efficient method for improving the dewatered sludge capability. However, heating fresh sewage sludge to high temperatures (500–600 °C) led to an increase in heavy metal concentrations: namely, Cr, Cu, Mn, Ni, Pb, and Zn [44].

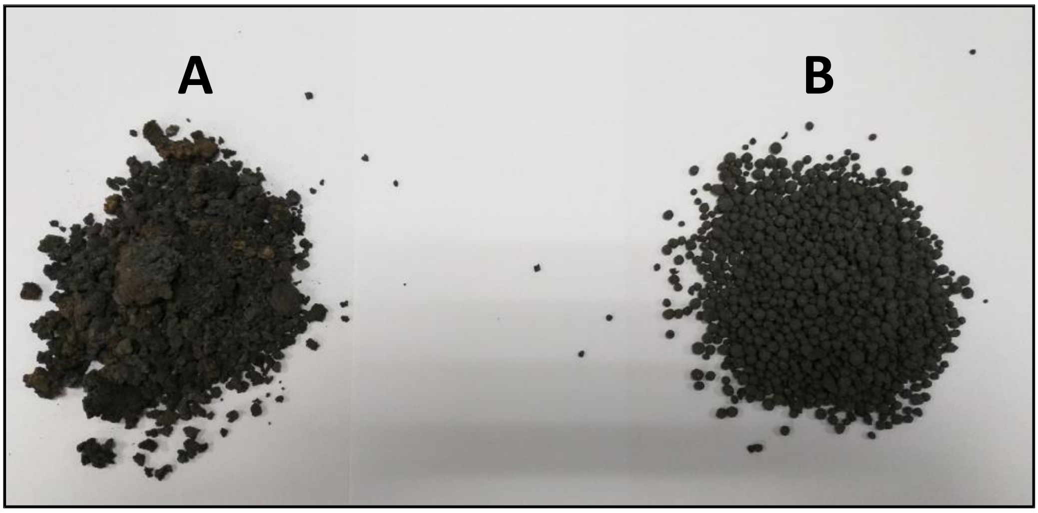

Figure 1. Dry sludge: (**A**) GTL sludge; (**B**) Sewage sludge (authors' own samples).

3.2. Aerobic Digestion

One of the recommended stabilization methods to implement for sludge management is aerobic digestion as it achieves a 70% reduction in solid content in two months [45]. Aerobic sludge digestion is a biological process that occurs in the presence of oxygen and results in organic carbon biodegradation and the release of carbon dioxide. The activation of aerobic digestion depends on air diffusion systems or jet aerators for sludge oxidation [46]. In order to enhance the effectiveness of aerobic digestion, several pre-treatment strategies have been proposed for biosolid stabilization such as alkali stabilization, ultrasonication, freeze-drying, and enzymatic digestion [47].

3.3. Electrochemical Treatment

The electrochemical treatment of sludge is a novel, clean, and efficient process used to reduce biosolid volume and eliminate pathogens due to low operating temperatures. This will further ensure the safe landfilling or land application of biosolids [48]. There are several electrochemical processes described in the literature that produce very powerful oxidants such as the Electro-Fenton (EF) process or the use of a microbial fuel cell (MFC) as a promising bio-electrochemical reactor [49,50].

4. Industrial Sludge

Industrial sludge is the by-product of wastewater treatment that does not have domestic/urban origins. More precisely, it originates from wastewater that does not contain any human excreta [51]. Major industrial activities that generate wastewater and sludge include food processing, PPMs, tanning, and the textile industries, as well as GTL processes [18,20,52]. The latter have been the least studied as they are regionally limited to the very specific process of natural gas conversion into several liquid hydrocarbon products and subsequent wastewater generation and treatment [53]. Australia and Qatar are currently the major producing and exporting countries of liquefied natural gas (87.6 and 77.4 million metric tons per year, respectively) followed by the United States, which has an annual capacity of 73.9 million metric tons [54]. Gas-to-liquid wastewater is generally characterized by high chemical oxygen demand (COD) and total organic carbon (TOC) content due to the presence of alcohol, ketones, and organic acids [53]. The final sun-dried GTL sludge is dark brown to black in color and has an earthy smell and a friable consistency. As an industrial sludge, the major difference from sewage sludge is the absence of human gastrointestinal pathogens and corresponding drug-resistant genes [51,55].

Pulp and paper mill wastewater is generated from various operations including washing raw wood materials before pulping, washing cooked pulp, bleaching pulp, and finally, from the chemical recovery system [56]. Solid materials are produced primarily from the rejection of screening, primary, and secondary sludge from wastewater management and lime sludge from the chemical recovery system [57]. As an important industrial activity worldwide, the PPM industry produces huge amounts of sludge from wastewater treatment, which constitutes an enormous environmental challenge [58]. Table 2 compares some chemical properties of GTL and PPM sludges. Both industrial sludges are characterized by high TOC and macroelement contents as well as low C/N ratios, which make them suitable for agricultural reuse in terms of fertilization potential [55,59]. Likewise, Xu et al. [60] reported on the properties of industrial ferric sludge (FS), which consists of a mixture of Fenton and waste-activated sludges. This FS contains C (16.2%) and N (3%) and a very high concentration of Fe (29.7%). Other types of abundant industrial sludges principally include those derived from the treatment of wastewater from agro-food processes, such as sugarcane mills [61] and the dairy industries [62]. Both of these sludges are, for instance, characterized by high organic carbon content (44% and 35.6%, respectively) and macroelement concentrations (24, 5.1, and 8.3 and 70.4, 14.6, and 6.1 mg kg^{-1} for N, P, and K, respectively), making them reusable for land application as potential biofertilizers as well.

Table 2. Comparison of chemical composition between GTL and PPM sludges.

