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Review article

A review on the fate and effects of contaminants in biosolids applied on land: Hazards and government regulatory policies

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ABSTRACT

The increase in world population growth and its resultant increase in industrial production to meet its need, have continued to raise the volume of wastewater received by treatment plant facilities. This has expectedly, led to an upsurge in the volume of sewage sludge and biosolids generated from wastewater treatment systems. Biosolids are best managed by application on land because of their agronomic benefits. However, this usage has been discovered to negatively affect humans and impact the environment due to the accumulation of minute concentrations of contaminants still present in the biosolid after treatment, hence the need for government regulations. This review article examined the fate and effects of pollutants, especially persistent organic pollutants (PoPs) of concern and emerging contaminants found in biosolids used for land applications, and also discussed government regulations on biosolid reuse from the perspectives of the two major regulations governing biosolid land application-the EU's Sludge Directive and USEPA's Part 503 Rule, in an attempt to draw attention to their outdated contents since enactment, as they do not currently meet the challenges of biosolid land application and thus, require a comprehensive update. Any update efforts should focus on USEPA's Part 503 Rule, which is less stringent on the allowable concentration of biosolid pollutants. Furthermore, an update should include specific regulations on new and emerging contaminants and persistent organic pollutants (PoPs) such as microplastics, pharmaceutical and personal care products (P&PCPs), surfactants, endocrine-disrupting chemicals, flame retardants, pathogens, and organic pollutants; further reduction of heavy metal standard limits, and consideration of soil phosphate-metal interactions to regulate biosolid agronomic loading rate. Future biosolid research should focus on the concentration of TCS, TCC, and emerging pharmaceuticals, as well as Microplastic transport in biosolid-amended soils, soil-plant transfer mechanism, and metabolism of PFAs in the soils; all of which will inform government policies on biosolid application on land.

1. Introduction

The proliferation of industries, urbanization, and industrialization of suburbs and rural areas over the past decades around the world, has resulted in an increased standard of living [1–3] together with its attendant increase in the volume of wastes generated, especially sewage and industrial wastewater [4–7] (with Asia generating highest volume according to a projection of global

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wastewater generation, Fig. 1), which has to be treated before discharged into the environment to prevent pollution [8,9]. According to Giacomo and Romano [10], municipal wastewater generated in the world in 2020 was between 360 and 380 cubic kilometers, which is a projection of a 24% and 51% increase by 2030 and 2050 respectively.

Wastewater treatment generates sewage sludge and effluent as end products [10,12,13]. Due to an increase in wastewater production as a result of population growth in the last couple of years [14], there has been a corresponding increase in sewage sludge production [15–17]. According to Collivignarelli and Abba et al. [18], the total sludge production from urban wastewater plants in the EU-15 countries has increased from 6.5 MT (dry weight) to 9.5 MT (dry weight) over the last two decades and, EU-27 countries saw sludge produced up to 10 MT (dry weight) during the same period; the yearly per capita sludge generation in Europe in kg dry matter per inhabitant per year (kg DM/inhabitant year) is shown in Fig. 2 which revealed that Portugal and Italy both members of EU-15 countries, had the highest and lowest sludge generation per inhabitant respectively. Chow and Pan [19], gave the total sewage sludge produced per day from different treatment plants in Hong Kong as 1200 tons and, this figure is projected to reach 2000 tons by 2030 as a result of the improvement of treatment plant operations [20,21], environmental discharge standard [22] and population increase [10,20,23].

Raw sewage sludge contains a high level of contaminants whose concentration has to be reduced by subjecting sludge to further treatment or processing before useful applications [25–27]. Land application of raw sludge is restricted because it can result in severe environmental risks [28–30] such as water pollution and soil contamination due to the presence of heavy metals including Chromium (Cr), Copper (Cu), Nickel (Ni), Zinc (Zn),Lead (Pb), Cadmium (Cd) [31,32],Ethers (PBDEs), Di(2-Ethylhexyl) Phthalate (DEHP), Polychlorinated Biphenyls (PCBs), Polycyclic Aromatic Hydrocarbons (PAHs), Polychlorinated Dibenzo-p-dioxins and Dibenzo-p-furans (PCDD/Fs) [33,34], pharmaceuticals, endogenous hormones, synthetic steroids [35] and, organic, antimicrobial pollutants known as Per- and Poly-fluoroakyl substances (PFAs) [36,37]which can all accumulate in soil or washed to surface water during any short or long term sludge use [38,39].

2. Sewage sludge treatment technologies

The large quantities of sewage sludge produced at water treatment plant facilities undergo certain treatment processes to convert it to biosolid (Fig. 3) for more beneficial use. Major reasons for the treatment of sewage sludge are to drastically reduce the amount of disease-causing pathogens and contaminants [40], reduce the volume of sludge for better handling [41], make sludge economically more viable [42], and to reduce the attraction of vectors such as flies, fleas, rodents, mosquitoes, birds; all these are referred to as vectors in the EPA's Biosolid Rule [43]. Some of these treatment technologies, which include anaerobic sludge digestion, incineration, sludge compost, sludge drying, deep-dewatering/thermal hydrolysis, and alkaline stabilization/pH treatment [19], are discussed below.

2.1. Anaerobic sludge digestion

This sludge stabilization process involves the use of biological microorganisms in the absence of air to convert volatile solids in raw sewage sludge to biogas, carbon dioxide, and water [44]. The biogas produced during this process can be further treated for energy recovery to utilize its methane component as fuel while the stabilized solids (biosolids) can be used for soil conditioning [44]. Anaerobic sludge digestion is the most commonly employed sludge stabilization process and unlike aerobic digestion, it is lower in

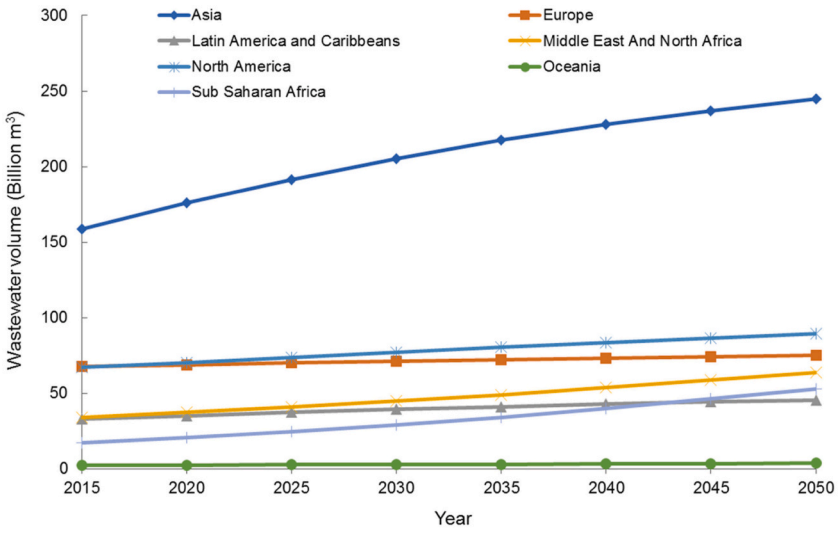


Fig. 1. Projected increase in global wastewater generation since 2015 [11].

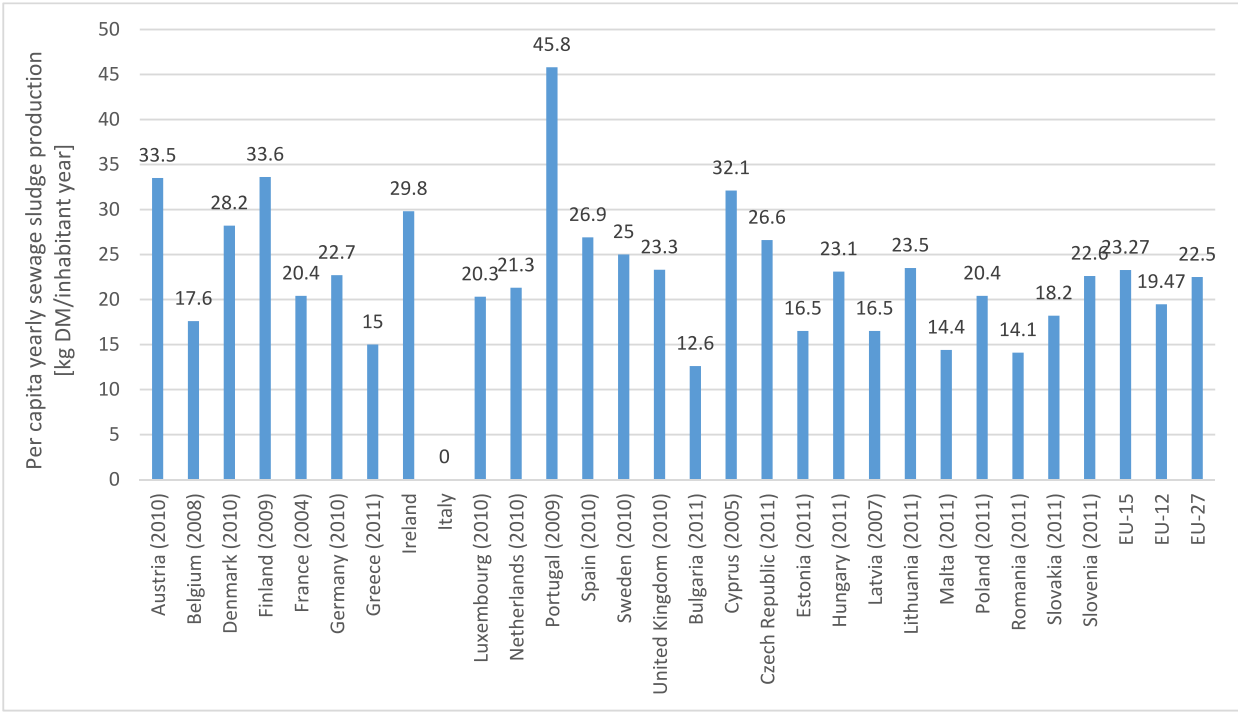


Fig. 2. Comparison of yearly sewage sludge generation per person in Europe [24].

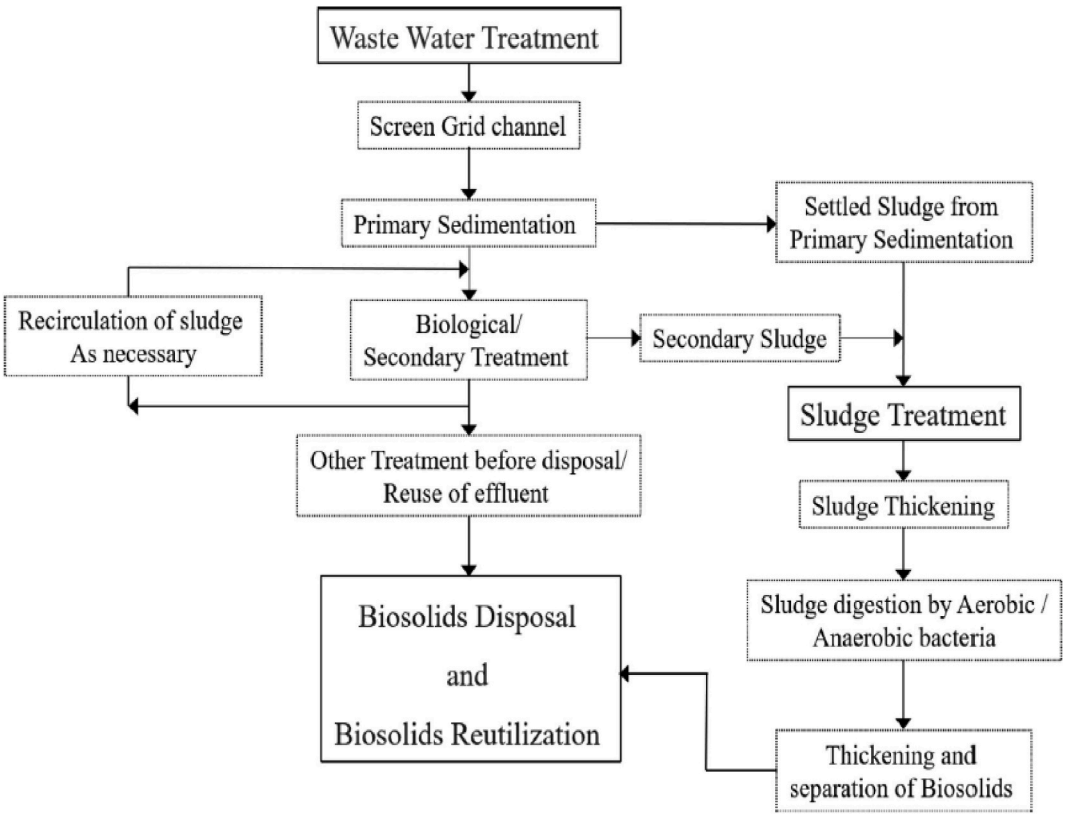


Fig. 3. Block flow diagram showing biosolid production from wastewater treatment [49].

energy utilization and generates fewer wastes; it also uses fewer chemicals than alkaline stabilization. However, it has a higher initial capital cost and complex operation than the other processes [44].

2.2. Sludge incineration

Incineration of sewage sludge is a thermal treatment process [45] that oxidizes organic sulphur, nitrogen, carbon, and phosphorus in raw sludge to inorganic, mineral solid, and gaseous products [44]. The process may be carried out in a furnace at a high temperature greater than 800 °C so that the organics in sewage sludge may be oxidized [45].

2.3. Sludge composting

This is an aerobic sludge digestion process that uses biological thermophilic or mesophilic microorganisms in the presence of air, to degrade raw sewage sludge combined with other waste materials such as rice hulls, straws, sawdust, and recycled compost (amendments) and, shredded green wastes, tyres, and wood scrapes (bulking agents) to produce a pasteurized solid product [44]. The stages involved in sludge composting include 1). Mixing raw sludge with amendments or bulking agents at the preprocessing stage, 2). Aerating the mixture for decomposition using air provided by a mechanical pumping device, 3). Recovery of undegraded additives (amendments and bulking agents), 4). Stabilizing the product formed by curing and storing and, 5). Screening, grinding, or comminution to remove undegraded materials and produce a uniform product [44].

2.4. Sludge drying

This is an intermediate thermal intervention process for treating wet sewage sludge that involves reducing/evaporating the moisture content of raw sludge thus, significantly lowering its weight and volume for better handling and recovery to meet important applications such as its use as fuel for incineration plants, compost material, and fertilizer [46].

2.5. Deep-dewatering/thermal hydrolysis

Raw sewage sludge can be treated by first subjecting the sludge to a deep-dewatering process before thermal hydrolysis. Deep-dewatering, which involves removing moisture from sludge to reduce its overall volume for better operational handling and subsequent cost of transportation [47], is carried out to thicken sludge to about 16–18% of dry weight [48]. The partially thickened sludge is then exposed to high temperature, between 160 and 180 °C, and a pressure of about 6 bars in the thermal hydrolysis step to sterilize the sludge and kill pathogens [48].

2.6. Alkaline stabilization/pH treatment

This is a chemical treatment method employed to reduce pathogenic colony, putrescence or decay, and odour during the sludge treatment process. It involves the use of slaked or hydrated lime (calcium hydroxide) to raise the pH of raw sludge to create a high alkaline system [44]. The amount of lime used may be reduced by dosing waste solid materials into the system thereby making the system cost-efficient [44].

3. Biosolid management and disposal methods

Biosolid management technologies are employed to dispose of the large quantities of biosolids generated from sewage sludge treatment. Many researchers have described biosolids as treated sewage sludge materials produced to meet Environmental Protection Agency (EPA) standards for agriculture and land application [12,36,43,50,51]. Kinney and Furlong et al. [52] in their study, defined biosolids as carbon-rich, organic beneficial solids, produced during wastewater treatment to meet local and United States Environmental Protection Agency (USEPA) regulations for pathogen, metal, and nutrient content, suitable for land application and, Thomas Burke [13], noted that biosolids are the main organic solids from municipal wastewater treatment that can be valuably recycled to amend agricultural soil.

Biosolid scan be classified as class A or class B solids and this classification is based on sewage sludge treatment methods [53] and the extent of pathogen reduction [13,43,54]. Class A biosolids are produced when sewage sludge is treated so that the presence of pathogens is below the detectable amount [27]. It can be bagged, sold to the public, and used without restrictions based on its pathogen content [43,53]. Class B biosolids, on the other hand, are produced when sewage sludge is treated to only reduce pathogen concentration which may still be detectable [27]. They are regulated for use on land to prevent man and animal exposure to contaminants, or environmental degradation [43,53].