Parameters (%)	GTL Sludge (Qatar) [55]	PPM Sludge (Thunder Bay, Canada) [59]
TOC	29.3	41.2
N	3.65	4.18
P	0.54	0.87
K	0.17	0.53
C/N	8.02	9.85
Na	0.82	7.04
Mg	0.72	0.81
Ca	4.97	1.42
Zn	0.02	0.06
Fe	2.1	0.48
Al	0.07	1.65
Mn	0.016	0.34

5. Sludge Management

The management of huge amounts of biosolids generated continuously during industrial and municipal wastewater treatment presents multifaceted issues, ranging from the risk of contaminant leaching, greenhouse gas emissions, and odor nuisance to high treatment costs [63]. Sustainable management of sludges as solid wastes is about developing innovative technologies to harness the benefits by maximizing waste utilization while considering the appropriate social, economic, and environmental conditions of the region of study [64]. For example, the destination of sewage sludge in Greece has included agriculture (42%), incinerators (27%), landfills (14%), and other applications (17%) [65]. In Germany, thermal disposal (incineration for energy production) constitutes by far the most common management option (55%), followed by land application (42%) and material recovery (3%), with no landfilling [66].

5.1. Landfilling

Sludge-to-landfill practice generally involves the mixing and disposal of concentrated (dried) sludge with other municipal solid wastes in open-air landfills. However, stringent legislation and higher landfill taxes have forced the wastewater treatment industry to adopt more sustainable management strategies, since sludge landfilling is not environmentally sound [6,66,67]. Many countries have established some regulations in order to minimize sludge landfilling. For instance, Germany had already stipulated that after 2005, the organic content of materials for landfill must be less than 5% to prevent groundwater pollution and greenhouse gas emissions [68]. In the EU, sludge landfilling was significantly reduced between 2002 and 2013 by 85.8%, 88.4%, 94.2%, 96.4%, and 100% in Norway, Slovakia, Poland, France, and Germany, respectively [69].

5.2. Biogas Production

Biogas production from sludge digestion has been a subject of interest for several researchers in the fields of energy and water/wastewater management, with the water–energy–carbon nexus gaining increasing importance in terms of research and analysis [70]. Sludge, as a biomass, can be used for the production of different fuels depending on the process. The anaerobic digestion, combustion, pyrolysis, and gasification of sludge produce biogas, flue gas, bio-oil, and biochar, respectively [71]. Anaerobic digestion is the most applied bioprocess technology but requires large reactor footprints due to the slow hydrolysis and methanogenesis rates. In addition, the process may produce high concentrations of corrosive hydrogen sulfide that needs to be efficiently removed using advanced technologies to increase bio-methane yield [72]. The usable energy in wastewater is determined by the organic fraction, which is measured by the chemical oxygen demand (COD) [73]. For instance, the COD of GTL wastewater is usually very high due to the presence of alcohol, ketones, and organic acids [53]. The energy produced by biogas may become profitable for the wastewater treatment plant itself [74].

5.3. Incineration

Fully dried sewage sludge could be a valuable source of renewable energy with a calorific value of approximately 11–13 MJ/kg, similar to lignite [75]. Sludge incineration produces much lower emissions of greenhouse gases than fossil fuel combustion. Accordingly, sewage sludge emits 58% and 80% less emissions than natural gas and hard coal or fuel oil, respectively, for the same amount of produced energy [57]. However, incineration can also be an expensive process when there is a need for forced drying or co-combustion and the use of auxiliary fuel. As a management option, sludge incineration is still widely practiced in countries such as Japan, Belgium, Denmark, France, and Germany, which have highly populated municipalities and advanced technologies [66,76].

5.4. Land Application

The degradation of agricultural lands has been of great economic concern due to its direct impact on crop production and the subsequent implications on world food security. Global estimates of degraded land surfaces vary from less than 1 billion ha to over 6 billion ha, with equally wide disagreement regarding their spatial distribution [77]. In any case, soil quality improvement is a mandatory practice in order to maintain productive lands in the long term. On the other hand, soils in arid and semi-arid regions are particularly vulnerable to degradation because they store little organic carbon [78]; however, these soils possess great potential for carbon sequestration after the application of organic materials [79]. Therefore, the incorporation of exogenous organic materials is required to restore depleted soils in these regions [6,80].

Since ancient times, it has been proven that organic amendments with on-farm agricultural wastes such as animal manure or plant residues improve the fertility of soils by enhancing their organic matter content, structure, nutrients, hydrodynamic properties, and biological activities [6,14,81]. Moreover, the slow release of nutrients in soil following the mineralization of added organic materials reduces the dependence on chemical fertilizers [18,55]. In recent years, agricultural intensification has led to continuous soil depletion, which has resulted in a high demand for traditional farm manure and has raised the need to seek organic matter supply from non-conventional sources. As indicated previously, sludge is naturally rich in organic carbon and macro- and micro-nutrients, which gives it unique fertilizing benefits [22,82]. Sewage sludge, and to a lesser extent some types of industrial sludge, have been proposed as alternatives to conventional organic amendments, with restrictions made to certain crops or soil conditions. In this regard, millions of metric tons of dry sludges generated worldwide may be subjected to land application in the most cost-effective way [6,18,55,83].

5.5. Benefits of Land Application

Sludge addition to croplands has been proposed as an alternative fertilization practice for farmers, particularly in arid and semi-arid regions, because it provides organic matter and plant nutrients at reduced costs and overcomes the issue of farm manure availability [6,14,18,22]. In addition, the use of biosolids in agriculture allows for compliance with new environmental management legislation, reducing the burden on landfills and recycling organic waste product [4,66]. In particular, the land application of sludge holds a great incentive in view of its fertilizing and soil-conditioning properties, unless it contains toxic compounds and pathogens at substantial levels [22]. The heterogeneous nature of sludge produced from different types of wastewater and the variations between treatment technologies necessitate a deep monitoring of sludge quality prior to land application. As such, sludge properties depend on the wastewater type, treatment processes, and sometimes the sampling season [84]. Sludge nutrients serve as a good source for plant growth, and organic materials provide beneficial soil-conditioning properties [6,8]. As compared to traditional agricultural systems using conventional inputs, expectations from the appropriate land application of sludge alone or in combination with chemical fertilizers include at least similar impact on the amelioration of soil physico-chemical properties, biological activities, and ultimately, crop yield [14,18,55]. Accordingly, the level of agricultural improvement also depends on intrinsic soil characteristics (texture, depth, and previous land use), as well as farming practices and climatic conditions [6].