Biosolid management and disposal techniques have evolved over the years with many countries favouring land/agricultural application to incineration, landfilling/surface disposal, or production of building materials, etc. [55]. In the United States for instance, of the 8.2 million tons of biosolid produced in 2010, 54% were land applied compared to 18% and 19% that were landfilled (surface-applied) and incinerated respectively (Fig. 4) [56]. In the same vein, the United Kingdom Water Industry Research (UKWIR) [50], noted that 150,000 ha **o**f agricultural land in the U.K. utilized a total, between 3 and 4 million tons of biosolids produced annually for land application, representing more than 75% of the total production quantity compared to other sludge management practices

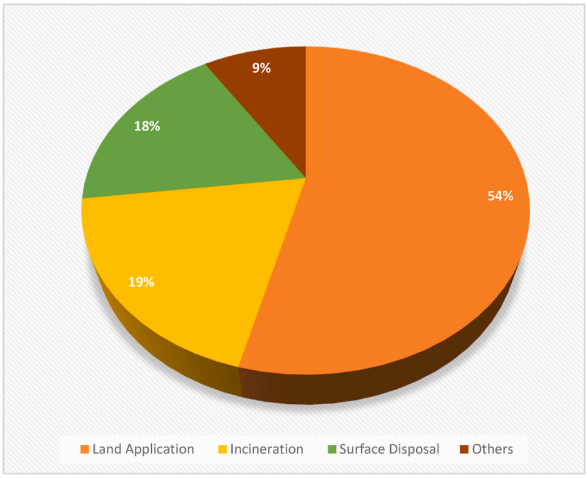


Fig. 4. Biosolid management and disposal practice in the U.S [5].

including incineration/energy recovery which utilized 12% of total sludge produced, land reclamation 5%, and others 5% (Fig. 5) while, in China, approximately 39.04 million tons of biosolids were produced annually (2019 data) from which 29.3% was disposed via land application, 26.7% incinerated, and 20.1% landfilled [57]. Some of the disposal routes of biosolids currently in use include land application, incineration, sanitary landfilling/surface disposal, and production of construction materials [51,57] are discussed in the following section.

The pie diagrams shown above (Figs. 4 and 5), clearly revealed that land application of biosolids is the most popular/common biosolid management practice in the U.S. and UK.

3.1. Land application

Biosolid land application involves spreading the stabilized, treated solids on surface soil or incorporating them into the ground on the basis of government regulations and guidelines [5]. This practice can occur at sites such as forests, mine reclamation sites, agricultural lands, disturbed lands, golf courses, or parks [5]. Land application of biosolid is sustainable, cost-effective, unlikely to cause environmental degradation, and generally improves soil fertility making it a widely accepted technique in many regions [50,56,57].

3.2. Biosolid incineration

Biosolid incineration is an "end-of-useful-life" approach to biosolid management. This is because after subjecting biosolid to high-temperature combustion in the presence of air and converting it to carbon, ash, and char, it is taken to a landfill site for surface disposal [57]. Incinerating biosolids before landfilling kills and inactivates pathogens that could cause environmental pollution [56]. Incineration as a biosolid management technique is discouraged as a result of the release of toxic, greenhouse gases into the environment thereby, causing air pollution [57].

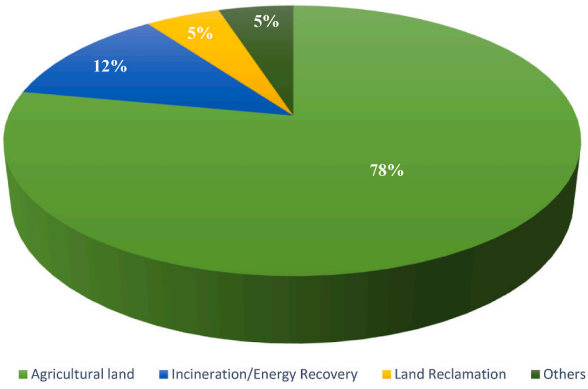


Fig. 5. The UK biosolid disposal pathways [50].

3.3. Sanitary landfilling/surface disposal

The biosolid landfilling/surface disposal management option involves dumping biosolids in excavated spaces and then compacting and leveling the area to reduce waste volume [58]. The disposal can be on a mono-disposal landfill where only solid wastes from wastewater treatment are dumped; this type of landfill is regulated by the 40 CFR Part 503 legislation or, on a mixed landfill (co-disposal landfill) where biosolid wastes are combined with municipal solid wastes (MSWs), which is regulated by the 40 CFR Part 258 legislation for MSWs [5,56]. Management of biosolids by landfilling should be distinguished from land application practice, where biosolids are applied to soil for enhancements [56]. It is important to point out that landfilling biosolids can result in severe public and environmental health complications due to the uncontrolled release of contaminants into the environment [56].

3.4. Production of construction materials

Application of biosolids in the field of construction is one of the sustainable disposal routes of biosolids, which is dependent on the material's geotechnical properties. Modified/stabilized biosolid composites and aggregates applied in construction can be used for embankment filling, land reclamation, brick manufacture, and the development of other construction materials [49]. Biosolid's use in this way is a major contribution to the circular economy ecosystem. The addition of substances like lime, cement, bauxson, glass, and fly ash has been reported to greatly enhance the stability, compaction characteristics, shear, and compressive strengths of biosolids [49]. Usually, before composites are applied in construction, standard tests such as large-scale direct shear tests, vane shear tests, cone penetration tests, triaxial tests, California bearing ratio, consolidation tests, compaction tests, etc. [49].

The present review article adopted the narrative research design to provide a contributory insight into the evidence of environmental hazards associated with biosolid land application disposal routes and discussed government legislations that regulate this biosolid management technique including how application practices can influence government policies. A flowchart of the research concept is shown in Fig. 6A scoping search was initially carried out to identify the knowledge gap in the field and develop a research question. Using keywords and phrases in the research question, which are combined with Boolean operators, an elaborate search for literature in major databases such as Scopus, Google Scholar, and Science-Direct was done. The total number of relevant articles obtained after applying inclusion/exclusion criteria such as year of publication, title, originality of the study, whether the study was peer reviewed or not, methodology, and reported outcome or conclusion, was 150. These articles were each assessed for limitation and bias using the Cochrane Collaboration Tool with a low overall risk of bias.

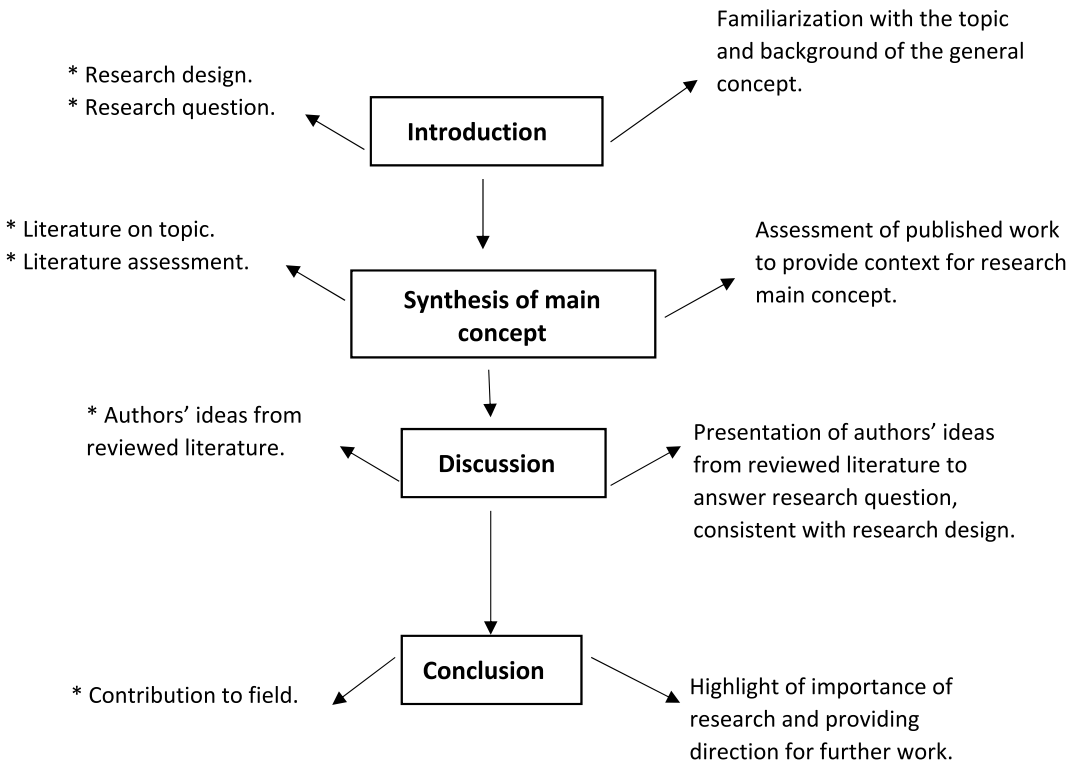


Fig. 6. Flow chart of research concept.

4. Biosolid land application and the circular economy

The need for land or agricultural soil amendment especially in an arid or semi-arid region where crop yields are very low, as a result of the combined effects of harsh climatic conditions like land degradation occasioned by erosion, long and persistent drought conditions, poor cultivation practices, etc. [59,60], has necessitated biosolid application practices on land for agricultural purposes [61]. This practice of recycling biosolids as organic fertilizer on land is an aspect of the circular economy [62] that tends to eliminate large volumes of sewage sludge produced annually around the world, especially in the developed world, and thus, reduce its environmental pollution by reprocessing and consuming it, forming a circle of sludge regenerative production and consumption [63].

Being a widely practiced and economically viable biosolid management method, biosolid land application in agriculture as soil amender and conditioner [12] has been reported to enhance the diversity of soil beneficial bacteria (rhizosphere bacteria) community, plant performance, and growth, improve soil properties [64], provide nutrient and stable organic matter to soil [12,50,65]. The organic nature of biosolids makes them better alternatives to mineral (inorganic) fertilizers [61] as they are high in plant nutrients such as nitrogen, potassium, phosphorus, organic matter and, can improve the yield and quality of crops [12]. Also, biosolids' slow-release of plant nutrients (which may potentially reduce pollution caused by leaching of excess nutrients, a common occurrence with mineral fertilizer)make them readily available to plants and increase their efficiency for plant growth and soil enhancement; these and together with their low cost make them more attractive than mineral fertilizer [12,54,66].

4.1. Biosolid land application best practices

Recycling biosolids to agricultural soils, rangelands, forests, and damaged lands to replenish soil organic matter, supply soil nutrients, and enhance land reclamation is known as biosolid land application [66]. Such land application has been observed to increase plant drought tolerance and improve soil texture and water holding capacity [67], which favour root and shoot growth and, remediate disturbed land area [66]. Application of biosolids on land, however, can result in environmental pollution due to the bioaccumulation of excess nutrients from repeated use as organic fertilizer [68]. It is therefore important to apply biosolids while observing certain management practices (Best Management Practices) which can allow for proper usage and protection of human and environmental health [54].

(a) Following Strict Biosolid Agronomic Loading Rate: This describes the frequency at which solid or liquid biosolid is applied (or injected) to an agricultural area of known dimension [69,70]. High loading rates may result in the accumulation of nutrients like phosphorus on soil surfaces thereby, increasing the likelihood of runoff (caused by erosion) to streams causing water pollution [54]. The application rate of biosolids should be determined by soil nitrogen requirement if the phosphorus index of the soil is low or medium and, by reducing phosphorus content if the index is high [54]. Application rate should also consider soil nutrient content and crop requirement (by considering trace element content of the soil) as well as, nutrient content of the biosolid (considering trace elements content of the biosolid) [54].

(b) Application Site Slope Limitation: Biosolid land application should consider site steepness or slope to avoid excess nutrient runoff to nearby plateaus or streams [70]. Usually, for sites with 0–6 unit slopes, the application rate is not limited but may be varied when erosion is a concern and soil conservation practices are employed on sites with 7–12 unit slopes [71]. Biosolids can be applied on grass vegetation sites with more than 80% of the site covered, for slopes of 12 units and above [71].

(c) Application Site pH Limitation: The availability of soil nutrients for plant growth is affected by soil pH value [72]. Depending on the soil solution pH test (whether salt or water solution test) conducted, biosolid application on sites should be avoided when pH is within the acidic region of less than 6.0 or alkaline region of greater than 7.5 for a salt test or, 6.5 and 8.5 for a water test [71].

(d) Establishing Buffer Zones: Biosolid application is discouraged on neutral land areas around natural water bodies like sink boreholes, ponds, wells, etc., for environmental protection [73]. These buffer zones (neutral areas) can be established about 300 feet from water bodies, 150 feet from residential areas or dwellings, 100 feet from wetlands, and 50 feet from intermittent flowing streams [71].

(e) Restriction of Public Access to Biosolids: General public use of biosolids should depend on the type and government approval for public use. Class A biosolids to be applied on home gardens, root and vegetable crops gardens, and other public sites must comply with strict regulations and must be approved by the government before distribution or use [71,74]. Class B biosolids should not be applied on public sites, turf lawns, or residential farms, unless they are produced by a regulated industry (i.e. incorporated) [71].

(f) Encouraging Biosolid Application Deferment on Grazing Land: Biosolid land application should be discouraged immediately after livestock grazing or crop harvesting on agricultural fields to prevent pollution caused by nutrient washout (run-off) which may occur after these activities as the soil becomes prone to erosion [69,70]. Generally, a 30-day period should be allowed before biosolid application on land after grazing or harvesting [69].

(g) Encouraging Soil Conservation via Biosolid Application: Biosolid application on land can protect topsoil from erosion and maintain soil productivity and fertility [75], as the organic matter present in biosolid help to increase soil water holding capacity, reduce compaction [67,76], and provide a platform for microbial activity [71].

(h) Discouraging Biosolid Application on Saturated or Frozen Soil: Biosolid land application should be discouraged on frozen, snow-covered, or saturated soil unless restriction controls can be employed to prevent pollution from snowmelt or water runoff [71]. If biosolid must be applied on frozen soil, then the land must have a maximum of 2 or 6 units of slope and a buffer area of 100 or 300 feet respectively and must be allowed between a water body and the application site [71,77,78].

(i) Protection of Threatened Biological Species: Biosolid land application should be practiced in such a way that it does not harm endangered soil biota such as bacteria, fungi, earthworms, protozoa, nematodes [71,79], insects [69] or, threaten their habitat [80].

5. Fate, environmental hazard, and effects of contaminants in Biosolid applied on land

Although biosolids are beneficial to crops when used as organic fertilizer and soil conditioner [81], they do contain contaminants at minute concentrations in nanogram/liter (ng/L) or lower (the concentrations of contaminants in biosolids are usually below regulatory standards), which can accumulate in soils [82] due to repeated land application and, eventually cause environmental pollution [83]. These emerging contaminants discovered in biosolids may include pharmaceuticals and personal care products (P&PCPs) such as prescription drugs that may be found in human faeces and urine [84,85] and, chemicals in soaps, detergents, shampoo, etc. [83], endocrine-disrupting chemicals (EDCs), surfactants, flame retardants, per-and poly-fluoroalkyl substances (PFAS) [84], microplastics (or nanoparticles) in toothpaste and laundry wastewater, etc. [86–88], which all enter the wastewater treatment plant and thus, sewage sludge through the sewer system, remaining in biosolid after sludge treatment as treatment does not completely remove these contaminants [27]. When contaminants enter the environment through biosolid application on land, they may be persistent, degraded, undergo additional transformations, or converted into by-products with hazardous effects on the environment [84].

Researchers have studied the effects and potential hazards of biosolid contaminants on the cell structure, reproduction, biochemistry, growth, and mortality of the flora and fauna colony in an environmental ecosystem [89,90]. While some of the literature reviewed agreed that contaminants may have a minor effect at low soil accumulation on soil microbes and animals [89], others presented evidence of a major effect on terrestrial and aquatic animals [91–93]. For instance, Diclofenac, a pharmaceutical drug detected in soil amended with biosolid, was linked to a drastic decrease in the vulture population in Pakistan [91]. Also, Garric et al. [92] in their study, demonstrated the effect of Ivermectin on the development of aquatic invertebrates and Lange et al. [93] demonstrated the endocrine disruptive effect of Ethinylestradiol (a drug used for contraception in humans) in fishes. It is therefore imperative to understand the fate and hazardous effects of emerging contaminants and persistent organic pollutants (PoPs) found in biosolid used for land application in order to prevent environmental pollution and inform government regulations on biosolid use.

5.1. Heavy metals

Biosolid application on land may be limited by its heavy metals content [35]. Heavy metals such as arsenic (As), mercury (Hg), selenium (Se), molybdenum (Mo), titanium (Ti), antimony (Sb) [94], zinc (Zn), cadmium (Cd), copper (Cu), chromium (Cr),lead (Pb), and nickel (Ni) [95] are commonly found in biosolids and, their concentrations depend on the nature of wastewater and region of origination (Table 1) [96], wastewater treatment process [96–98], and the industrial manufacturing process [95]. Domestic or household wastewater usually has a lower heavy metal concentration than industrial wastewater [95]. While some heavy metals in biosolids are appropriate for plant growth at low concentrations (Mn, Zn, Cu), others (Pb, Cd, Hg, As) are not [99]. The accumulation of heavy metals in the soil becomes high over a long-term period of biosolid application and even, short-term use has been observed to raise their concentrations considerably [95].

5.2. Per- and poly-fluoroalkyl substances (PFAs)

These synthetic, organic chemical compounds, used in several applications such as water and heat-resistant fabrics [100,101], cleaning products, dental floss [102], shampoo, and cosmetics [103], paint production, industrial emulsifier and surfactant manufacture, non-sticky cookware, oil-repelling containers, aqueous film-forming foams for firefighting operations [84], etc., are found in sewage sludge and have been detected in soil amended with biosolids [36,104,105].

The number of PFA variants (Table 2) synthesized after the first (Polytetrafluoroethylene or PTFE) was produced in 1938, has been on a steady increase since the early 1950s and presently, there are over 5000 PFA compounds that have been commercially manufactured [106]. Of these PFA variants, long-chain PFAs such as polyfluorooctane sulfonic acid (PFOS) and perfluorooctanoic acid (PFOA) are most detected in polluted soils [107] as they are more persistent and bioaccumulative in the environment [36,106,108] due to their preferential bonding to the hydrophobic or protein binding site of the soil [106]. Because of the persistent fate of PFAs especially PFOA and PFOS in environmental matrices, their concentrations are elevated within a short period of biosolid application on

Table 1
Variation in concentrations of heavy metals in biosolids due to region and nature of wastewater generated.

Country	Zn	Cu	Cr	Pb	Ni	Cd	Hg
USA (1977)	1740.0	850.0	890.0	300.0	82.0	19.0	3.0
USA (2002)	750.0	511.0	35.0	65.0	23.0	2.3	1.5
France (1999)	761.0	286.0	4.5	107.0	35.0	4.5	2.1
France (2017)	60–200	20–100	30–100	70–100	15–70	0.5–1.5	0.1–1.0
China (2018)	674.0	204.0	236.0	26.0	334.0	0.9	0.9
Concentration limit, EPA (2002)	7500.0	4300.0	ND	840.0	420.0	85.0	57.0

ND = not determined. All values are in mg/kg [98].

agricultural soil [109,110] with the attendant phytotoxic effect on certain plant species [111]. This characteristic of PFAs has been demonstrated by researchers in various research, for instance, Damien [106], reported the phototoxic effect of PFOA on Thale Cress (used in genetic experiments) at a concentration in excess of 181 ng/L in a study to investigate the fate, behaviour, and ecological impact of biosolid-derived PFAs and, Li et al. [111], carried out a metabolomic study of Lettuce exposed to elevated concentrations of PFOS and PFOA, between 500 and 5000 ng/L and observed between 88 and 95 significantly disturbed metabolites (amino acids, prenol lipids, phenolic compounds, carboxylic acids)for PFOS and PFOA exposure respectively. Public health hazards of human and animal exposure to high levels of PFAs include reproductive defects, neurological development delays, low birth weight, bone variation, accelerated puberty, changes in liver and kidney functions, thyroid disease, testicular cancer, etc. [94,112].

In a study conducted by Oliveira et al. [113], the researchers reported that the concentration of Zn, Ni, Cr, and Cu in a biosolid-amended soil rose sharply over a relatively short period of two years. The speciation, bioavailability, and fate of heavy metals after biosolid application to the soil ecosystem is a function of the soil's (ionic strength, pH, and dissolved organic carbon) and biosolid's chemical characteristics [95]. Most metals are not biodegradable or chemically degradable and are, thus persistent in the environment [114,115] leading to metal leaching through soil profile and contamination of groundwater [99]. Heavy metals in the soil's rhizosphere (where toxicokinetic and toxicodynamic processes take place) in the form of dissolved free cations and negative, positive, or neutral species, are absorbed by plants through the root system [116], causing bioaccumulation of these metals [114]. Environmental effects related to heavy metal accumulation in the soil are severe on plants, animals, and humans alike [117]. Plant exposure to high concentrations of heavy metals may result in stunted growth, reduced yield, altered metabolism, etc. [116]. The effect of heavy metals on humans is the result of the biomagnification of these toxic chemicals in the body which affects different organs [118]. These effects on humans, which may be acute or chronic [118,119] include nervous system disorders, gastrointestinal and kidney dysfunction, immune system dysfunction, skin lesion, birth defect, vascular damage, and cancer [118].

5.3. Excess nutrients in biosolid

The major plant nutrients in biosolids are magnesium (Mg), nitrogen (N), potassium (K), phosphorus (P), sulphur (S), calcium (Ca), and micronutrients [120]. Among these nutrient types, P is commonly found in excess in biosolid-amended soils [121] because most of the phosphates in wastewater are retained in sewage sludge (biosolid)during the tertiary treatment step [122] and, because farmers by following government regulatory guidelines, apply biosolid to soil based on plant requirement of N, [121]; P accumulation in the soil thus increases rapidly as the ratio of N to P varies [123]. Long-term application of biosolid leads to excess concentration of P in top soils [121] which can be washed into surface water resulting in algae invasion and river eutrophication [123]. Increased phosphate concentration in the soil has also been observed to affect soil metal lability [124]. Mossa et al. [124], in a study, reported that with rising levels of metal concentration due to prolonged biosolid application, there is an interaction with increasing phosphate concentration, which reduces the lability of Pb and Cd metals and, increases lability of Zn, Ni, and Cu metals as a result of weak soil binding. These interactions between soil phosphate and heavy metal concentrations should be taken into account when the government sets regulatory standards [124].

5.4. Microplastics (MPs)

These emerging contaminants are plastic particles with diameters less than 5 mm [86–88]. MPs commonly found in biosolids include microbeads (Fig. 7) and microfibers (both primary MPs) manufactured by industries to act as fillers, abrasives [125], viscosity control agents [126], and for aesthetics purposes in fabrics, personal care products, consumer plastics [127,128], etc.; secondary MPs are formed by processes such as biodegradation, photo-oxidation, or mechanical abrasion of large plastic debris in the environment [129].

Most MPs are collected in sewage sludge during wastewater treatment and are not completely removed when the sludge is treated to produce biosolids [86]. When MPs enter the environment through the application of biosolids on land, they are hardly removed (they can persist in the environment for many years) but can be fragmented into smaller-sized particles (nanoplastics have a size range less than 1 μm) by environmental forces [86–88]. MPs have been reported to act as vectors (carriers) for pollutants, heavy metals, hazardous additives, and pathogens, which can cause health complications in humans and animals [131]; these pollutants can be spread through contamination pathways such as water and food ingestion [132] as water and food samples have been discovered to

Table 2
Pollutants and emerging contaminants recently discovered in biosolid. Cl-PFCA: Chlorine substituted PFCA; UPLC-ESI-MS/MS: Ultra Performance liquid Chromatography with Mass Spectrometry and Positive Electrospray Ionization; PFHxS: Perfluorohexanesulfonic acid; ASE: Accelerated solvent extraction; PFHpA: Perfluoroheptanoic acid; PFEA: Perfluoroalkyl ether alcohol; H-PFCA: Hydro substituted PFCA; HPFSA: Hydro substituted PFSA; HPLC-FD: High performance liquid chromatography with fluorescence detection; LC-MS/MS: Liquid chromatography with mass spectrometry; LOD-limit of detection; FTS: Fluorinated telomer sulfonate; LOQ-limit of quantification; OBS: P-perfluorous nonenoxybenzenesulfonate; PFBS: Perfluorobutanesulfonic acid; Cl-PFESA: Chlorine substituted perfluoroalkyl ether sulfonate; GC/ECNI-MS: Gas chromatography/mass spectrometry operated in negative ionization mode; PFBA: Perfluorobutanoic acid; PFOS: Perfluorobutanesulfonic acid; PFOA: Perfluorooctanoic acid; PFPeA: Perfluoropentanoic acid; P&PCPs: Pharmaceutical and personal care products; PFSM: Perfluoroalkyl sulfonamide; SPE: Solid-phase extraction; UPFCA: Unsaturated PFCA; PFHxA: Perfluorohexanoicacid; HPFESA: Hydro substituted perfluoroalkyl ether sulfonate; PFAS: Poly & Per fluoroalkyl substances; UPFA: Unsaturated perfluoroalkyl alcohol [84].

Contaminant category in Biosolid.	Contaminant Detected	Research Location/year	Limit of Detection/Limit of Quantitation (LoD/LoQ)	Biosolid Sample-specific Treatment	Analytical Technique	Amount of Contaminant detected/Result
1. P&PCPs.	*Ciprofloxacin. * Diclofenac. * Enrofloxacin.	Brazil/2021	No data.	*Sample centrifugation at 14000 rpm. *Extraction with methanol and n-hexane, methanol in formic acid. * Filtration at 0.45 mm.	UPLC-ESI-MS/MS.	*10.9-158.4 µg/kg. * 13.345µg/kg. *45 µg/kg.
	*Pharmaceutical 24 active Compounds.	Portugal/2021	LOD-1:66-10.5 ng/g. LOQ-5:49-34.5 ng/g.	*Accelerated solvent extraction (ASE) of dried samples. Solid-phase extraction and extract evaporation under a nitrogen gas stream.	LC-MS/MS.	*Each contaminant has a mean conc. in the range of 0-70.8 ng/g.
	*Triclosan.	USA/2012.	No data.	*Solvent extraction of the sample with 50:50 ratio of dichloromethane (DCM): n-hexane mixture and homogenized with sodium sulfate and spiked with a standard. *Solid phase extraction conditioned with 10 mL of 50:50 DCM: hexane. *Further elution with DCM: hexane * Samples spiked with recovery standard, extracts reconcentrated, and solvent-switched to methanol under a gentle nitrogen gas stream.	GC/ECNI-MS.	*490-13,866 ng/g.
	*Pharmaceutical 42 active Compounds.	Czech Republic/2021	LOD-0:3-5.4 ng/g. LOQ: 1-12 ng/g.	*Sonication (sound) extraction of the sample with 6 ml of methanol/water mixture i.e. 0.5% HCOOH and 0.1% Na_2EDTA	UPLC-ESI-MS/MS.	*Each compound has a meanconc. in the range of 2.2 to 3.8 µg/g.
	*Pharmaceutical 35 active Compounds.	South Korea/2021.	LOD-0:1-10 ng/L. LOQ-0:5-20 ng/L.	*Supernatant centrifugation at 13000g for 10 min and 200ml, 5-sulfosalicylic acid added and centrifuged at 2 ◦C at 16,000 g for 10 min. * Filtered samples spiked with 80 mL of citric acid buffer mixture and 100 mL of standard solution. *Solid phase extraction with 5 mL methanol and 10 mL deionized water, samples loaded into the cartridge for approximately 1h and then eluted with alkaline/acidic solutions.	GC-MS/MS.	Mean conc. of all compounds is 12000 ng/L.
	*Pharmaceutical 35 active compounds.	Italy/2021.	LOD-0:11-14.38 ng/g. LOQ- 0:21 -47.92 ng/g.	* Homogenized and spiked (with standard) sample was solvent extracted (accelerated extraction) with water: methanol mixture at a 1:1 ratio and then solid-phase extraction was carried out for purification.	HPLC-MS/MS.	Mean conc. of each compound in the range of <LOQ- 4889 ng/g.
	*Bisphenol A. *Estradiol. *Triclosan. *Pharmaceutical 27 active compounds.	Czech Republic/2021.	No data.	*Purified samples extracted with ethyl acetate were evaporated in a mixture of dimethylformamide and trimethylsilylated. *Samples were then reconstituted in 800 ml ethyl acetate and 100 ml standard stock solution. * P&PCPs were extracted by ASE using methanol at 80 °C temp. and 10.3 MPa pressure.	GC-MS. LC-MS/MS.	*1023 ng/g. *29 ng/g. *2236 ng/g. *Mean conc. of eachcompound in the range of 0.1 to 50 ng/g.
	*Aminoacetophenone-2, 3. *Dimethyl naphthalene-1, 4-dione.	China/2021.	No data	*Sample extraction using acetone/n-hexane mixture at 1:1 (vol/vol) ratio with n-hexane at 100 °C and 10.35 MPa. *Spiked (with 50µl surrogate standard solution) eluted mixture of n-pentane, DCM, and methanol used.	GC-MS.	0.3 mg/kg dw 0.1 mg/kg dw 1750-6358 ng/g

2. Flame Retardants.	Polybrominated diphenyl ethers.	USA/2012.	No data.	*Solvent extraction of the sample with a 50:50 ratio of dichloromethane (DCM): n-hexane mixture and homogenized with sodium sulfate and spiked with a standard. *Solid phase extraction conditioned with 10 mL of 50:50 DCM: hexane and the elute treated with 1 mL concentrated sulfuric acid *3 mL aliquots of hexane were used to extract the mixture in triplicate and samples were spiked with recovery standard.	GC/ECNI-MS.	*0.17 ng/g.
	*2,4,6-tribromophenyl allyl ether (TAE). *2,3-dibromopropyl tribromophenyl ether (DPTE). *1,2- bis (2,4,6-tribromophenoxy) ethane (BTBPE). *Bis (2-ethylhexyl)-3,4,5,6-tetrabromophthalate (BTBP). *Decabromodiphenyl ethane (DBDPE).	China/2020.	No data.	*Water samples were filtered through a GF/C 1.2 mm grade glass filter. *3x200ml DCM solvent was used to extract the filtered sample spiked with 5 ng of surrogate standard. *Activated copper powder was used to remove the sulfur-containing compounds. *The extract was fractionated and cleaned on a 5% Al_2O_3 column deactivated with water. The Column was packed with 2g of anhydrous sodium sulfate, 8g, 5% water-deactivated Al_2O_3, and an additional 2g of anhydrous sodium sulfate.	GC/ECNI-MS.	*0.47 ng/g. *0.65 ng/g. *8.9 ng/g. *184 ng/g.
	*2,4,6-tribromophenyl allyl ether (TAE). *1,2- bis(2,4,6-tribromophenoxy) ethane (BTBPE). *Bis (2-ethylhexyl)-3,4,5,6-tetrabromophthalate	China/2020.	No data.	*Sample was eluted with a solution containing 30mL n-hexane and a 30mL mixture of n-hexane and DCM at a 3:1 ratio (vol/vol).	GC/ECNI-MS.	*0.05-0.07 ng/L. *0.11-0.14 ng/L. *0.68-1.0 ng/L.
	(TBPH). *Decabromodiphenyl ethane (DBDPE).					*9.9-18.0 ng/L.
	*Polybrominated diphenyl ethers.	China/2016.	No data.	*Surrogate standard spiked sample was filtered through 0.45 µm, millipore glass filter and then extracted by liquid-liquid extraction process with DCM.	GC/ECNI-MS.	*503 ng/g.
	*Decabromodiphenyl ethane (DBDPE).			*Oscillation extraction was carried out with 10mL hexane/ethyl acetate mixture at a 1:1 ratio (vol/vol) and soxhlet extraction was carried out with 120ml DCM for 24.		*33.86-607.32 ng/g.
	*Tris (2-chloro-isopropyl) Phosphate. *Triphenyl phosphate. *Triethyl phosphate.	China/2021.	No data.	*Sample extraction using acetone/n-hexane mixture at 1:1 ratio (vol/vol) with n-hexane at 100 °C and 10.35 MPa. *Sample was eluted with a mixture of n-pentane, DCM, andMethanol and spiked with 50µL standard.	GC-MS.	*19-70 mg/kg dw. *14 mg/kg dw. *39-269 mg/kg dw.
3. PFAS.	*PFAS 32 active compounds.	Czech Republic/2020.	LOD-0:01-0.25 ng/ml. LOQ- 0:05-0.7 ng/ml.	*ASE treatment was carried out on dried samples followed by solid-phase extraction and nitrogen stream extract evaporation.	LC-MS/MS.	Mean conc. was in therange of 5.6 to 963.2 ng/g.
	*PFBS. *PFHxS. *PFOS. *PFBA. *PFPeA. *PFHxA. *PFHpA. *PFOA.	Australia/ 2021.	LOD-0:01-0.25 ng/ml. LOQ- 0:05-0.7 ng/ml.	*ASE treatment was carried out on dried samples followed by solid-phase extraction and nitrogen stream extract evaporation.		*0.002 mg/kg. *0.0006 mg/kg. *0.0148 mg/kg. *0.0020 mg/kg. *0.0023 mg/kg. *0.0037 mg/kg. *0.0013 mg/kg. *0.0086 mg/kg.
	*Perfluorinated acids (C_3-C_{14} PFAs).	China/2012.	LOD-0:01-0.25 ng/ml.	*Freeze-dried sample (24 hrs freeze-drying) at 50 °C was homogenized	HPLC-MS/MS.	Mean conc. in

			LOQ- 0:05-0.7 ng/ml.	by sieving, through a 60-mesh stainless steel sieve followed by sonication solvent extraction, solid-phase extraction, and dispersive carbon adsorbent cleanup.		the range of 126-809 ng/g dw.
	*PFAS 44 active compounds.	Australia/ 2021.	LOD-0:01-0.25 ng/ml. LOQ- 0:05-0.7 ng/ml.	*Freeze-dried samples were homogenized, grounded, and then, spiked with 4.65mL, 10 mM NaOH methanol solution, and 25ng isotopic PFAS followed by sonication solvent extraction. *Filtration was carried out on samples to which 100mL glacial acetic acid, 100mg C18 compound, and 50mg PSA were added to remove interfering compounds.	LC-MS/MS.	Mean conc. of all compounds was 260 ng/g dw.
	*PFAS 13 active compounds.	Canada/2020.	LOQ-2:32-38.8 ng/g.	*Solid samples were spiked with surrogated standards.	LC-MS/MS.	Mean conc. of each compound was in the range of 3.3-15 ng/g dw.
	*PFAS 13 active compounds.	Canada/2020.	LOQ-2:06-22.9 ng/g.	*Samples were extracted with 15ml 3% acetic acid, 5ml 0.3% methanolic ammonium hydroxide, and 15ml 0.3 % methanolic ammonium hydroxide, followed by solid-phase extraction. *Samples were then washed with 50% 0.1M formic acid solution and 50% methanol and followed by elution with 0.3% methanolic ammonium hydroxide spiked with recovery standards.	LC-MS/MS.	Mean conc. of each compound was in the range of 3.8-21 ng/g dw.
	*PFOA. *PFOS.	Italy/2021.	LOD-0:11 ng/g. LOQ- 0:36 ng/g. LOD-0:34 ng/g. LOQ- 1:12 ng/g.	*Sample spiked with the standard was homogenized followed by ASE with water: methanol mixture at a 1:1 ratio. before solid-phase extraction.	HPLC-MS/MS.	*2.5-3.4 ng/g.
4. Surfactants	*Linear alkyl benzene Sulfonates.	South Korea /2021.	No Data.	*Samples were centrifuged at 10,000rpm for 3min to form pellets and then stored at -80 °C.	CHEMets test kits.	*18.9-22.4 ng/g. *0.6-4.5 mg/L.
	*Linear alkylbenzene Sulfonates.		No Data.	*Samples were extracted using microwave-assisted extraction (LAS) with 25 mL methanol.	HPLC.	*1.27-8.06 g/kg.
	*Linear alkylbenzene sulphonates.	Spain/2012.	LOD: 500 ng/L. LOQ: 1000 ng/L.	*Samples were filtered using a 0.45 mm glass filter followed by the addition of surrogated standard and elution with methanol.	HPLC-FD.	*2,166,667 ng/L.
	*Alkyl benzyl ammonium chlorides compounds.	Austria/2007.	LOD: 11-23 ng/L. LOQ: 6-11 ng/L.	*Liquid-liquid extraction was carried out on the samples using chloroform before evaporation and re-dissolution in $CHCl_3$ and then extraction with 4ml millipore-Q water.	LC-MS/MS.	*55,111 ng/L.
	*Dialkyl ammonium chlorides and Trialkyl ammonium chlorides.		LOD: 11-23 ng/L. LOQ: 6-21 ng/L. LOD: 12-46 ng/L. LOQ: 6-23ng/L.	*Samples were evaporated to dryness using nitrogen and made up to the final volume using methanol.	LC-MS/MS.	*68,444 ng/L.
	*Nonylphenol		LOD: 10 ng/L. LOQ: 20 ng/L.	*Acidified samples (with sulphuric acid) were filtered with a 0.45mm glass filter followed by the addition of 25ml surrogated standard and then eluted with a solution of acetone and a mixture of methanol/methyltertbutylether at a ratio of 1:9. *Evaporation of extracts was carried out with a nitrogen stream and made up to the volume of 1.0ml with acetonitrile.		*2,933 ng/L.
	*Nonylphenol mono-ethoxylate.		LOD: 20 ng/L. LOQ: 40 ng/L.			*9,167 ng/L.
	*Nonylphenol di-ethoxylate.		LOD: 10 ng/L. LOQ: 20 ng/L.			*2,300 ng/L.
	*Octylphenol.		LOD: 10 ng/L. LOQ: 20 ng/L.			*302 ng/L.