5.5.1. Physico-Chemical Properties

The benefits of organic amendments could apply to any biowaste that contains organic carbon and macro- and micro-nutrients. In this regard, similar improvements to those observed for farm manures have been noticed when sewage sludge is added. Ramulu [85] showed that the addition of sewage sludge improved soil properties, such as bulk density, porosity, and water-holding capacity. Soil structural stability reflected by soil aggregation has been consistently improved by sewage sludge addition as well [86–88]. Kogbara et al. [18] reported that the addition of GTL sludge to an arid soil caused a noticeable dose-dependent increase in the microporosity before planting. In a typical south Mediterranean light-textured soil, Zoghlami et al. [89] observed a net dose-dependent improvement in TOC, N, P, and K contents up to an excessive sewage sludge application rate of 120 t ha^{-1} $year^{-1}$ (1.6%, 0.09%, 233, and 21 mg kg^{-1}, respectively) after two successive annual amendments. The same observations have been consistently reported over multiple years for the same experiment, with significant effects derived from soil texture variation (sandy loam > sandy) [6,8,14].

5.5.2. Biological Properties

The enhancement of soil physico-chemical properties is generally accompanied by a net improvement in biological activities when moisture and temperature conditions are adequate [90]. In this regard, the application of various sludges to agricultural soils may enhance soil microbial communities because of the effects of biostimulation (nutrient supply) and bioaugmentation (the addition of exogenous microorganisms) [11]. The effect of sludge on biological activity may also be used as an indicator of soil health or contamination depending on the observed microbial responses [91]. In this regard, enzymatic activities have often been measured to establish the indices of soil enhancement [6,8,14]. Microorganisms as well as plants synthesize enzymes, which act as biocatalysts of important reactions to produce essential compounds for both soil microorganisms and plants. In general, sewage sludge application improves soil's biological activity, including higher respiration rate and enzyme production [92]. After repetitive annual sewage sludge amendments, Hamdi et al. [6] and Hechmi et al. [8,14] noticed a dose-dependent enhancement of the microbial biomass and soil enzymes involved in organic matter mineralization: namely, dehydrogenase, protease, and phosphatase. In addition to sludge type, dose, and quality, soil textural effect is very important in conditioning biological activities. Nutrient retention and the subsequent biological processes are generally proportional to fine particle content in soils [8]. Dar [93] found that sewage sludge application improved soil microbial biomass by 8–28% when added at 0.75%, being highest in clay loam and least in the sandy loam soil. In addition, soil enzyme activities—namely, dehydrogenase, alkaline phosphatase, and arginine-ammonification—were all increased by 18–25%, 9–23%, and 8–12%, respectively, compared to control soils.

5.5.3. Crop Yield and Quality

In general, it has been shown that the improvement in soil properties following sludge amendments positively influences the growth and production of agricultural plants. The increase in plant performance because of sludge application often exceeded that of well-managed fertilized controls [18,55,94]. Sludge effect may be tangible since seed germination and the early stages of plant growth due to the confined conditions of moisture and temperature conferred to amended soils and the growth promoters contained in biosolids. In some cases, delayed germination or lower germination rates were observed in sludge-amended soils as compared to chemical fertilizers or unamended soils, but plant growth could be significantly promoted at later stages [2,95]. A study carried out by Kumar and Chopra [96] stated that the addition of sewage sludge changed the soil properties and resulted in the highest growth rates of common bean (*Phaseolus vulgaris* L.). Sewage sludge application also increased the chlorophyll a and b content of the same plant compared to the non-amended control [97].

As consequence of plant growth promotion when an adequate sludge amount is added, multiple studies have correlated sludge application with crop yield improvement under different conditions over a long period. For instance, it was found that sludge amendments at the rate of 0, 80, 160, and 320 t ha^{-1} in soil increased the average dry weight of sunflower plants (*Helianthus annuus* L.) in a dose-dependent manner [98]. Teixeira et al. (2009) reported that sewage sludge application might have a higher agronomic efficiency on banana yield than mineral fertilizer added at equal rates [99]. They showed that sludge has additional advantages beyond nutritional contributions, such as improved physical properties of soil and longer a gradual nutrient release because of slow microbial mineralization. Antolín et al. [100] evaluated the effects of sewage sludge amendment on the growth and yield of barley (*Hordeum vulgare* L.) under semi-arid Mediterranean conditions for four years. The repeated sludge application reduced the soil pH and increased soil TOC and CEC, which improved grain yield as compared to conventional fertilizer. Working also under semi-arid pedo-climatic conditions, Lassoued et al. [101] observed a dose-dependent increase in canola yield (*Brassica napus* L.) up to a sewage sludge dose of 100 t ha^{-1}. The effect of increasing sludge application rates (0, 12.5, 25, 50, and 75 t ha^{-1}) and farm manure (47 t ha^{-1}) on apple trees were studied for two consecutive years [102]. Sewage sludge significantly enhanced apple yield with respect to unamended control and barnyard manure. In addition, a higher cumulative yield efficiency (up to 105% at 75 t ha^{-1}) was also noticed for apple trees grown on sludge-amended soils.