	*Sodium dodecyl sulfate. *Sodium tetradecylsulfate. *Sodium hexadecylsulfate. *Sodium octadecyl sulfate.	Spain/2020.	LOD: 0.03 ng/g. LOQ: 0.11ng/g. LOD: 2.54 ng/g. LOQ: 8.48 ng/g. LOD: 2.63 ng/g. LOQ: 8.76 ng/g. LOD: 0.03 ng/g. LOQ: 0.09 ng/g.	*Freeze-dried samples were homogenized, crushed, and sieved with a <100mm mesh size sieve followed by ultrasound-assisted extraction with a 3mL solution of methanol and glacial acetic acid before dispersive solid-phase extraction for clean-up.	LC-MS/MS and HPLC.	*2,452 ng/g. *6,210 ng/g *4,790 ng/g. *7,111 ng/g.
	C_{10}-Linear alkyl benzenes. C_{11}-Linear alkyl benzenes. C_{12}-Linear alkyl benzenes. C_{13}-Linear alkyl benzenes. C_{14}-Linear alkylbenzenes.	China/2021.	No data.	*Samples were extracted using an acetone/n-hexane mixture at a 1:1 ratio (vol/vol) with n-hexane at 100 °C and 10.35 MPa followed by elution with a mixture of n-pentane, DCM, and methanol, spiked with 50mL surrogated standard.	GC-MS.	*1-8 mg/kg dw. *1-15 mg/kg dw. *2-15 mg/kg dw. *2-16 mg/kg dw. *1 mg/kg dw

contain them (e.g. salt, seafood, tap water, honey, and bottled water) [133].

5.5. Pharmaceuticals and personal care products (P&PCPs)

P&PCPs (structure of common pharmaceuticals in biosolids are shown in Fig. 8) are a wide range of manufactured products used for medical care and cosmetic purposes and, include substances like anti-inflammatory agents, steroidal hormones, antibiotics, detergents, perfumes and active ingredients in soaps [84]. Prescription drugs used by patients for the treatment of certain diseases are not fully absorbed by the body as remnants of these drugs and their metabolites, are passed out of the body during excretion into the sewer system [134,135]. From the sewer, the remnant drugs are transported to the wastewater treatment plant and then degraded by the secondary treatment process [136]. Most of the remnant drugs are trapped in the sewage sludge after wastewater treatment while a small concentration is contained in biosolid after sludge treatment [136].

The fate and persistence of pharmaceuticals in the soil following biosolid application depend on the drug's biodegradability [84], its physicochemical properties, and the molecular structure of its constituent chemicals but not on the soil's properties or bioactivity [137]. This persistence can affect plant uptake of, and bioaccumulation of these drugs which can determine their effect on humans as a result of ingestion [136]. For example, the pharmaceutical drug Carbamazepine was found to have a limit of quantitation (LOQ) as high as 52 μg/g and 33 μg/g in radish and ryegrass respectively, in a study to compare plants' uptake of pharmaceuticals in soil; whereas the LOQ for sulfamethazine was far lower at 0.01 μg/g in the same study [136]. The effects of bioaccumulation of Carbamazepine on plants include leaf chlorosis, tissue necrosis, inability to produce fruit, etc. [138]. Phototoxicity of Sulfamethazine at high levels in plants on the other hand causes root and stem elongation, root decay, necrosis, etc. [139]. In humans, the toxic effects of Carbamazepine trigger life-threatening allergic reactions such as Toxic Epidermal Necrolysis (TEN) and Stevens-Johnson syndrome (SJS), decrease the number of blood cells produced in the body, causing unusual bleeding in the gum, nose, heavy menstruation, shortness of breath, mouth sores, etc. [140]. Also, Sulfamethazine causes anaphylaxis, angioedema, and urticaria hypersensitive reactions in the human body and can result in skin rash, drug fever, agranulocytosis, polyarthritis, and hemolytic anemia, etc. [141]. Another effect of applying pharmaceutical-contaminated biosolids on agricultural land is the development of antibiotic-resistant bacteria or ARBs in the soil [136].

Triclocarban (Fig. 9a), Triclosan (Fig. 9b) and Triclocarban (Fig. 9c) are used as antibacterial ingredients in fast-moving consumer (FMCG) or personal care products that enter the wastewater treatment system from household wastewater (kitchen and bath wastewater) because of their use in liquid and bar soaps [142], are not completely eliminated from the sludge during wastewater treatment [143]. These chemicals also called 3, 4, 4′-trichlorocarbanilide and 2, 4, 4′-trichloro-2′-hydroxydiphenyl ether respectively, whose structure are shown in Fig. 9b and (c) respectively, have a low biodegradation potential and high persistence in the soil after biosolid application [143–145]; their degradation may increase with the amount of aeration available in the soil i.e. degradation rate is faster in aerobic than in anaerobic soil [144,145]. Negative effects of TCS and TCC in the environment can be seen in algae growth inhibition [146], fauna endocrine disruption [126], toxic or carcinogenic compound (such as chlorinated anilines, dioxins, and chloroform) formation on environmental biota, and chemical bioaccumulation especially in snails and earthworms [143].

Fig. 7. Microbeads used as abrasive in toothpaste [130].

5.6. Surfactants

These are organic chemicals often called surface-active agents because they help to reduce the surface tension of liquid media [84]. They are used for a wide range of applications including soap, detergent, dish cleaner and shampoo manufacture, production of lubricants and mining flocculates, and in wastewater treatment, textile industries, and petroleum recovery operations [84]. Commonly found surfactants in biosolid-amended soil are quaternary ammonium compounds, linear alkyl benzene sulfonates, alkyl ethoxylate, alkyl phenol ethoxylates, alkyl sulfates, and alkyl ethoxy sulfates [84]. The biodegradability of certain surfactants (linear alkyl benzene sulfonates) in environmental matrices, which is dependent on their isomerization process is more rapid under aerobic conditions (with removal rate up to 99%) than anaerobic conditions [84]. Thus, linear alkyl benzene sulfonates have short-term persistence in the environment than alkyl phenol ethoxylates, which are further degraded into by-products such as nonyl and octyl phenols, increasing their environmental persistence and bioaccumulation [84,147]. The effect of benzalkonium chloride exposure is evident in soil

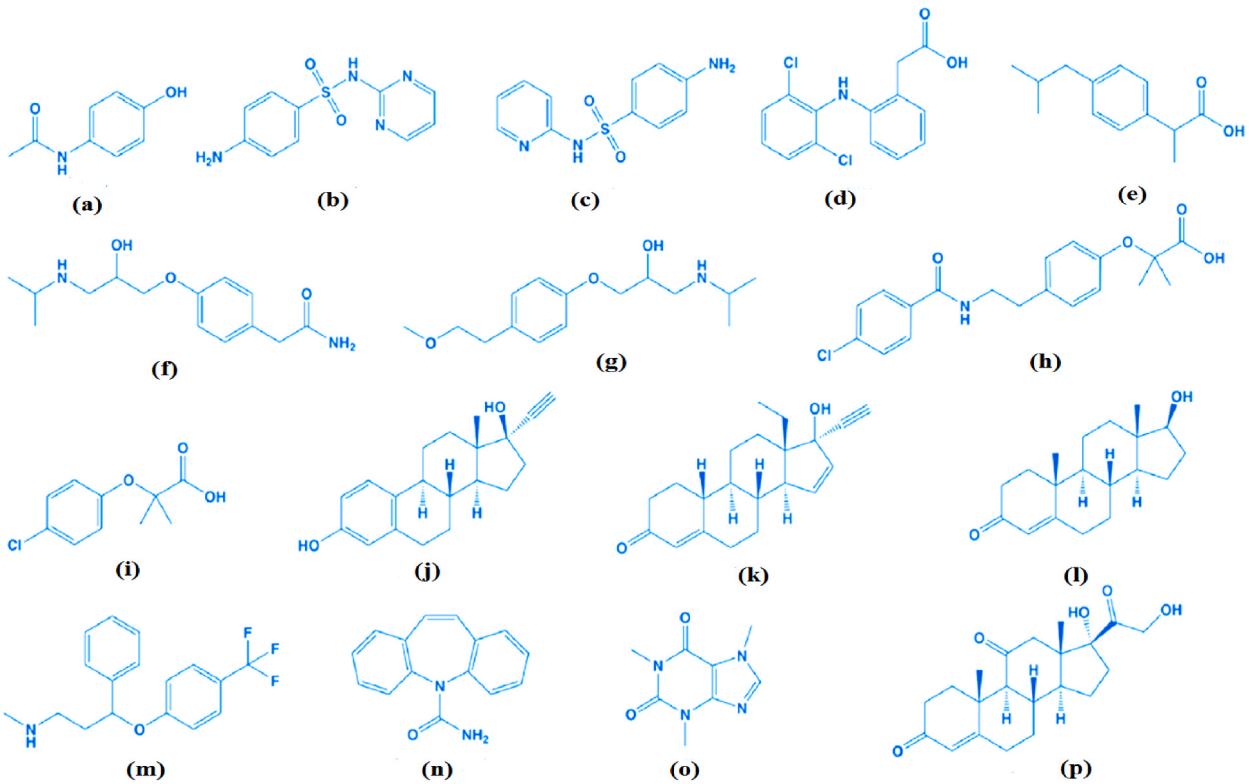


Fig. 8. Chemical structure of (a) acetaminophen (b) sulfadiazine (c) sulfapyridine (d) diclofenac (e) ibuprofen (f) atenolol (g) metoprolol (h) bezafibrate (i) clofibric acid (j) ethynylestradiol (k) gestodene (l) testosterone (m) fluoxetine (n) carbamazepine (o) caffeine and (p) cortisone which are commonly found pharmaceuticals in Biosolid [84].

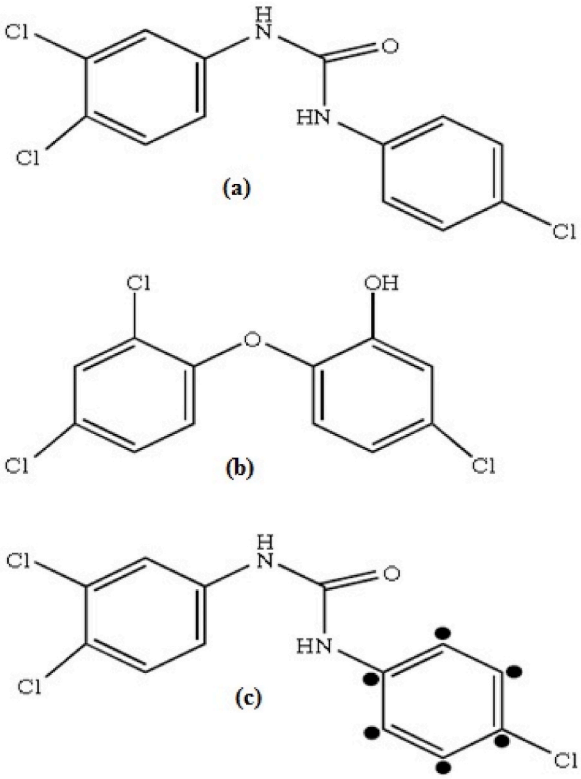


Fig. 9. Structure of (a) Triclocarban (b) Triclosan, and (c) Triclocarban [143].

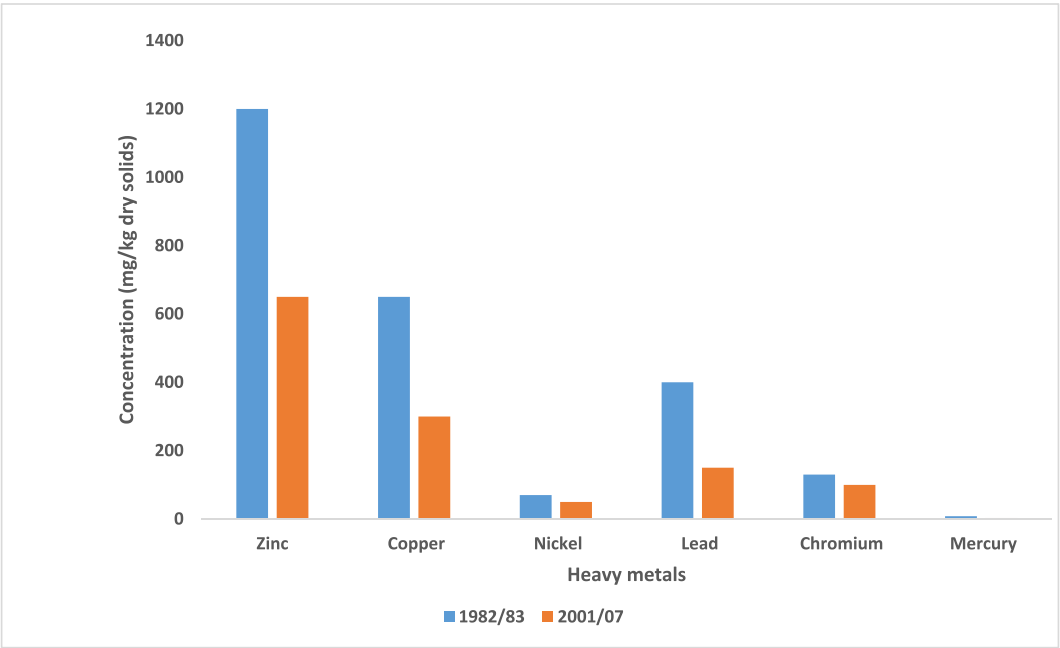


Fig. 10. Heavy metal improvement in biosolid in the UK over a period of two decades [50].

microbe resistance to antibiotics [84]. Also, alkyl phenol ethoxylate exposure causes the production of vitellogenin in male fishes at low concentrations [148] just as its by-products, nonyl, and octyl phenols have been associated with certain types of cancer and, low sperm count in men [147].

Table 3
Soil and Biosolid analyses as stipulated by The Sludge Directive [156].

S/N	Soil Analysis	Sludge Analysis
Frequency	Determined by soil mass.	Bi-annually or annually if there's no change in parameters.
Sampling method	Samples should be taken from 25 cm below the surface and not less than 10 cm minimum depth, a minimum of 25 core samples of soil over an area of 5 ha should be taken.	Wet or dry sludge samples can be used for analysis.
Parameters	To be tested should include Zn, Pb, Ni, Cd, Cu, Cr, and pH.	Should include organic matter, pH, dry matter, Zn, Pb, Ni, Cd, Cu, Cr, pH, Hg, N, and P.
Test method (s)	Atomic absorption spectrometry	Atomic absorption spectrometry, Spectrophotometry.

5.7. Flame retardants

These are lipophilic compounds used in applications such as firefighting equipment manufacture, transportation, construction, electrical/electronic industries, etc. [84]. They are designed to provide lifesaving opportunities in a disaster. For instance, they can extend personnel escape time in an industrial fire incident thus, reducing the number of casualties [84]. Flame retardants enter the wastewater treatment plants through the drainage system and because of their lipophilic (hydrophobic) nature, they are trapped in sewage sludge and reach the environment through biosolid land application, incineration, plastic, and e-waste recycling, etc. [84]. Common flame retardants such as organo-phosphate and brominated flame retardants (BFRs) [84] have long-term persistence in the soil and will undergo transformation, translocation, sorption, and root-uptake processes once in the environment [149]. They negatively affect public health and are toxic to the environment [84].