As for sewage sludge, the best management approach from economic and ecological standpoints in the long run is to reuse nutrient-rich industrial biosolids in agriculture as biofertilizers. However, data on such recycling options are limited as compared to sewage sludge. For instance, the application of PPM sludge could be beneficial to improving soil fertility, biomass production, and plant growth [103]. According to Méndez et al. [104], the addition of deinking sludge improved the chemical and hydrophysical properties of peat and coir. Roszalin et al. [83] found that the application of raw and composted PPM sludge on African mahogany trees (*Khaya senegalensis*) resulted in greater height increment, diameter growth, and total plant biomass than the control and inorganic fertilizer. Java tea (*Orthosiphon stamineus* L.) also produced greater biomass with PPM sludge application, especially during the first cropping cycle [83]. In another study, sludge from a sugarcane molasses distillery added at 10% to garden soil increased the germination rate, root and shoot length, number of leaves, biomass, photosynthetic pigments, protein content, and starch in green gram plants (*Phaseolus mungo* L.). However, these parameters were inhibited at higher application rates ($\geq$40%) [105]. GTL sludge is the least studied industrial biosolid in terms of agricultural applications; most recently published data have been obtained by members of Qatar-based research teams [18,21,55]. Early outcomes showed that GTL sludge application generally increased the total porosity and volumetric abundance of different pore types in soil, which in turn influenced alfalfa performance depending on sludge dose. GTL sludge application rates of 0.75–3% resulted in comparable or better yields than soil, chemical fertilizer, and compost controls. However, alfalfa height, aboveground fresh biomass weight, and the number of tillers decreased with excessive application rates in soil [18]. Likewise, another study indicated that GTL sludge application rates for up to 3% led to better buffel grass growth compared to the above-mentioned control treatments [55].

All of these studies confirm the improvement of crop yield after sludge application when cropping trials have been carried out under adequate farming conditions. These include mainly the choice of appropriate sludge doses depending on plant species and the pedo-climatic conditions of the region of study [8]. In terms of contamination risks, most published studies have focused on heavy metal accumulation in soil and crops and have generally shown accumulation rates below thresholds for edible products [100,102,106]. However, the chemical and/or biological contamination of crops following sludge application is complex and very case-specific as it involves several parameters simultaneously: namely, sludge quality, application rate and frequency, crop species, farming practices, and pedo-climatic conditions [2,6,107].

6. Risks Related to the Agricultural Valorization of Sludge

Various sludge types, mostly sewage sludge, have been used as organic amendments in different countries, whether for agro-environmental research or commercial production purposes. However, the environmental risks related to sludge reuse have often been less highlighted in comparison to the agronomic effects [2,90,108]. As a biowaste with complex chemical composition originating from wastewater treatment, major concerns related to sludge reuse include risks of soil degradation, crop contamination, and ultimately, human health issues. These issues have been generally observed in sludge-amended soils when one or more of the following conditions occur: sludge of low quality (severely contaminated), excessive application rates, adverse pedo-climatic conditions, and inappropriate farming practices.

6.1. Physical Land Degradation

The physical degradation of agricultural soils is primarily monitored by addressing changes in soil pH and electrical conductivity (EC), which respectively reflect acidification or alkalinization and salinization risks after sludge application [11,84,109]. These parameters affect the adsorption/desorption of chemicals, soil structure, and aggregate stability, resulting in dispersive soils with poor agricultural aptitude and high contamination potential when severely affected [110]. All of these variations are closely linked to sludge quality and the dose and frequency of applications [6,8]. Depending on prevailing conditions, the rate of organic matter mineralization directly affects soil physical properties as well [11,111]. Soil pH decreases and EC increases with sludge amendment rates have previously been observed in several studies [6,8,11,22,84]. In particular, under aerated and warm conditions, with light-textured topsoils in arid and semi-arid regions, the microbial degradation of organic matter is generally fast when soil moisture is adequate. Released CO_2 reacts with the H_2O of the soil solution to form a weak carbonic acid (H_2CO_3), which in turn releases H^+ protons by double dissociation, causing gradual soil pH to decrease over time [11]. Eventually, Hamdi et al. [11] reported that one application of sewage sludge at 40 t ha^{-1} at pot-scale resulted in significant decreases in pH (to up to 5.7) compared to the original pH of the experimental soil (7.8) after 15 months. Five successive sewage sludge applications up to 120 t ha^{-1} $year^{-1}$ at field scale also led to a significant dose-dependent decrease in pH in an agricultural sandy soil (from 8.1 to 7.17) [8]. Buni [112] showed that in most acidic soils with pH levels lower than 5.5, the major plant growth limitations are due to lower nutrient availability and phytotoxicity effects of trace metals due to increased mobility under acidic conditions.

Another risk related to agricultural sludge reuse is soil salinization because sludge contains more soluble salts and exchangeable bases than arable lands [11]. Therefore, sludge addition to croplands is known to increase soil salinity, reflected by higher EC values in amended soils than those of soil before sludge addition [113]. This increase could be further aggravated in the absence of vegetation or under deficient water regimes [8,11]. The repetitive or mismanaged land application of sludge may increase soil EC over 4000 µS cm^{-1}, a value above which soils are considered to be at risk of salinization [114]. Soil salinity is mainly attributed to the accumulation of soluble ions in the soil solution: namely, Na^+, K^+, Ca^{2+}, Mg^{2+}, Cl^-, NO_3^-, and SO_4^{2-} [115]. In their pot-scale experiment, Hamdi et al. [11] found that one application of sewage sludge (EC = 2700 µS cm^{-1}) at an equivalent field application rate of 40 t ha^{-1} provoked a significant long-term increase in soil salinity up to 5257 µS cm^{-1} under deficient irrigation. EC values as high as 4200 µS cm^{-1} were also reached in an uncropped sandy soil amended with sewage sludge at 120 t ha^{-1} $year^{-1}$ for five consecutive years under natural semi-arid conditions [8]. Under similar pedoclimatic conditions, Navas et al. [116] applied a sewage sludge with an average salinity of 4010 µS cm^{-1} at an excessive dose of 320 t ha^{-1} to cultivate barley. They noticed a significant increase in soil EC after 6 months, reaching alarming values of 10,500 µS cm^{-1}. In any case, the significant accumulation of soluble salts has a long-term destructive effect on soil structure and stability by affecting particle aggregation and water retention, resulting in fine particle erosion and, ultimately, land degradation [117].

6.2. Soil Contamination

Heavy metals are known as the main class of trace contaminants that sludge application delivers to amended soils. For this reason, all sludge quality guidelines have maximum levels of heavy metals above which the sludge cannot be landfilled or reused [118,119]. It is commonly known that urban wastewater has generally lower heavy metal content than industrial wastewater. As mentioned before, toxic metals such as lead, cadmium, mercury, nickel, and chromium may be present in the municipal wastewater stream due to various urban activities (households, gas stations, SMEs, etc.) and the intentional or accidental entry of untreated industrial wastewater into the public wastewater network [120]. During the wastewater treatment process, heavy metals are partitioned between the solid and liquid phases depending on the metal partition coefficient. For instance, Karvelas et al. [121] found that Mn and Cu primarily accumulate in the sludge fraction (>70%), while 47% to 63% of Cd, Cr, Pb, Fe, Ni, and Zn remain in the treated effluent. Two groups of heavy metals can be identified based on their relative toxicity to plants and animals. The first group comprises Cd, Hg, and Pb, which are highly toxic metals to humans and animals but less toxic to plants. The second group is composed of Zn, Ni, and Cu, which are more damaging to plants than to humans and animals when present at high concentrations [122].

Once applied to croplands, sludge-borne metals will enter the soil ecosystem; their bioavailability to biota including crops is influenced by several soil properties such as pH, redox potential (Eh), cation exchange capacity, sesquioxide, and organic matter content, as well as sludge quality, application rate, frequency, and farming practices [8,22,123]. For instance, assuming that Zn, Cu, and Ni behave similarly as pH varies, the maintenance of a pH above 6.0 for grassland and 6.5 for arable soil to which the sewage sludge is applied is recommended. In this regard, high levels of $CaCO_3$ in sludge coupled to the buffering capacity of soils may maintain a pH in the neutrality range, thus preventing metal solubilization and bioavailability [6]. On the other hand, most studies have pointed out a dose-dependent accumulation of total trace elements in soil with sludge dose as compared to unamended controls [8,11,30,124]. However, very few studies have highlighted a significant heavy metal accumulation that exceeded permissible levels in agricultural soils and plants [125,126]. This is because most experimental trials consist of a single sludge application or repetitive applications with moderate amounts, in addition to plant resistance mechanisms against metal accumulation [6,127]. Likewise, water (in the form of rainfall or irrigation) can provoke the leaching of most soluble heavy metals (e.g., Cu, Zn, and Ag) from the sludge incorporation zone to deeper profiles and may even reach shallow groundwater [128].

Organic and emerging pollutants represent any synthetic or naturally occurring chemical or any biological entity that has potentially known or suspected adverse ecological and human health effects but is not commonly monitored or regulated in the environment [129]. The list includes mainly petrochemical products, pesticides, industrial and household products, surfactants, pharmaceuticals, personal care products, industrial additives, solvents, nano-enabled products, and pathogens [130]. Raw wastewater constitutes the main reservoir of various types of emerging pollutants that end up split between the liquid and the solid phase depending on their hydrophobicity. In this regard, Khadhar et al. [131] found total concentrations of 16 EPA-priority polycyclic aromatic hydrocarbons (PAHs) varying from 96 to the highest level of 7718 mg kg^{-1} in mixed urban and industrial Tunisian sludges. Many emerging pollutants are released continuously into the environment, even in very low amounts, and some may cause chronic toxicity, endocrine disruption in humans and aquatic wildlife, and the development of pathogenic bacteria resistance [51]. In this regard, the COVID-19 outbreak has induced an overconsumption of personal care products and partially metabolized antibiotics that resist wastewater treatment processes and end up adsorbed onto the sludge fraction in significant amounts as compared to normal conditions [132].

In addition to chemical pollutants, the type and number of pathogens in different sludges, and primarily in sewage sludge, can vary according to several parameters such as sludge origin, the type of treatment, and the season of collection [22]. For instance, the nature and concentrations of pathogens in sewage will depend on the health and size of the population in the catchment [133]. The nature of sewage, and hence its sludge, is such that it may contain enteric pathogens such as coliforms and streptococci, which are excreted with fecal material and are generally infective by the oral route via contaminated food after land application [22]. Humans are obviously the most likely source but, depending upon local conditions, the wastewater may also contain excreta from pets arising from storm runoff or from farm animals and birds. However, the fate of these pathogens after sludge application is quite complex because of interactions with soil physical components, prevailing climatic conditions, and native microorganisms, which can favor or hinder their proliferation [6,134].

One of the most recent concerns about biological contamination in soils after sewage sludge application is the proliferation of antibiotic-resistant bacteria (ARB) or genes (ARGs) [51]. Accordingly, Zhang et al. [135] found that the application of sewage sludge compost led to ARB enrichment with a special resistance to cefotaxime (β-lactam antibiotic) in soil. They also observed the influence of soil types on ARB dynamics after amendments. In another study, ARB were detected in long-term sewage sludge-amended semi-arid soils, with co-resistance of some strains such as *Chryseobacterium indoltheticum* (ATCC 27950) to the toxic metals Cd, As, and Be [51]. In general, the human health risks associated with different drug-resistant pathogens present in urban effluents are of particular concern, especially after the advent of the COVID-19 pandemic [51,136].

6.3. Human Health

Recent studies have essentially estimated the burden of disease associated with direct and indirect exposure of humans to chemicals and pathogens as a result of wastewater and sludge management operations from treatment to reuse [133,136]. Many of the risks to people, agriculture, and the environment posed by the land application of sludge are chronic and may only be evident after long-term exposure. Consequently, harmful effects associated directly with sludge are difficult to distinguish, measure, and document. Other than the accidental direct ingestion of contaminated food, the highest public risks of infection from sludge land application are associated with airborne exposure [137]. For instance, illnesses were reported by residents living near sludge land application sites in a variety of locations across the US, especially for Class B sewage sludge [138]. Allegations ranged from headaches and respiratory problems to death. Crop contamination after sludge application has also been documented with *Listeria monocytogenes* in alfalfa plants [139], low molecular weight PAHs in clover [140], and heavy metals in beans, maize, pepper, and sugarcane [141]. However, the contamination of foods with pathogens remains the most reported negative effect caused by sludge, in particular sewage sludge [139,142,143]. Lately, growing concerns about the risk of food contamination with sludge-borne ARB have been pointed out, especially after the outbreak of the COVID-19 pandemic [51,144]. Nevertheless, contaminant transfer from sludge-amended soils through the food chain is complex and depends on several parameters, including soil properties, contaminant mobility, plant species, agricultural practices, and bioconcentration factors. In this regard, many countries impose restrictions on the choice of crops that can be grown on sludge-amended soils, varying from a total ban for food crops to allowing only plants with low bioaccumulation capacity such as cereals, fodder, and fruit trees [118,145,146].