5.8. Endocrine-disrupting chemicals

These are natural (e.g. daidzein, coumestrol, and genistein found in human and animal foodstuff) or synthetic (e.g. bisphenol, perchlorate, and polychlorinated biphenyls used in manufacturing plastics, epoxy resins, lubricants, industrial solvents, pesticides, electrical equipment, and will certain pharmaceuticals) organic chemicals found in industrial/domestic wastewater, sewage sludge, and biosolids and, can impede the normal functioning of the endocrine or hormonal system [84,150]. These chemicals, which are examples of a group of PoPs found in biosolids, get into the environment via waste discharge including sewage effluent disposal and biosolid land application [151], and are extremely stable and persistent, resulting in high bioaccumulation [84]. The accumulation of endocrine disruptors (EDs) has made them ubiquitous and human exposure pathways included ingesting contaminated food, and water, breathing contaminated air, and contact with domestic/industrial chemicals [84]. Exposure to bisphenol can lead to infertility and the early onset of puberty in adolescents [84] while high levels of perchlorate impede thyroid gland iodine intake, reducing the production of hormones in the thyroid gland [97].

6. Regulations on biosolid land application

For the benefits of biosolid application in agriculture and land reclamation to be fully realized while mitigating its adverse effects on humans and the environment, government agencies around the world have promulgated guidelines for biosolid application practices [152]. Some of these legislations are examined in the following section.

6.1. The sludge directive and regulations (directive 86/278/EEC)

Biosolid land application in the European Union (EU) is controlled by the number of heavy metals it contains which is specified in the EU regulation called The Sludge Directive or simply, The Directive, first enacted in 1986 [153]. This regulation sought to promote sustainable biosolid use in agriculture and control its land application in order to prevent harmful effects on public health and environmental contamination; since treatment and recovery of biosolid on land is one of its important uses [154,155]. Contaminants such as organic compounds and pathogens also present in biosolids, are not considered under The Sludge Directive [18]. Since the enactment of the regulation, the United Kingdom (UK) has made great improvements in the heavy metal content (measured in mg/kg dry solids) of biosolids used for land application as seen in Fig. 10, which showed that between 1982 and '83, the concentration of Zinc, Copper, Lead, and Chromium allowable in biosolid were 1200, 650, 400, and 130 mg/kg dry solids compared to more recent years between 2001 and 2007, when the allowable concentration of these metals had been scaled down to 650, 300, 150, and 100 mg/kg dry solids respectively, as a way of reducing their environmental impact [50]. The Sludge Directive specified that the soil and biosolid samples are to be analyzed (Table 3) before biosolid usage on land to limit heavy metal concentrations below maximum permissible levels in the soil (Table 4) [156].

The standard permissible limits for heavy metals in soil and biosolid samples used for agricultural applications in 1986, according to The Sludge Directive are given in Table 4; the high concentrations of metals allowable in both soil and sludge samples are noticeable from this table. Although this regulation is almost four decades since it was first enacted, it is still relevant as many other regulations on biosolid reuse have been derived from it (EU member states' individual regulations) [35], its provisions may be reviewed and updated

Table 4
The EU Council standard limit for heavy metals in soil and biosolid for agricultural applications.

Parameter	Soil (mg/kg dm)	Sludge (mg/kg dm)	Loading Amount (kg/ha/yr)
Hg	1–1.5	16–25	0.1
Cd	1–3	20–40	0.15
B	30–75	300–400	3.0
Cu	50–140	1000–1750	12.0
Pb	50–300	750–1200	15.0
Zn	150–300	2500–4000	30.0

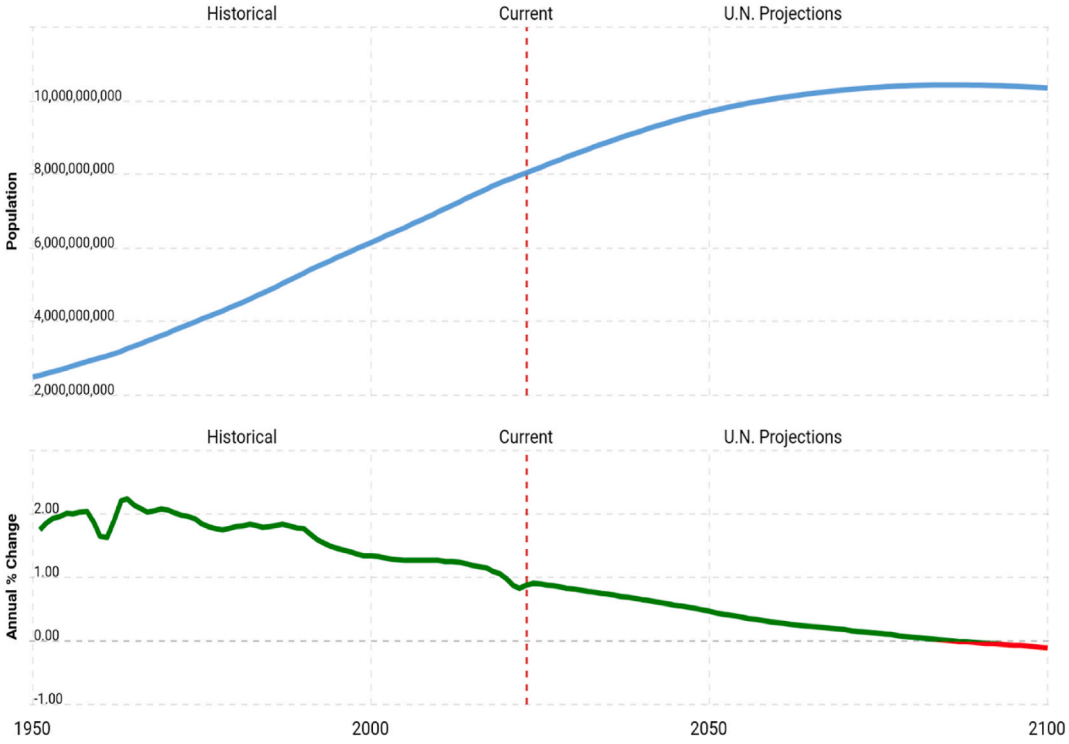


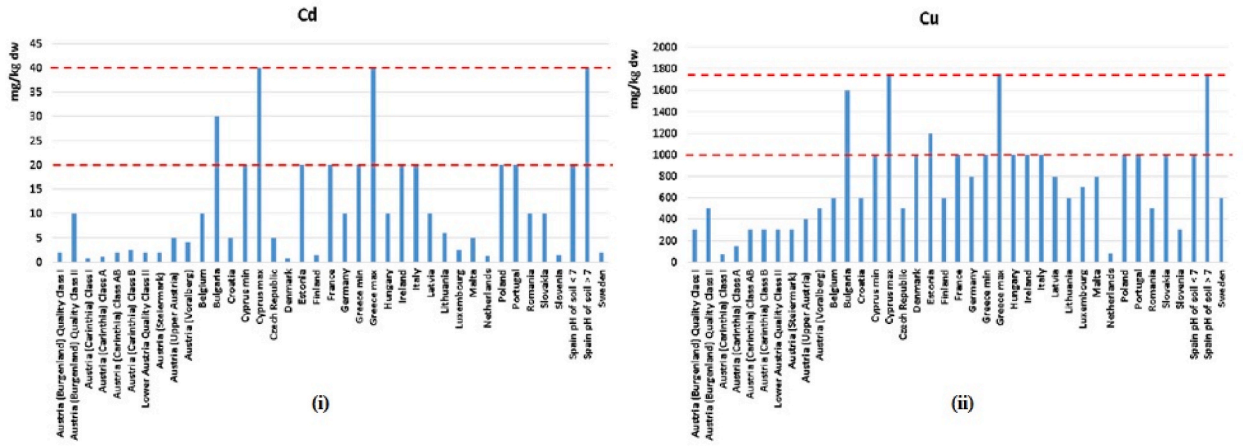
Fig. 11. World population data from 1950 and projections up to the year 2100 [157].

to meet present-day realities [155]. With new and emerging pollutants not covered by the regulation now discovered in biosolids [86–88,136,144,145], any potential environmental pollution caused by biosolid samples containing these pollutants will affect far more people in the world today, with the population in 2023 put at 8,045,311,477, at a percentage growth rate of 0.88% than in 1986 (year The Sludge Directive was passed) when the world's population was just 85,804,185 and 2.65% growth rate (Fig. 11) [157].

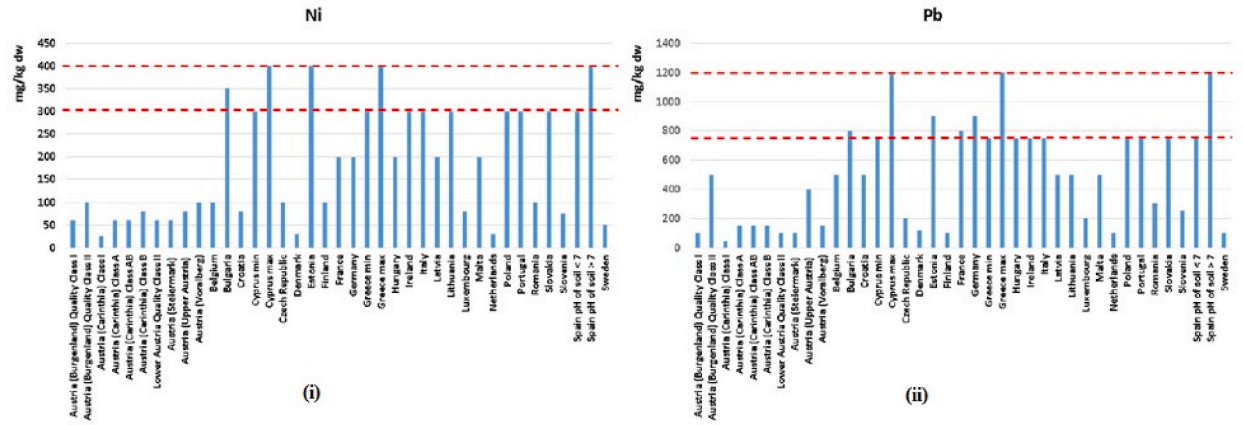
[156].

EU member states have scaled down the concentration level of heavy metals in biosolids since the enactment of The Sludge Directive, by promulgating individual national legislations on further reduction in the limits of these metals, keeping values lower than the EU Council's directive, in a bid to further protect humans and the environment (Fig. 12a, b, c, and d showed the comparison of standard limits of selected heavy metals) [35]. In Fig. 12, it can be seen that the majority of EU states including Belgium, Croatia, Denmark, Finland, Germany, Hungary, Latvia, Malta, Netherlands, Slovakia, etc., allowed a national standard limit of heavy metals much below the EU Council's lower limit, which is shown on the figure by the red dotted lines (upper and lower limits). The comparison in Table 5 is important as it gives a glance-view of the national heavy metal standard limits and organic compounds concentration between EU member states.

Permissible heavy metal constituent of biosolid shown in Table 4 (EU Council Sludge Directive, data from 1986), Table 5 (national regulation of EU member states, data from 2019) and Fig. 12 revealed a significant reduction over 3 decades, indicative of the need for lower values and thus, better environmental health. Maria et al. [18] provided the standard limit for pathogens in biosolids in selected EU states based on national legislation, which is not captured by The Sludge Directive. This is represented in Table 6. Data in Table 6 showed that most pathogen standard limits allowable in biosolids are less than or equal to 1000 CFU per gram dry matter (CFU/gDM); this has been adopted in many other states in Europe today [18].



a: Comparison of the limits of (i) Cadmium and (ii) Copper in biosolid for land application set by EU member states and that set by the EU council's Sludge Directive. Red dotted lines represent the EU Sludge Directive's lower and upper limits.



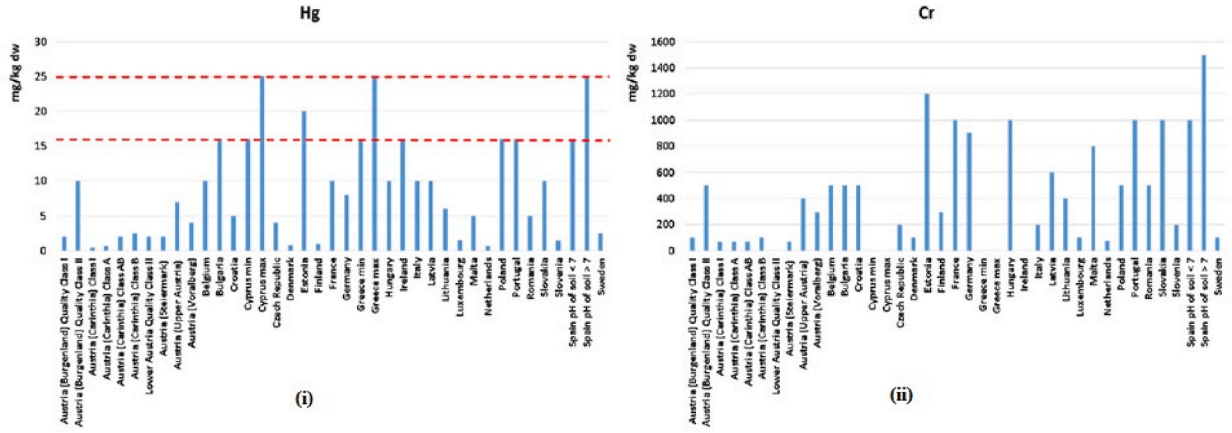
b: Comparison of the limits (in mg/kg dw) of (i) Nickel and (ii) Lead in biosolid for land application set by EU member states and that set by the EU council's Sludge Directive. Red dotted lines represent the EU Sludge Directive's lower and upper limits.

Fig. 12. a: Comparison of the limits of (i) Cadmium and (ii) Copper in biosolid for land application set by EU member states and that set by the EU council's Sludge Directive. Red dotted lines represent the EU Sludge Directive's lower and upper limits. b: Comparison of the limits (in mg/kg dw) of (i) Nickel and (ii) Lead in biosolid for land application set by EU member states and that set by the EU council's Sludge Directive. Red dotted lines represent the EU Sludge Directive's lower and upper limits. c: Comparison of the limits (in mg/kg dw) of (i) Mercury and (ii) Chromium in biosolid for land application set by EU member states and that set by the EU council's Sludge Directive. Red dotted lines represent the EU Sludge Directive's lower and upper limits. d: Comparison of the limits (in mg/kg dw) of (i) Zinc and (ii) Arsenic in biosolid for land application set by EU member states and that set by the EU council's Sludge Directive. Red dotted lines represent the EU Sludge Directive's lower and upper limits [35].

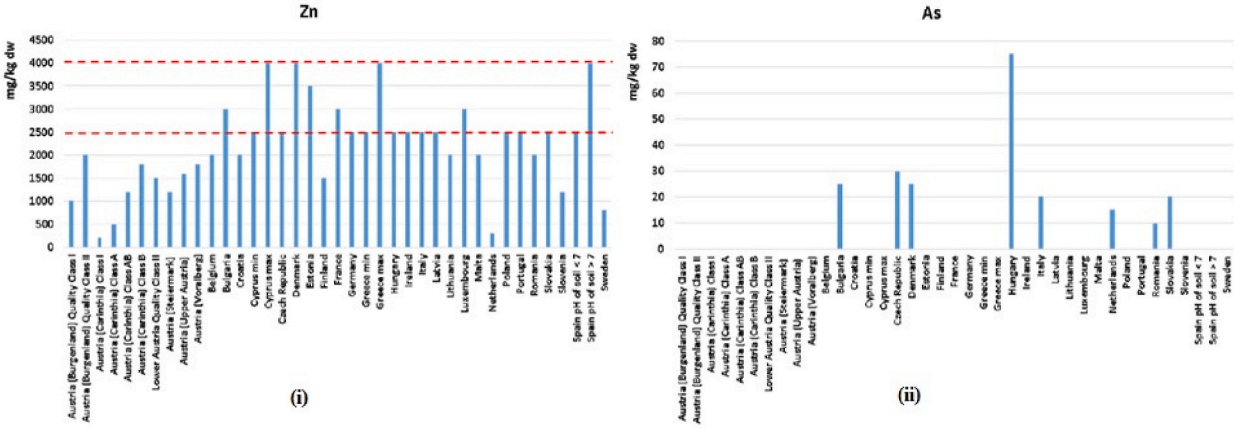
6.2. *The 40 CFR part 503 regulation*

The United States Environmental Protection Agency (USEPA) developed a regulation titled Standards for the Use of Sewage Sludge (popularly referred to as the Part 503 Rule, enacted in 1993), as part of the Clean Water Act of the U.S. Congress [158]. The Rule specifies general minimum requirements for applying biosolids on land for agronomic applications in the United States [159,160]. According to the Part 503 Rule, biosolids used for agricultural applications must meet the ceiling concentration (maximum limit) for heavy metal content before usage; this is given in Table 7.

The 40 CFR Part 503 Rule specifies pathogens reduction in two biosolid categories, Class A and Class B biosolids. The Rule specifies that pathogens in Class A biosolids be reduced below detectable concentrations [160]. In Class B biosolids, however, pathogens are reduced to concentrations unlikely to pose threats to the environment and public health [160]. There are, however, new pathogens (hepatitis A virus and adenovirus) with heat-resisting capabilities that have been detected in biosolids since the enactment of the Part 503 Rule in 1993 [161]. The general requirements for land application of biosolids based on the Part 503 Rule specified that the supplier of biosolid must provide the user with a guide so as to comply with the requirements of Table 7 and the user, on the other hand, must apply biosolids to meet these requirements [159,162]. The supplier must notify government authorities in writing, about



c: Comparison of the limits (in mg/kg dw) of (i) Mercury and (ii) Chromium in biosolid for land application set by EU member states and that set by the EU council's Sludge Directive. Red dotted lines represent the EU Sludge Directive's lower and upper limits.



d: Comparison of the limits (in mg/kg dw) of (i) Zinc and (ii) Arsenic in biosolid for land application set by EU member states and that set by the EU council's Sludge Directive. Red dotted lines represent the EU Sludge Directive's lower and upper limits [35].