It is obvious that the potential health and safety implications of sludge agricultural reuse are of serious concern worldwide, and we should balance these with the need for this management option. To define a safe operating domain, it is imperative that the concepts of risk assessment be employed to establish safe levels of pollutants in the applied wastes, amended soils, or harvested crops [147]. As such, guidelines and recommendations for the safe reuse of sludge in agriculture as a biofertilizer have been established in different countries/regions, generally based on local conditions. In this regard, public awareness and acceptance of this recycling method play a crucial role as well [148]. At a time when environmental protection and food safety are a major issue in almost every aspect of life, greater emphasis has been placed on advanced wastewater treatment and stringent thresholds to make the agricultural valorization of sludge acceptable, profitable, and safe [23].

7. Guidelines and Regulations of Bio-Sludge Land Application

Over the past three decades, advisory guidelines and regulations have been established by international organizations and local regulatory/environmental agencies for the safe reuse of treated wastewater and sludge in agriculture [23]. Most international standards set an upper limit for pollutant concentrations in sludge or for the permissible pollutant concentration in the soil, or limit the maximum sludge/pollutant loading. In general, the most considered class of chemicals for quality evaluation of sludge are heavy metals [22,23,147]. Heavy metals are often assumed to be ubiquitous, transferable into the soil-plant system, and hazardous to the environment [149]. Accordingly, the toxicity of trace elements has been deeply investigated for a long time due to the fact that they are not biodegradable or only partly metabolized for certain metals [150,151]. In this regard, some trace metals are potentially toxic to plants (namely, Cr, As, Cd, Hg, and Pb), while certain others, such as Cu, Ni, and Zn, have physiological functions but are only harmful if accumulated in excess or transformed to different forms. Table 3 illustrates the variation in maximum permissible heavy metal concentrations in sludge across some countries/regions.

Table 3. Maximum permissible heavy metal concentrations in sludge destined for land application in selected countries/regions.

Element (mg kg^{-1})	USA Class A [118]	Qatar [152]	EU [153]	Tunisia [154]	France [119]	Oman [106]
Zn	2800	2500	2500–4000	2000	3000	3000
Cu	1500	1000	1000–1750	1000	1000	1000
Cr	1200	300	1000	500	1000	1000
As	41	10	-	-	-	-
Cd	39	20	20–40	20	20	20
Pb	300	300	750–1200	800	800	1000
Hg	17	10	16–25	-	10	10
Ni	420	200	300–400	200	200	300
Se	36	50	-	-	-	50

It is obvious that there are disparities in the target metal listing and concentrations among these selected guidelines (Table 3). This is probably influenced by the local sludge properties, agricultural activities, environmental priorities, and pedo-climatic conditions that characterize each country/region of study. On the other hand, some countries such as Tunisia and Qatar simply chose the strictest maximum permissible values for each metal from a compilation of foreign guidelines. For instance, Tunisia's national norm NT 106-20 is heavily inspired by the French NF U44-095 but further recommends a specific list of crops and a maximum application rate of 6 t ha^{-1} per year for a maximum period of five consecutive years to reduce the risk of long-term contamination [146]. In addition to heavy metals, decision makers have gradually introduced other contaminants of concern into sludge quality guidelines. This consists mainly of an extensive list of organic/emerging pollutants that is being regularly updated according to emerging environmental concerns [51,118,155].

The significant health hazard of sludge relates also to the risk of biological contamination due to the presence of a wide array of pathogens [156]. As previously mentioned, enteric pathogens are present mainly in sewage sludge because the raw urban wastewater stream receives principally human (and animal) excreta, but in some cases also contains food-processing wastewater and abattoir and hospital discharges. According to the US.EPA [157], pathogens are divided into four categories: namely, viruses, bacteria, parasites, and fungi. Within each group, there are various species with different occurrences and virulence. With lower levels of sanitation and less public awareness, pathogens can cause many diseases and infections, such as gastroenteritis, hepatitis, typhoid fever, etc. Gut pathogens are consistently prone to mutagenesis under various exposure conditions, which drives higher virulence and the spread of drug-resistance traits [51,158]. The risk of human contamination arises from direct contact with contaminated sludge and sludge-amended soils and the consumption of products that have been contaminated via the soil–crop–(host) pathway. Therefore, several countries have included pathogens in their guidelines to mitigate the risks of biological contamination (Table 4).

Table 4. Standards for maximum content of selected pathogens in sewage sludge.

	USA Class A 40 CFR Part 503 [118]	Bulgaria (EEA-BG) [155]	France (NF U44-095) [119]	Tunisia (NT 106-20) [146]	Jordan (JS 893/2002) [159]
Salmonella sp.	<1000 MPN */g	No occurrence in 20 g	<8 MPN/10 g	-	<3 MPN/4 g
E. coli	<1000 MPN/100 g	<100 MPN/g	<1000 CFU	-	-
Fecal coliforms	-	-	-	<2 $\times 10^6$ MPN/g	<1000 MPN/g
Enteroviruses	<1 PFU/g	-	<3 MPN/g	-	-
Helminth eggs	<1 viable/g	<1 viable/kg	<3 viable/10 g	-	<1 viable/5 g

* MPN, most probable number.

Updated guidelines also include an extensive list of organic micropollutants for sludges destined for land application, as shown in Table 5. While the burden of heavy metals in the raw wastewater stream is almost totally split between sludge and TSE at the outlet of WWTP, organic pollutants undergo biotransformations during treatment stages according to their availability to microbial degradation. This generally results in a reduction in the initial concentration of a given organic compound in effluents and the generation of several degradation by-products of different toxicities [160]. As most of these organic pollutants and their degradation by-products are hydrophobic, it is expected that a large fraction will be adsorbed on sludge particles [131].

In a similar manner to heavy metals and pathogens, there are variations in sludge quality regulations for the type and maximum permissible concentrations of organic micropollutants (Table 5). Interestingly, many countries have included polycyclic aromatic hydrocarbons (PAHs) in their guidelines. PAHs are persistent organic pollutants widely spread in the solid phases of the terrestrial environment due to natural and anthropogenic activities [11]. Therefore, PAHs can be found in effluents, mostly in those generated by industrial activities [131,161]. Polycyclic aromatic hydrocarbons have been recognized as hazardous compounds because of their intrinsic chemical stability and potential harmful effects on biota and human health [162]. Khadhar et al. [131] reported concentrations varying between 96 and 7718 mg kg^{-1} of 16 EPA-priority PAHs sampled from nine Tunisian WWTPs. These variations depend on effluent type and treatment conditions. Accordingly, the highest PAH concentrations were found in sludge originating from mixed urban (81%), hospitality (11%), and industrial (8%) wastewaters treated by natural lagooning. There have also been recent efforts to include emerging pollutants such as pharmaceuticals into sludge guidelines. Fijalkowski et al. [163] cited that bioactive substances in sewage sludge include mostly analgesics, antibiotics, hormones, psychiatric drugs, antiseptics, and stimulants. In fact, the recent COVID-19 outbreak has resulted in an overconsumption of antibiotics worldwide and raised the necessity of setting limits for antibiotic concentrations in TSE and sludge by-products as well [51,164].

Table 5. Limits of organic micropollutants for sludge use in agriculture.

	EU * [155]	Denmark [155]	Germany [155]	Portugal [155]	France [155]	Luxembourg [155]	Qatar [152]
AOX (mg kg^{-1})	500	-	500	-	-	-	-
DEHP (mg kg^{-1})	100	50	-	-	-	-	-
LAS (mg kg^{-1})	2600	1300	-	5000	-	-	-
NP/NPE (mg kg^{-1})	50	10	-	450	-	-	-
PAH (mg kg^{-1})	6 [a]	3 [a]	-	6 [a]	9.5 [b]	20 [c]	-
PCB (mg kg^{-1})	0.8	-	0.2	0.8	0.8	0.2	-
PCDD/F (ng TEQ/kg)	100	-	100	100	-	20	-
Pesticides (mg kg^{-1})							
DDT/DDE/DDD (Banned in 2001)	-	-	-	-	-	-	0.5

Table 5. *Cont.*

	EU * [155]	Denmark [155]	Germany [155]	Portugal [155]	France [155]	Luxembourg [155]	Qatar [152]
Aldrin (Banned in 2001)	-	-	-	-	-	-	0.02
Dieldrin (Banned in 2001)	-	-	-	-	-	-	0.02
Chlordane (Banned in 2001)	-	-	-	-	-	-	0.02
Heptachlor (Banned in 2001)	-	-	-	-	-	-	0.02
HCB (Banned in 2001)	-	-	-	-	-	-	0.02
Lindane (Banned in 2001)	-	-	-	-	-	-	0.02
BHC (Banned in 2001)	-	-	-	-	-	-	0.02

AOX—absorbable organic halogen; DEHP—di(2-ethylhexyl)phthalate; LASs—linear alkylbenzene sulphonates; NP/NPEs—nonylphenols and nonylphenol ethoxylates; PAHs—polycyclic aromatic hydrocarbons; PCBs—polychlorinated biphenyls; PCDD/Fs—polychlorinated dibenzo-p-dioxins and dibenzo-p-furans; DDT/DDE/DDD—dichlorodiphenyl-trichloroethane/-dichloroethylene/-dichloroethane; HCB—hexachlorobenzene; BHC—benzene hexachloride. * Proposed but withdrawn (European Commission 2000); [a] 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; [b] sum of fluoranthene, benzo(b)fluoranthene, benzo(a)pyrene; [c] sum of 16 EPA-priority PAHs.

After sludge application, biodegradable compounds such as PAHs, pesticides, and pharmaceuticals are partly or entirely metabolized by soil microorganisms. As such, the biostimulation and bioaugmentation effects of sludge on soil microbial communities could enhance biodegradation [11,165]. However, some organic pollutants can also be strongly adsorbed onto the solid phase due to aging, which limits their bioavailability for future degradation [11,166]. In this regard, Hamdi et al. [2] found that the concomitant presence of plant roots and PAH-degrading microorganisms in sludge-amended soils further stimulated the biodegradation of residual PAHs as part of a phytoremediation process. When assessing the biodegradation of three antibiotics—namely, clindamycin, sulfamethoxazole, and trimethoprim—over 61 days of incubation at 20 °C, Koba et al. [167] observed degradation rates in soil varying between 13% and 99% depending on antibiotic type and soil texture. For all these reasons, decision makers have focused principally on heavy metals as non-biodegradable compounds for sludge quality classification and for calculating application rates.

8. Prevention of Pollutant Accumulation in Sludge-Amended Soils

Due to the complex composition of different types of sludge, pollutants are inevitably introduced via land application and will subsequently accumulate in agricultural soils. Accumulation rates depend on several factors but are mostly related to sludge quality and the dose and frequency of applications [6,8]. As a result, potential sludge reuse in the long term may become limited in case of severe contamination [6,148]. Therefore, regulating the agricultural valorization of sludge has become inevitable to prevent the significant accumulation of contaminants in amended soils. Ultimately, sludge reuse should not increase contamination levels in croplands [90]. In other words, contaminant input after sludge incorporation must be equal to or less than output through plant uptake and other loss pathways. Consequently, soil quality could be preserved and contaminant transfer via the food chain could be consistently kept at non-harmful levels [118].