Fig. 12. (*continued*).

the location and time of application, and provide contact information while the user, must notify authorities before the initial application of bulk biosolids [159].

The majority of literature reviewed for this study agreed that The Sludge Directive (i.e. Directive 86/278/EEC) regulations and the Part 503 Rule formed the basis for biosolid use on land in most countries around the world including developing countries [153,154, 160–162]. Iranpour et al. [163], compared the standards (Table 8) specified by both regulations and, a national standard of the Netherlands in a review study in which values for heavy metals were normalized against the U.S. pollution standard limit. These pictorials clearly showed that the national limit of the Netherlands is much lower than EU or U.S limits., further underscoring efforts by EU member states to reduce national pollutant standards for better protection of the environment. Also, Table 8 showed that concentration limits in the EU are expected to reduce significantly, over a long-term period [163].

[163].

According to Iranpour et al. [163], the EU Council proposed a reduction in heavy metal concentration in biosolids over a 10-year period in line with member states' leanings. Iranpour and co-researchers compared these proposed (projected) standards with mean values from the U.S. EPA National Sewage Sludge Survey (NSSS), Pennsylvania, and Los Angeles Surveys over a given period (Table 9). The authors observed a general decline in the value of heavy metals except for Pennsylvania (95th percentile) which has the highest values in spite of the average. They concluded that if the U.S. is to adopt EU proposed limits, new technologies (such as adsorption or precipitation from a suitable porous solid) must be developed to further reduce pollutant concentrations in biosolids.

7. Discussion

In terms of agronomic usage and as a fit for the circular economy, biosolid has had beneficial applications, whether in agricultural land application or as a base material for the manufacture of construction materials. It has been used as organic fertilizer (due to their high organic matter and nutrient content, which is slowly released to plants, making biosolids better than chemical fertilizers in

Table 5
Comparison of the standard limits of heavy metals and organic compounds in biosolid among EU states with the EU Council's Sludge Directive.a, b, c, d, etc. represents values from different categories measured. PCDD/F: polychlorinated dibenzo-p-dioxins and furan; AOX: absorbable organic halogens; DEHP: di (2-ethylhexyl) phthalates; NP: nonylphenols; PCB: polychlorinated biphenyls; NPE: nonylphenol ethoxylates; PAH: polycyclic aromatic hydrocarbons; LAS: linear alkylbenzene sulfonates [18].

Legislation	Heavy Metals								Organic Compounds						
	Cd	Cr	Cu	Hg	Ni	Pb	Zn	As	PCB	AOX	LAS	DEHP	NP/NPE	PAH	PCDD/F
Directive 86/278/EEC	20–40	-	1000–1750	16–25	300–400	750–1200	2500–4000	-	-	-	-	-	-	-	-
EU-15															
Germany	10	900	800	8	200	900	4000	-	0.1 [o]	400	-	-	-	-	100
UK	-	-	-	-	-	-	-	2	-	-	-	-	-	-	-
Spain [a]	20–40	1500	1000–1750	16–25	300–400	750–1200	2500–4000	-	-	-	-	-	-	-	-
France	20	1000	1000	10	200	800	3000	-	0.8 [l]	-	-	-	-	2–5 [p]	-
Italy	20	200	1000	10	300	750	2500	20	0.8	-	-	-	-	6	25
Netherlands	1.25	75	75	0.75	30	100	300	15	-	-	-	-	-	-	-
Austria [b]	2–10	50–500	70–500	0.4–10	25–100	45–500	200–2000	20	0.2–1	500	-	-	-	6	50–100
Sweden [c]	0.75	40	300	1.5	25	25	600	-	0.4 [l]	-	-	-	50	3 [f]	-
Portugal	20	1000	1000	16	300	750	2500	-	0.8	-	5000	-	450	6	100
Finland	1.5	300	600	1	100	100	1500	25	-	-	-	-	-	-	-
Denmark	0.8	100	1000	0.8	30	120	4000	25	0.2 [l]	-	1300	50	10	3 [f]	-
Ireland	20	-	1000	16	300	750	2500	-	-	-	-	-	-	-	-
Greece [d]	20–40	500	1000–1750	16–25	300–400	750–1200	2500–4000	-	-	-	-	-	-	-	-
Belgium [b]	6–10	100–150	600–800	1–1.6	100	300–500	1500–2000	20–150	0.6–0.8 [l]	-	-	-	-	3–20	20
Luxembourg [d]	2.5	100	700	1.6	80	200	3000	-	0.2 [m]	-	-	-	-	20 [i]	20
EU-13															
Poland	20	500	1000	16	300	750	2500	-	-	-	-	-	-	-	-
Hungary	10	1000	1000	10	200	750	2500	75	1 [l]	-	-	-	-	10 [i]	-
Czech Republic	5	200	500	4	100	200	2500	30	0.6 [l]	500	-	-	-	10 [j]	-
Romania	10	500	500	5	100	300	2000	10	0.8 [l]	500	-	-	-	5 [k]	-
Lithuania [e]	1.5–20	140–400	75–1000	1–8	50–300	140–750	300–2500	-	-	-	-	-	-	-	-
Slovakia	10	1000	1000	10	300	750	2500	20	0.8 [l]	500	-	-	-	6 [h]	-
Bulgaria	30	500	1600	16	350	800	3000	25	-	-	-	-	-	-	-
Estonia	20	1000	1000	16	300	750	2500	-	-	-	-	-	-	-	-
Cyprus [d]	20–40	-	1000–1750	16–25	300–400	750–1200	2500–4000	-	-	-	-	-	-	-	-
Latvia [f]	2–10	100–600	400–800	3–10	50–200	150–500	800–2500	-	-	-	-	-	-	-	-
Slovenia	1.5	200	300	1.5	75	250	1200	-	-	-	-	-	-	-	-
Malta	5	800	800	5	200	500	2000	-	-	-	-	-	-	-	-
Croatia	5	500	600	5	80	500	2000	-	0.2 [n]	-	-	-	-	-	100

preventing the leaching of nutrients to surface water), as soil amender, and as a land repairer. Although biosolids are treated before land application, they still contain low concentrations of harmful emerging and persistent organic pollutants (PoPs) (Table 2) including per- and poly-fluoroalkyl substances (PFAs), heavy metals, P&PCPs, and microplastics, etc., which can have hazardous effects on humans, animals, and the environment at high concentrations without controlled application of biosolids. Some of the effects in the soil include reproductive defects, neurological development delays, low birth weight, bone variation, accelerated puberty, changes in liver and kidney functions, thyroid disease, and testicular cancer caused by PFAs; algal invasion, endocrine disruption, carcinogenic and toxic compound formation such as chlorinated anilines, dioxins, and chloroform, and, chemical bioaccumulation in snails and earthworms caused by the cosmetic antibacterial chemicals TCS and TCC; nervous system disorders, gastrointestinal and kidney dysfunction, immune system dysfunction, skin lesion, birth defect, vascular damage, and cancer caused by heavy metals; water eutrophication and algae invasion caused by excess soil phosphorus content; Toxic Epidermal Necrolysis (TEN) and Stevens-Johnson syndrome (SJS), reduction in red blood cell production, hemolytic anemia, anaphylaxis, etc. These effects on humans and the environment may be mitigated by biosolid land application practices (Section 4.1) which account for the concentration, fate, and persistence of pollutants in the soil and form part of government regulations.

Government regulations are effective in limiting the number of heavy metals, pathogens, and organic pollutants allowable in biosolids applied on land, imposing restrictions on sites, and fostering good agronomic application practices. The EU's Sludge Directive (Directive 86/278/EEC) and USEPA's Part 503 Rule are the two major legislations on biosolid land application from which many

Table 6
Standard limit for pathogens in Biosolids used in agriculture in the EU.1 = three regions were involved in measurement, 2 = advanced sewage sludge treatment was employed; DM: dry matter; CFU: colony-forming unit; MPCN: most probable cytopathic number u.m.: units of measure; WW: wet weight; MPN: most probable number [18].

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

countries around the world and even, EU member states have derived national biosolid legislation. A comparison of both legislations revealed that the EU's Sludge Directive specified stricter regulation, with allowable biosolid pollutant concentration values much lower than USEPA's Part 503 Rule (Table 8). Since the enactment of these major legislations (the Sludge Directive was enacted in 1986 and the Part 503 Rule was passed in 1993), there have been new and emerging contaminants detected in biosolids, making it necessary to review and update these regulations to meet current realities and challenges of today's biosolid land applications.

Updating these government regulations (Directive 86/278/EEC and the Part 503 Rule) should focus on their expansion to accommodate specific legislation on new pathogens, organic pollutants, and emerging contaminants such as microplastics and pharmaceuticals. The updated regulations should review downwards, the standard limits of heavy metals in biosolids (with a focus on USEPA's Part 503 Rule with less stringent allowable standard limits) in line with the national regulation of EU member states, using regulations in the Netherlands as a guide (Table 8). The new heavy metal regulations should take into account soil phosphate interaction with metals which causes metal lability in the soil due to high phosphorus content occasioned by the high agronomic loading rate of biosolid on land. Biosolid land application best practices should form an integral part of government regulatory policies on biosolid use.

Table 7
The EPA Part 503 heavy metal requirements in biosolids applied to land.

Type of Heavy Metal	Concentration limits for all biosolid types applied to land. (mg/kg)[a]	Concentration limits for exceptional quality (EQ) and pollutant concentration biosolids (mg/kg)[a]	Cumulative pollutant loading rate (CPLR) limits for biosolid (kg/ha)	Annual pollutant loading rate (APLR) for biosolids (kg/ha.yr)
Chromium	3000	1200	3000	150
Arsenic	75	41	41	2.0
Cadmium	85	39	39	1.9
Mercury	57	17	17	0.85
Lead	840	300	300	15
Copper	4300	1500	1500	75
Nickel	420	420	420	21
Molybdenum[b]	75	–	–	–
Zinc	7500	2800	2800	140
Selenium	100	36	100	5.0
Applies to:	All biosolids applied on land.	Bulk and bagged biosolids.	Bulk biosolids	Bagged biosolids[c].

ab,c = Dry mass basis measurement; molybdenum limits were deleted from the Part 503 Rule after February 1994 amendment, awaiting EPA consideration; biosolids delivered in bags or other similar containers [159].

Table 8
Comparison of heavy metal standard limits for the EU, Netherlands, and the U.S.

Heavy Metal	EU Limits					Netherland Limits	U.S. EPA Limits	
	Upper Limits	Lower Limits	Short-term Limits	Medium-term Limits	Long-term Limits		Ceiling Concentration	Limit Concentration
Chromium	–	–	1000	800	600	75	–	–
Arsenic	–	–	–	–	–	15	75	41
Cadmium	40	20	10	5	2	1.25	83	39
Mercury	25	16	10	5	2	0.75	57	17
Lead	1200	750	750	500	200	100	840	17
Copper	1750	1000	1000	800	600	75	4300	1500
Nickel	400	300	300	200	100	30	420	420
Molybdenum	–	–	–	–	–	–	75	–
Zinc	4000	2500	2500	2000	1500	300	7500	2800
Selenium	–	–	–	–	–	–	100	100

Table 9
Comparison of EU projected heavy metal limits in biosolid to U.S. current average values. NSSS = National Sewage Sludge Survey; TITP: Terminal Island Treatment Plant [163].

	Proposed Ceilings in EU		U.S. Sewage Sludges and Biosolids		
Pollutant	Year 2015	Year 2025	NSSS 1988 (mean)	Pennsylvania, 1996–97 (median / 95th percentile)	TITP, 2001, Los Angeles, CA (mean)
Arsenic	–	–	9.9	3.6/18.7	11.5
Cadmium	5	2	6.9	2.26/7.39	3.2
Chromium	800	600	119	35.1/314	–
Copper	800	600	741	511/1382	265
Lead	500	200	134.4	64.9/202	59
Mercury	5	2	5.2	1.54/6.01	3
Molybdenum	–	–	9.2	8.18/36	25
Nickel	200	100	42.7	22.6/84.5	46.5
Selenium	–	–	5.2	4.28/8.47	58
Zinc	2000	1500	1202	705/1985	964

8. Conclusion and future perspectives

This review study examined the fate of contaminants in biosolids used for land application and the potential effects of this biosolid management practice on humans and the environment while discussing government policies regulating land application. Although there are other methods (including incineration and energy recovery, landfilling, land reclamation, etc.) employed to dispose of the increasing quantities of biosolids generated from large volumes of wastewater treated daily due to an increasing world population, land application on agricultural farms has been adjudged the most economical and sustainable disposal route aside using biosolid as a

base material for the manufacture of construction materials. Land application of biosolids is regulated by the government due to the negative effects of its contaminants and the need to protect the environment. Two major biosolid regulations, the EU's Sludge Directive, and USEPA's Part 503 Rule were identified. These regulations require a review and an update, with a focus on USEPA's Part 503 Rule with less stringent biosolid regulations, to meet the current challenges of biosolid land application. The update should include specific regulations on new and emerging contaminants such as microplastics, pharmaceuticals, pathogens, organic pollutants, and other persistent organic pollutants (PoPs) in biosolid, further reduction of heavy metal standard limits (which should be lower than the projected EU regulation in Table 9), and consideration of soil phosphate-metal interactions to regulate biosolid agronomic loading rate. Future biosolid research should focus on the concentration of TCS, TCC, and newly discovered pharmaceuticals, as well as Microplastic transport in biosolid-amended soils, soil-plant transfer mechanism, and metabolism of PFAs in soils, all of which should inform government policies on biosolid application on land.

Author contribution statement

All authors listed have significantly contributed to the development and the writing of this article.

Data availability statement

Data will be made available on request.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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References

[1] J. Boyle Michael, D. Jasperson Hans. How Does Industrialization Lead to Urbanization?, The Investopedia Team, 2021. Available at: https://www.investopedia.com (Accessed 14 February 2023.
[2] David Satterthwaite, McGranahan Gordon, Cecilia Tacoli, Urbanization and its Implication for Food Farming, vol. 365, Philosophical Transaction of the Royal Society, 2010, pp. 2809–2820, https://doi.org/10.1098/istb.2010.0136.
[3] The World Bank, Understanding poverty: urban development, The International Bank for Reconstruction and Development (IBRD) and, International Development Association (IDA) (2022). Available at: https://www.worldbank.org. (Accessed 14 February 2023).
[4] Ferronado Navarro, Vincenzo Torretta, Waste management in developing countries: a review of global issues, Int. J. Environ. Res. Publ. Health 16 (6) (2019) 1060, https://doi.org/10.3390/ijerph16061060.
[5] USEPA, Guide for industrial waste management, United States Environmental Protection Agency (2015). Available at: https://www.epa.gov/epaoswer/non-hw/industd/index.htm. (Accessed 14 February 2023).
[6] I. Kanu, Ome Achi, Industrial effluents and their impact on water quality of recurring rivers in Nigeria, Journal of Applied Technology in Environmental Sanitation 1 (1) (2011) 75–86.
[7] Marcos V. Sperling, Alliance House, 12 Caxton Street, Wastewater Characteristics, Treatment, and Disposal, vol. 1, Iwa Publishing, London SW1HOQS, UK, 2007.
[8] Robert Edwin White, Silvana I. Torri, Rodrigo Stuart Correa, Biosolids soil application: agronomic and environmental implications, 10.1155/2011/928973-biosolid-soil-application, Applied and Environmental Soil Science (2011), 928973.
[9] Shahrzad Kabirinejad, Mehran Hoodaji, The effect of biosolids application on soil chemical properties and zea mays nutrition, 1:4, Kabirinejad and Hoodaji International Journal of Recycling of Organic Wastes in Agriculture (2012), https://www.ijrowa.com/content/1/1/4.
[10] Gabriel Di Giacomo, Pietro Romano, Evolution and Prospects in Managing Sewage Sludge. Environment Agency Strategy for Safe and Sustainable Sludge Use (Policy Paper), UK Govt. Publication, 2020.
[11] Manzoor Quadir, Pay Drechsel, Blanca Jimenez Cisneros, Youngy Kim, Pramanik Amit, Praem Mehta, Oluwabusola Olaniyan, Global and regional potential of wastewater as a water, nutrient and energy source, Nat. Resour. Forum 44 (1) (2020) 40–51, https://doi.org/10.1111/1477-8947.12187.
[12] Yecui Hu, Shuang Pang, Junjie Yang, Xiaoguang Zhao, Jirong Cao, Changes in soil microbial community structure following amendment of biosolids for seven years, 2019, Environmental Pollutions and Bioavailability 31 (1) (2019) 24–31, https://doi.org/10.1080/26395940.2019.1569478.
[13] Thomas A. Burke, Biosolids Applied to Land: Advancing Standards and Practices, National Academies of Sciences, Engineering, and Medicine, National Academies Press, Washington D.C, 2002. https://nap.nationalacademies.org/10426.
[14] Augusto Blanchini, Marco Pellegrini, Cesare Sarcani, Sewage Sludge Management in Europe: A Critical Analysis of Data Quality, 2016, Int'l Journal of Environment and Waste Management, 2016, https://doi.org/10.1504/IJEWM.2016.10001645.
[15] Victor E. Akpan, David O. Omole, Daniel E. Bassey, Assessing the public perceptions of treated wastewater reuse: opportunities and implications for urban communities in developing countries, Heliyon 6 (10) (2020), e05246, https://doi.org/10.1016/j.heliyon.2020.e05246.
[16] A.J. Englande Jr., Peter Krenkel, J. Shamas, Waste Treatment and Water Reclamation, Encyclopedia of Physical Science and Technology, 2015, pp. 639–670.
[17] Sana Khalid, Muhammad Shahid, et al., A review of environmental contamination and health risk assessment of wastewater use for crop irrigation with focus on low and high income countries, International Journal of Environmental Resource and Public Health 15 (5) (2018) 895, https://doi.org/10.3390/ijerph15050895.
[18] Maria M. Collivignarelli, Allessandro Abba, Andrea Frattarola, Marco C. Mino, Sergio Padovani, Joannis Katsoyannis, Vincenzo Torretta, Legislation for the reuse of biosolids on agricultural land in Europe: overview, Sustainability 219 (11) (2019) 6015.
[19] Hoi Yan Chow and Min Pan, Fertilization value of biosolids on nutrient accumulation and environmental risks to agricultural plants, 2020, Journal of Water, Air, and Soil Pollution 231 (2020) 578, https://doi.org/10.1007/s//270.020.049468.