Heavy metals have been the major chemical pollutants assessed during the quality control of sludge destined for land application [22,118]. Several studies indicated that heavy metals naturally occur in soils [168], and even the conventional practice of mineral fertilization or manure amendments would result in a gradual increase in metal concentration in croplands [5,18,169]. In addition to their toxic potential, heavy metals are not readily assimilated through biogeochemical cycles, and neither are they biologically transformed to completely different chemical species [151]. As previously mentioned, if the concentration of a given contaminant should not be increased in soil, its input through sludge application must be balanced by output via various processes including adsorption, surface runoff, leaching, atmospheric loss, and uptake by plants. In case of trace elements, plant uptake is probably the most significant output pathway [148]. Under these circumstances, the permissible metal inputs are expected to be small to avoid any significant contamination. Based on this principle, sludge reuse guidelines have been then drafted by employing one of the following strategies:

- Setting rigorous pre-treatment requirements to minimize metal discharge in the wastewater collection networks and treatment systems;
- Requiring the sludge to undergo advanced treatment processes to remove metals;
- Establishing strict guidelines for maximum metal concentrations in sludge and for loading limits in different soil types;
- Employing a combination of all of these approaches.

In most countries, heavy metal levels in wastewater are often variable, which in turn affects sludge quality. Therefore, maximum permissible heavy metal concentrations in sludge destined for land application have been established [155]. To prevent excess metal accumulation, some guidelines also include limits not to be exceeded in amended soils after application of the "regulated" sludge (in mg kg^{-1} year or in kg ha^{-1} $year^{-1}$) [118,155]. As such, this will allow for the calculation of the maximum amount of sludge that could be added to a given soil (generally expressed in t ha^{-1} $year^{-1}$) without provoking the transfer of toxic metals via the food chain in a significant manner. Consequently, environmental risks and human exposure to pollutants via the land application of sludge could be kept to a minimum. Table 6 illustrates examples of the heavy metal concentrations not to be exceeded in sludge-treated soils. It is worth noting that in some countries/regions, other factors that influence heavy metal availability in soil, such as long-term accumulation, pH, and texture, have been taken into consideration when proposing these limits.

Table 6. Maximum permissible heavy metal concentration in soil in selected countries/regions.

	USA (kg ha^{-1}) [118]	**EU (mg kg^{-1}) [155]**	**Oman (mg kg^{-1}) [106]**	**Bulgaria (mg kg^{-1}) [155]**		**Lithuania (mg kg^{-1}) [155]**	
	Cumulative (20 years)	6 < pH < 7	pH > 7	6 < pH < 7.4	pH > 7.4	Sand Sandy loam	Clay Clay loam
Zn	2800	150–300	300	250	300	120	200
Cu	1500	50–140	150	140	200	40	60
Cr *	3000	-	400	200	200	40	60
As *	41	-	-	-	-	-	-
Cd *	39	1–3	3	2	3	0.8	1.1
Pb *	300	50–300	30	100	120	40	60
Hg *	17	1–1.5	1	1	1	0.5	0.8
Ni	420	30–75	75	-	110	35	45
Mo	-	-	3	-	-	-	-
Se	100	-	50	-	-	-	-

* Most toxic heavy metals [170].

Evidently, soils with low pH values (acidic soils) require more stringent guidelines in terms of maximum metal content, as pH influences metal mobility through enhancing cation exchange capacity and metal release into the soil solution [171]. In this regard, Oman, for instance, completely bans sludge application in soils with pH < 7 [106]. Fine-textured soils can sustain higher loads of heavy metals because they possess stronger adsorption capabilities than coarse soils due to the presence of negatively charged and layer-structured clay minerals [172]. Even within coarse-textured soils that characterize semi-arid and arid regions, small variations in the clay fraction may play a significant role in the retention of cations [6,14]. In some countries such as Tunisia and the USA, thresholds of heavy metal content in soil are also proposed as cumulative concentrations over a certain period of sludge repetitive applications: namely, 5 and 20 years, respectively [118,146]. Organic matter content in soil may also play the role of clay in attenuating metal bioavailability due to its colloidal structure and adsorption capacity [173]. Under certain soil conditions, the adsorption capacity of contaminants could be further enhanced when clay minerals and organic materials form clay-humic complexes [174]. However, these specific soil conditions have not been considered in sludge reuse guidelines. In any case, maximum permitted heavy metal concentrations in soil will influence sludge application rates by taking into account the individual concentration of each metal in the applied sludge [118]. In this regard, the analysis of heavy metals (and other potential contaminants) in a given soil should be carried out before each sludge application to calculate the appropriate sludge dose that will not increase metal content above threshold concentrations. Simultaneously, sludge doses should also be readjusted to consider several other parameters, such as plant nutrient requirements, soil topography, salinization risks, and the agricultural practices adopted on the farm [8,175].

9. Conclusions

The literature harnessed in the present review highlights that the reuse of sludge as an organic fertilizer in agriculture has become a popular waste recycling practice and could be environmentally sound and economically profitable if well managed. The available data are mostly derived from experimental studies that used sewage sludge as the main wastewater treatment by-product to be applied to croplands. Industrial sludge reuse in agriculture is the least addressed because it is less abundant in terms of volume and also due to greater concerns about contamination risks. Sludge application enhances the physico-chemical and biological properties of depleted soils by providing organic matter and nutrients, consequently improving crop yield. Globally, the presence of contaminants in sludge is variable and depends on population habits and awareness, environmental regulations, and technological advances in wastewater treatment for a given country. Accordingly, it is required that country-specific guidelines be prepared or updated, taking into account the local sludge quality, target agricultural crops, and pedo-climatic conditions. In addition, new classes of emerging pollutants such as pharmaceuticals and related resistant pathogens/genes have increasingly been detected in sewage sludge, which might cause more virulence to humans in the case of crop contamination.

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