[20] Javier Mateo-Sagasta, Liqa Rasehid Sally, L. Anne, Thebo, Global wastewater and sludge production, treatment, and use, Researchgate (2015), https://doi.org/10.1007/978-94-017-9545-6_2.
[21] Avram Karagiannidis, Petro Samaras, Themistoklis Kasampalis, Evaluation of sewage sludge production and utilization of Greece in the frame of integrated energy recovery, Desalination and Water Treatment, Researchgate (2011), https://doi.org/10.5004/dwt.2011.2613.
[22] Yuting Liang, Ling Ye, Bo Yue, Bang-bang Meng, Xusheng Yuan, Bibliometric analysis of related literatures on sludge disposal technologies, 2022, IOP Conf. Ser. Earth Environ. Sci. 1035 (2022), 012015, https://doi.org/10.1088/1755-1315/1035/1/012015.
[23] E. Corcoran, E. Nellemann, R. Baker, D. Bos, H.S. Osborn, Sick water? The central role of wastewater management in sustainable development – a rapid response assessment. United nations environment programme, UN-habitat, grid, arendal. https://www.grida.no, 2010.
[24] Eurostat, Population Connected to Wastewater Treatment Plant (online) (2014). Available at: https://www.eea.europa.eu/data-and-maps/data/external/population-connected-to-wastewater-collection. September 2015.
[25] Martyna Buta, Jaku Hubeny, Wiktorzie Inski, Monika Harnisz, Ewa Korzeniewka, Sewage sludge in agriculture: the effects of selected chemical pollutants and emerging genetic resistance determinants on the quality of soil and crops- A review, Ecotoxicol. Environ. Saf. 214 (2021), 212070.
[26] A. Hamood, J.M. Khatib, Sustainability of Sewage Sludge in Construction, Sustainability of Construction Materials, Science Direct, 2016, pp. 308–415 (Chapter 24).
[27] Krzysztof Fijalkowski, Agnieszka Rorat, Anna Grobelak, et al., The presence of contaminations in sewage sludge – current situation, J. Environ. Manag. 203 (2017), 1206-1136.
[28] Elif Yakamercan, Akif Ari, Aygun Ahmed, Land application of municipal sewage sludge: human health risk assessment of heavy metals, J. Clean. Prod. 319 (2021), 128568.
[29] Rajeev Pratap Singh, Madhoolika Agrawal, Potential benefits and risks of land application of sewage sludge, Waste Manag. 28 (2) (2008) 347–358.
[30] B.A. Maureen Reilly, The case against land application of sewage sludge pathogens, Can. J. Infect Dis. Med. Microbiol. 12 (4) (2001) 205–207.
[31] Jessica Briffa, Emmanuel Sinagra, Reynald Blundell, Heavy metal pollution in the environment and their toxicological effects on humans, Heliyon 6 (9) (2020), e04691.
[32] Alysson Ronberto Baizie Silva, Fabio Camilotti, Risks of heavy metal contamination of soil-plant system by land application of sewage sludge: a review with data from Brazil, IntechOpen (2014), https://doi.org/10.5772/58384.
[33] Serdar Koyuncu, Occurrence of organic micropollutants and heavy metals in the soil after the application of stabilized sewage sludge, Journal of Environmental Health Science and Engineering 20 (1) (2022) 385–394.
[34] J. Aparicio, J.L. Santos, E. Alonso, Limitation of the concentration of organic pollutants in sewage sludge for agricultural purpose: a case study of south Spain, Waste Manag. 29 (5) (2009) 1747–1753.
[35] Andrea Gianico, M. Camilla, Braguglia, et al., Land application of biosolids in Europe: possibilities, constraints, 2021, and Future Perspective Water 13 (2021) 103, https://doi.org/10.3390/w/3/0103.
[36] Rebecca A. Dickman, Dana S. Aga, Efficient workflow for suspect screening analysis to characterize novel and legacy per-and polyfluoroakyl substances (PFAs) in biosolids, Anal. Bioanal. Chem. 414 (2022) 4497–4507.
[37] Sara J. Lupton, Francis Casey, David J. Smith, Heldurr Hakk, Perfluorooctanoic acid transport in soil and its absorption and distribution in alfalfa (medicago sativa), J. Food Protect. 85 (1) (2022) 164–172, https://doi.org/10.4315/jfp-21-276.
[38] Muhammed Z. Hashimi, Shuhong Wang, Zulkfil Ahmed, Environmental Micro-pollutants, Elsevier, 2022, https://doi.org/10.1016/c2020-0-03749-7.
[39] Sarah M. Elliot, Melinda Erickson, Aliesha Krall, Micropollutants in Groundwater and Soil Downgradient from On-Site Wastewater Discharges, ResearchGate, 2018. https://www.researchgate.net/publication/321371850.
[40] Agnieszka Rorat, Pauline Coiurtois, Vandenbulcke Frank, et al., Sanitary and Environmental Aspects of Sewage Sludge Management. Industrial and Municipal Sludge, 2019, https://doi.org/10.1016/B978-0-12-815907-1.00008-8.
[41] Aryadeep Roychoudhury, Nilanjana Das, Sewage sludge treatment and involvement of microbes, in: Sustainable Management and Utilization of Sewage Sludge, Springer, 2022, pp. 165–181, https://doi.org/10.1007/978-3-030-85226-98.
[42] Malgorzata Kacprzak, Ewa Neczaj, Krzysztof Fijalkowski, et al., Sewage sludge disposal strategies for sustainable development, Environ. Res. 156 (2017) 39–46.
[43] Tina Lovingood, Jill Toynosky, et al., EPA Unable to Assess the Impact of Hundreds of Unregulated Pollutants in Land-Applied Biosolids on Human Health and Environment [Report], U.S. Environmental Protection Agency, 2018.
[44] Sludge Processing, Sludge Treatment (online). Benenv (2020). Available at: https://www.sludgeprocessing.com/, 15-03-2023.
[45] Veolia,), Incineration (online (2021). Available at: https://www.veoliawatertechnologies.co.uk/technologies/incineration. (Accessed 15 March 2023).
[46] SUEZ, Sludge Drying (online) (2022). Available at: www.suez.com/en/water/sludge-and-by-products-management/sludge-drying/. (Accessed 15 March 2023).
[47] Franco Mangone, Soledad Gutierrez, et al., Sludge Dewatering. 28th European Symposium on Computer-Aided Process Engineering, Computer-aided Chemical Engineering, 2018, 2018.
[48] CAMBI, How Does Thermal Hydrolysis Work (online)? (2020). Available at: www.cambi.com/what-we-do/thermal-hydrolysis/how-does-thermal-hydrolysis-work/. (Accessed 15 March 2023).
[49] Wanare Ram, Iyer Kannan, Trudeep Dave, Application of biosolids in civil engineering: state of the art, Mater. Today: Proceedings, Elsevier (2022), https://doi.org/10.1016/j.matpr.2022.04.166.
[50] UKWIR, Biosolids: Agricultural Good Practice. United Kingdom Water Industry Research Guidance Leaflet, 2014. Project 14/SL/11/7.
[51] Eliot Epstein, Land Application of Sewage Sludge and Biosolids, Lewis Publishers, CRC Press LLC, 2000NW, Corporate BLVD, Boca Raton, Florida 33431, 2003.
[52] Chad A. Kinney, Edward T. Furlong, Steven D. Zangg, et al., Survey of organic wastewater contaminants in biosolids destined for land application, Environmental Science Technology 140 (2006) 7207–7215.
[53] Savakumar Patel, Sazal Kundu, et al., A critical literature review on biosolids to biochar: an alternative biosolid management option, Revised Environmental Science and Biotechnology 202 (19) (2020) 807–841.
[54] Qin Lu, Zhenli L. He, Peter J. Stoffellla, Land Application of Biosolids in the U.S.A.: A Review, Applied and Environmental Soil Science 2014 (62) (2012), https://doi.org/10.1155/2012/201462.
[55] Gefa, Biosolid Assessment and Prepared Study (online). Georgia Environmental Finance Authority, Black and Veatch, Available at: https://www.bv.com/, 2021. (Accessed 24 February 2023).
[56] USEPA, Control of Pathogen and Vector Attraction in Sewage Sludge, U.S. Environmental Protection, Office of Research and Development, National Risk Management Research Laboratory, Centre for Environmental Research Information. EPA/625/R-92-013, Cincinnati, OH, 1999.
[57] Liangliang Wei, Fengyi Zhu, Qiaoyang Li, et al., 2020, Development, Current State, and Future Trends in Sludge Management in China Based on Exploratory Data and CO2-equivalent Emission Analysis, vol. 144, Environmental International, Elsevier, 2020, 106093.
[58] Olawale T. Ogunwumi, Lukumon Salami, Perspective Chapter: Industrial Waste Landfill, in: Dr Suhaiza Zailan (Ed.), Solid Waste Management - Recent Advances, IntechOpen, Princes Gate Court, London, SW7 2QJ, UK London, UK, 2023, pp. 1–32, https://doi.org/10.5772/intechopen.108787.
[59] Anandkhumar Naorem, Somasundaram Jayaraman, P. Yash, Dang, et al., Soil Constraints in an Arid Environment- Challenges, Prospects, and Implications, Agronomy, MDPI 13 (1) (2023) 220, https://doi.org/10.3390/agronomy13010220.
[60] Sonia Boudjabi, Haroun Chenchouni, On the Sustainability of Land Applications of Sewage Sludge: How to Apply the Sewage Biosolid in Order to Improve Soil Fertility and Increase Crop Yield? Chemosphere 282 (2021), 131122.
[61] Bhavisha Sharma, Abhijit Sarkar, Pooja Singh, Rajeev Pratap Singh, Agricultural Utilization of Biosolids. A Review on the Potential Effects on Soil and Plants Growth, Journal of Waste Management, ScienceDirect 64 (2017) 117–132, https://doi.org/10.1016/j.wasman.2017.03.002.

[62] Sergio Siqueira Jr, Mariana Antonio Desouza Perreira, Marjuli Morishigue, et al., Circular Economy in the Biosolid Management by Nexus Approach: A View to Enhancing Safe Nutrient Recycling-Pathogens, Metals, and Emerging Organic Pollutants of Concern, Sustainability 14 (2022), 14693, https://doi.org/10.3390/sul142214693.

[63] S.O.M. Apec, Understanding the Bio-Circular Green (BCG) Economic Model, August 2022, APEC SOM Committee on Economic and Technical Co-operation, 2022.

[64] Evagelica Stavridou, Ioannis Giannakis, Ioannis Karamichiali, Natalie N. Kamou, Lagiolis George, et al., Biosolid-amended Soil Enhances Defense Responses in Tomato Based on Metagenomic Profile and Expression of Pathogenesis-related Genes, Plants 2789 (10) (2021), https://doi.org/10.3390/plants10122789.

[65] M.K. Banks, A.P. Schwab, N. Cofield, J.E. Alleman, M. Switzenbaum, J. Shalabi, P. Williams, Biosolid-amended Soils: Part I: Effects of Biosolids Application on Soil Quality and Ecotoxicity, Water Environ. Res. 78 (11) (2006) 2217.

[66] USEPA, Biosolid Technology Factsheet: Land Application of Biosolids, U.S. Environmental Protection Agency, Office of Water Management, Washington D.C, 2000. EPA 832-F-0064.

[67] Christos D. Tsadilas, I.K. Mitsios, Evangelia E. Golia, Influence of Biosolid Application on Some Soils' Physical Properties, Communication in Soil Science and Plant Analysis 36 (4–6) (2005) 709–716, https://doi.org/10.1081/CSS-200043350.

[68] Simin Li, Li Zhu, Li Jin, Ke Xin, et al., Influence of Long-term Biosolid Applications on Communities of Soil Fauna and their Metal Accumulation: A Field Study, Environ. Pollut. 260 (2020), 114017, https://doi.org/10.1016/j.envpol.2020.114017.

[69] Amira Oun, Arun Kumar, Timothy Harrington, Andreas Angelakis, Irene Xagoraraki, Effects of Biosolids and Manure Application on Microbial Water Quality in Rural Areas in the, U.S. Water 6 (12) (2014) 3701–3723, https://doi.org/10.3390/w6123701.

[70] Lawrence Wang, Nazih Shammar, Gregory Evanylo, Engineering and Management of Agricultural Land Application of Biosolid, in: Biosolid Engineering and Management, Handbook of Environmental Engineering and Management, Springer, 2009.

[71] David Brune, Ken Arnold, John Dunn, Jerry Carpenter, Best Management Practices for Biosolids Land Application: A Review, University of Missouri, USA, 2022.

[72] Rodolfo Gentili, Roberto Ambrosini, Chiara Montagnani, Sarah Carroni, Sandra Cittero, Effect of Soil pH on the Growth, Reproductive Investment, and Pollen Allergenicity of Ambrosia Artemisifolia L, Front. Plant Sci. 9 (2018), https://doi.org/10.3389/fpls.2018.01335.

[73] Silvana Torri, Studart Rodrigo, Giancarlo Renella, Alejandro Vadecantos, Leonid Perelomov, Biosolid Soil Application: Agronomic and Environmental Implication 2014, Applied and Environmental Soil Science, Hindawi, 2015, 6277819, https://doi.org/10.1155/2015/627819.

[74] Jim Newton, Land Application of Biosolid and Domestic Septage. Process Design Manual, United States Environmental Protection Agency, Office of Research and Development, Washington D.C., 2020, 20460.

[75] B.J. Chambers, Fiona Nicholson, M. Aitken, et al., Benefits of Biosolids to Soil Quality and Fertility, Water Environ. J. 17 (3) (2007) 162–167. https://10.1111/j.1747-6593.2003.tb004555.

[76] L. Virginia Jin, N. Kenneth Potter, V. Mari-Vaughn Johnson, R. Daren Harmel, G. Jeffrey Arnold, Surface-appllied Biosolids Enhance Soil Organic Carbon and Nitrogen Stocks but Have Contrasting Effect on Soil Physical Quality. Biosolid Soil Application: Agronomic and Environmental Implication 2014 Special Issue, Applied and Environmental Soil Science, 2015, 715916, https://doi.org/10.1155/2015/715916, 2015.

[77] Mpca, Animal Feedlots Minnesota Rules (Chapter 7020) (online), Minnesota Pollution Control Agency, 2019. Available at: https://www.pca.state.mn.us. (Accessed 22 February 2023).

[78] NRCS, Nutrient Management, Code 590 (online), Conservation Practice Standard. Naional Resources Conservation Service (2022). Available at: https://www.nrcs.usda.gov/sites/default/files/2022-09/NRCS_590_standard/. (Accessed 22 February 2023).

[79] N.P. Nikolaidis, L. Demetropoulou, M.L. Saru, Land Application of Biosolids: Benefits, Risks, and Costs. International Water Association, Water Purification and Reuse, WWPR12, ResearchGate, 2012.

[80] Anja Coors, Mark Edwards, Pascale Lorenz, et al., Biosolids Applied to Agricultural Land: Influence on Structural and Functional Endpoints of Soil Fauna on a Short-and Long-term Scale, J. Sci. Teach. Educ. 562 (2016) 312–326, https://doi.org/10.1016/j.scitotenv.2016.03.226.

[81] E. Margui, Monica Iglesias, Francesc Camps, et al., Long-term Use of Biosolids and Organic Fertilizers in Agricultural Soils: Potential Toxic Element Occurrence and Mobility, Environ. Sci. Pollut. Control Ser. 23 (5) (2016), https://doi.org/10.1007/s11356-015-5618-9.

[82] Nuno Nunes, Carla Ragonezi, Carla Gouveia, Miguel Pinheiro de Cavalho, Review of Sewage Sludge as a Soil Amendment in Relation to Current International Guidelines: A Heavy Metal Perspective, Sustainability 13 (4) (2021) 2317, https://doi.org/10.3390/su13042317.

[83] D.P. Mohapatra, M. Cledon, S.K. Brar, R.Y. Surampalli, Application of Wastewater and Biosolids in Soils: Occurrence and Fate of Emerging Contaminants, Water Air Soil Pollut. 227 (2016) 77, https://doi.org/10.1007/s11270-016-2768-4.

[84] Ravinder Kumar, Arun K. Vuppaladadiyam, Elsa Antunes, et al., Emerging Contaminants in Biosolids: Presence, Fate, and Analytical Techniques, 2022, Emerging Contam. 8 (2022) 162e194. keaipublishing.com/cn/journals/emerging-contaminants.

[85] Barbara Gworek, Marta Kijenska, Justyna Wrzosek, Magdalena Graniewska, Pharmaceuticals in the Soil and Plant Environment: A Review, Water Air Soil Pollut. 232 (145) (2021).

[86] Kuok H.D. Tang, Tony Hadibarata, Microplastics Removal through Water Treatment Plants: Its Feasibility, Efficiency, Future Prospects and Enhancements by Proper Waste Management – A Review, Environmental Challenges 5 (2021) (2021), 100264.

[87] Amir Hossein, Hamidianab Minyangabe, A Review on the Characteristics of Microplastics in Wastewater Treatment Plants: A Source for Toxic Chemicals, Journal of (Leaner Production 2021 (295) (2021), 126480.

[88] Zhiqiang, et al., Are Rural and Small Community Aerated Wastewater Stabilization Ponds a Neglected Source of Microplastic Pollution? Water 2021 13 (2021) 2833.

[89] F. Pomati, C. Orlandi, M. Clerici, F. Luciani, E. Zucialo, Effects and Interaction in an Environmentally Relevant Mixture of Pharmaceuticals, Toxicol. Sci. 102 (2008) 129–137.

[90] M. Farre, S. Perez, C. Gonclaves, M. Alpendurada, D. Barcelo, Green Analytical Chemistry in the Determination of Organic Pollutants in the Aquatic Environment, Trends Anal. Chem. 29 (2010) 1347–1362.

[91] J.L. Oaks, M. Gilbert, M.Z. Virani, R.T. Watson, C.U. Meteyer, B.A. Rideout, et al., Diclofenac Residues as the Cause of Vulture Population Decline in Pakistan, Nature 427 (2004) 630–633.

[92] J. Garri, B. Vollat, K. Dunis, A. Pery, T. Junker, M. Ramu, G. Fink, T.A. Ternes, Effects of the Parasiticide Ivermectin on Cladoceran Daphnia Magna and the Green Algea Pseudo-Kirchneriella Subcapitata, Chemisphere 69 (2007) 903–910.

[93] R. Lange, T.H. Hutchinson, C.P. Croudace, F. Siegmund, H. Schweinfurth, P. Hampe, et al., Effects of the Synthetic Estrogen 17-α-ethinylestradiol on the Life Cycle Fathead Minow (Pinephales promelas), Environ. Toxicol. Chem. 20 (2001) 1216–1227.

[94] Raymond A. Wuana, Felix E. Okieimen, Heavy Metals in Contaminated Soils: A Review of Sources, Chemistry, Risks, and Best Available Strategies for Remediation, vol. 402647, International Scholarly Research Network, 2011, pp. 1–20, https://doi.org/10.5402/2011/402647.

[95] Maria L. Azevedo, Luis R. Ferracciu, Luiz R. Guimaraes, Biosolids and Heavy Metals in Soils: A Review, Sci. Agric. 60 (4) (2003), https://doi.org/10.1590/s0103-90162003000400029.

[96] Mojeed Agoro, Abiodun Adeniji, Adefisoye Martins, Omobola Okoh, Heavy Metals in Wastewater and Sewage Sludge from Selected Municipal Treatment Plants in Eastern Cape Province, South Africa. Water, MDPI 12 (10) (2020) 2746, https://doi.org/10.3390/w12102746.

[97] S.M. Shasheen, M.S. Shams, F.A. Elbehiry, S.M. Ibrahim, Influence of Stabilized Biosolid Application on Availability of Phosphorus, Copper, and Zinc, 2012, Applied and Environmental Soil Science, Hindawi, 2012, 817158, https://doi.org/10.1155/2012/817158.

[98] Emna Ammar, Loic Morin, Abdelghani Sghir, Environmental, Economic, and Ethical Assessment of the Treated Wastewater and Sewage Sludge Valorization in Agriculture, Handb. Environ. Chem. (2020), https://doi.org/10.1007/698_2020_606.

[99] Decong Xu, Zhangjun Shen, Chaugming Dou, Zhiyong Dou, Li Yang, Yi Gao, Qingye Sun, Effects of Soil Properties on Heavy Metals Bioavailability and Accumulation in Crops Grains under Different Farmland Use Patterns, Sci. Rep. 12 (2022) 9271, https://doi.org/10.1038/s41598-022-13140-1.

[100] USEPA, Our Current Understanding of the Human Health and Environmental Risks of PFAs, The U.S. Environmental Protection Agency, Office of Research and Development, National Risk Management Research Laboratory, Centre for Environmental Research Information. EPA/625/R-92-013, Cincinnati, OH, 2022.
[101] Raveena Dhore, Ganti Murthy, Per-and Poly-fluoroalkyl Substances Production, Applications, and Environmental Impacts, Bio-resources Technology, ScienceDirect 341 (2021), 125808, https://doi.org/10.1016/j.biortech.2021.125808.
[102] Juliane Gluge, Martin Scheringer, Ian Cousins, et al., An Overview of the Uses of Per-and Poly-fluoroalkyl Substances, Environmental Science Processes Impact 22 (2020) 2345–2373, https://doi.org/10.1039/DOEMOO29IG.
[103] Fda, Per- and Poly-fluoroalkyl Substances (PFAs) in Cosmetics, United States Food and Drug Administration. 10903, New Hampshire Avenue, Silver Spring, MD (2022), 20993-0002.
[104] Lazcano Kim, T.Y. Choi, M.L. Mashtare, L.S. Lee, Characterizing and Comparing Per-and Poly-fluoroalkyl Substances in Commercially Availability Biosolids and Organic Non-biosolid-based Products, Environ. Sci. Technol. 39 (11) (2020) 3946–3956.
[105] A.C. Blaine, C.D. Rich, L.S. Hundal, C. Lau, M.A. Mills, K.M. Harris, et al., Uptake of Perfluoroalkyl Acids into Edible Crops via Land Applied Biosolids: Field and Greenhouse Studies, Environ. Sci. Technol. 47 (24) (2013) 14062–14069.
[106] Damien Terrence Moodie, Fate, Behaviour, and Ecological Impacts of Biosolid-Derived Per-And Poly-Fluoroalkyl Substances [thesis], Environmental Science Department, RMIT University, 2021.
[107] Blake Langebach, Mark Wilson, Per-and Poly-fluoroalkyl Substances (PFAs): Significance and Considerations within Regulatory Framework of the United States, Int. J. Environ. Res. Publ. Health 18 (21) (2021), 11142, https://doi.org/10.3390/ijerph182111142.
[108] Sibel Barisci, Rominder Suri, Electro-oxidation of Short- and Long-chain perfluoroalkyl Substances (PFASs) Under Different Process Conditions, Journal of Environmental Chemical Engineering, ScienceDirect 9 (4) (2021), 105323, https://doi.org/10.1016/j.jece.2021.105323.
[109] Hamidreza Sharifan, Majid Bagheri, Dan Wang, et al., Fate and Transport of Per-and Poly-fluoroalkyl Substances (PFAs) in the Vadoze Zone, Sci. Total Environ. 771 (10) (2021), 145427, https://doi.org/10.1016/j.scitotenv.2021.145427.
[110] Hong Chen, Chan Zhang, Jianbo Han, Yixuan Yu, Peng Zhang, PFOS and PFOA in Influents, Effluents, and Biosolids of Chinese Wastewater Treatment Plants and Effluent-receiving Marine Environments, Environmental Pollution, ScienceDirect 170 (2012) 26–31, https://doi.org/10.1016/j.envpol.2012.06.016.
[111] P. Li, X. Oyang, X. Xie, Z. Li, H. Yang, J. Xi, Y. Guo, X. Tian, B. Liu, J. Li, Z. Xiao, Phototoxicity Induced by Perfluorooctanoic Acid and Perfluorooctane Sulfonate via Metabolics, J. Hazard Mater. 389 (2020), 121852.
[112] CDC, Perfluorooactanoic Acid. National Biomonitoring Program, Centre for Disease Control and Prevention. The U.S. Department of Health and Human Services, 2017.
[113] F.C. Oliveira, M.E. Mattiazzo, Heavy Metals in an Oxisol Treated with Sewage Sludge and in Sugarcane Plants, Scientia Agricola, ResearchGate 58 (3) (2001) 581–593.
[114] AnYan, Yamn Wang, Swee Ngin Tan, et al., Phytoremediation: A Promising Approach for Revegetation of Heavy Metal Polluted Land, Front. Plant Sci. 11 (2020) 369, https://doi.org/10.3399/fpls.2020.00359.
[115] Bieby V. Tanyahu, Siti R.S. Abdullah, Hassan Basri, Mushrifah Idris, Nurina Anuar, and Muhammad Mukhlisin, A Review on Heavy Metals (As, Pb, and Hg) Uptake by Plants through Phytoremediation. International Journal of Chemical Engineering (2011), 939161.
[116] Minori Uchimiya, Desmond Bannon, Hiromi Nakanishi, McBride Murray, Marc Williams, Toshihiro Yoshihana, Chemical Speciation, Plant Uptake, and Toxicity of Heavy Metals in Agricultural Soils, J. Agric. Food Chem. 68 (46) (2020) 12856–12869.
[117] Saikat Mitra, Arka J. Chakraborty, Abu M. Tareq, et al., Impact of Heavy Metals on Environmental and Human Health: Novel Therapeutic Insights to Counter the Toxicity, Journal of King Saud University-Science, ScienceDirect 34 (3) (2022), 101865.
[118] Naseri Balali-mood, Tahergorabi, et al., Toxic Mechanisms of Five Heavy Metals: Mercury, Lead, Chromium, Cadmium, and Arsenic, Frontier Pharmacology 12 (2021), https://doi.org/10.3389/fphar.2021.643972.
[119] Monisha Jaishanker, Tenzin Tseten, Naresh Anbalagan, et al., Toxicity Mechanism and Health Effects of Some Heavy Metals, Interdiscipl. Toxicol. 7 (2) (2015) 60–72, https://doi.org/10.2478/intox-2014-0009.
[120] Hailin Zhang, Using Biosolids as a Plant Nutrient Source, Division of Agricultural Sciences and Natural Resources, Oklahoma State University Extension, Agricultural Hall, Oklahoma State University, Stillwater, OK 74078, 2017.
[121] Jorge Antonio Silva-Leal, Andrea Perez-Vidal, Patricia Torres-Lozada, Effects of Biosolids on the Nitrogen and Phosphorus Content of the Soil Used for Sugar Plantation, National Library of Medicine, Heliyon 7 (3) (2021), e06360, https://doi.org/10.1016/j.heliyon.2021.e06360.
[122] Yulia Markunas, Vadim Bostan, Andrew Larsen, Michael Payne, Lynda McCarthy, Vertical Phosphorus Migration in a Biosolid-Amended Sandy-loam Soil in a Laboratory Setting: Concentration in Soils and Leachates, Applied and Environmental Soil Sciences, Hindawi, 2016, 3460939, https://doi.org/10.1155/2016/3460939.
[123] McBride Murray, Long-term Biosolid Application on Land: Beneficial Recycling of Nutrients or Eutrophication of Agroecosystem? Soil System 2022 (6) (2022) 9, https://doi.org/10.3390/soilsystems6010009.
[124] Adul-Wahab Mossa, H. Elizabeth, Bailey, Abida Usman, Scott D. Young, Neil M.J. Crout, The Impact of Long-Term Biosolid Application (>100 Years) on Soil Metal Dynamics, School of Biosciences, University of Nottingham, Sutton, Bonington Campus, Leicester, LE12 SRD, UK, 2020.
[125] Daniel Venghaus, Mathias Berjenbruch, Microplastics in Urban Water Management, 2017, Technical Transaction I, Environmental Engineering, 2017, https://doi.org/10.4467/235373XCT.17.011.6108.
[126] Rolf U. Halden, Avery E. Lindeman, Allison E. Aiello, et al., The Florence Statement on Triclosan and Triclocarban. Environmental Health Perspectives, National Library of Medicine 125 (6) (2017), 064501.
[127] Saidu M. Bashir, Sam Kimiko, Chu-Wa Mak, James Kar-Hei Fang, David Goncalves, Personal Care and Cosmetic Products as a Potential Source of Environmental Contamination by Microplastics in a Densely Populated Asian City, Front. Mar. Sci. 8 (2021), https://doi.org/10.3389/fmars.2021.683482.
[128] Sarva M. Praveena, Siti N.M. Shaiffudin, Shinichi Akizuki, Exploration of Microplastics from Personal Care and Cosmetic Products and its Estimated Emissions to Marine Environment: An Evidence from Malaysia, Mar. Pollut. Bull. 136 (2018) 135–140, https://doi.org/10.1016/j.marpolbul.2018.09.012.
[129] Lara D. Jak, Nora Exposito, Joaquin Rovira, Karim Macher, Fernando Valero, Jordi Sierra, Screening of MIS in Water and Sludge Lines of a Drinking Water Treatment Plant in Catalonia, Spain, Water Res. 2022 (225) (2022), 119185.
[130] Cleanpng, Microbeads Denitifricio Microplastic Esfoliazione per la Cura Personale, Available at: https://www.it.cleanpng.com/png.p810bz, 2018. (Accessed 28 February 2023).
[131] Claudia Campanale, Carmine Marssarelli, Illaria Savion, et al., A Detailed Review Study on Potential Effects of Microplastics and Additives of Concern on Human Health, International Journal of Environmental Research and Public Health, National Library of Medicine 17 (4) (2020) 1212, https://doi.org/10.3390/ijerph17041212.
[132] Concentta Pironti, Maria Ricciadi, Oriana Motta, et al., Microplastic in the Environment: Intake through Food Web, Human Exposure and Toxicological Effects, Toxics, MDPI 9 (9) (2021) 224, https://doi.org/10.3390/toxics9090.224.
[133] S.M. Mintenig, M.G.J. Loder, S. Pimpke, G. Gerdts, Low Members of the Microplastics Detected in Drinking Water from Groundwater Sources, J. Sci. Teach. Educ. 648 (2019) 631–635.
[134] Aaron Z. Garza, Sharon B. Park, Remek Kocz, Drug Elimination: StatPearls (online). National Library of Medicine (2022). Available at: https://www.ncbi.nlm.nih.gov/books/NBK547662. (Accessed 7 March 2023).
[135] Victoria Blanco, Carolina Hernandora, Paula Scibona, Waldo Belloso, et al., Acute Kidney Injury and Pharmacokinetic Changes and, its Impact on Drug Prescription, Healthcare, MDPI 7 (1) (2019) 10, https://doi.org/10.3390/healthcare7010010.
[136] Laura J. Carter, Eleanor Harris, Mike Williams, Jim J. Ryan, Rai S. Kookana, B.A. Alistair, Boxall, Fate and Uptake of Pharmaceuticals in Soil-Plant Systems, J. Agric. Food Chem. 64 (2014) 816–825.
[137] Sara C. Monteiro, Alistair B.A. Boxall, Factors Affecting the Degradation of Pharmaceuticals in Agricultural Soils, Environ. Toxicol. Chem. 28 (2) (2009) 2546–2554, https://doi.org/10.1897/08-657.1.

[138] Emma R. Knight, L.J. Carter, M.J. McLaughlin, Bioaccumulation, Uptake, and Toxicity of Carbamazepine in Soil-Plant Systems, Environ. Toxicol. Chem. 37 (4) (2018) 1122–1130, https://doi.org/10.1002/etc.4053.
[139] A. Piotrowicz-Cieslak, B. Adomas, Nalecz-Jawecki Michalczyk, Phototoxicity of Sulfamethazine Soil Pollution to Six Legume Plant Species, J. Toxicol. Environ. Health, Part A 73 (18) (2010), https://doi.org/10.1080/152873794.2010.492006.
[140] MedlinePlus, Carbamazepine (online). National Library of Medicine (2022). Available at: https://www.medlineplus.gov. (Accessed 13 February 2023).
[141] Mellisa A. Mercer, Sulfonamides and Sulfonamide Combination Use in Animals, Merck Manual, Maryland College of Veterinary Medicine, Virginia, 2022.
[142] Nuria Lozano, Clifford P. Rice, Mark Ramirez, Alba Torrents, The Fate of Triclocarban, Triclosan, and Methyltriclosan during Wastewater and Biosolid Treatment Processes, Water Resources, National Library of Medicine 47 (13) (2013) 4519–4527, https://doi.org/10.1016/j.waters.2013.05.015.
[143] Jongmun Cha, Alison M. Cupples, Detection of the Antimicrobials Triclocarban and Triclosan in Agricultural Soils Following Land Application of Municipal Biosolid, Water Res. 43 (2009) 2522–2530.
[144] Ban Yu, Guodi Zheng, Xuedong Wang, Min Wang, Tongbin Chen, Biodegradation of Triclosan and Triclocarban in Sewage Sludge during Composting Under Three Ventilation Strategies, Front. Environ. Sci. Eng. 13 (41) (2019), https://doi.org/10.1007/s11783-019-1125-4.
[145] Guang-Guo Ying, Yu Xiang-Yang, Kookana Rai, Biological Degradation of Triclocarban and Triclosan in a Soil under Aerobic and Anaerobic Conditions and Comparison with Environmental Fate Modeling, Environ. Pollut. 150 (3) (2008) 300–305, https://doi.org/10.1016/j.envpol.2007.02.013.
[146] Gurpreet Singh Dhillon, Surinder Kaur, Rama Pulicharia, et al., Triclosan: Current Status, Occurrence, Environmental Risks, and Bioaccumulation Potential, International Journal of Environmental Resources and Public Health, National Library of Medicine 12 (5) (2015) 5657–5684.
[147] Matthew J. Scott, Malcolm N. Jones, The Biodegradation of Surfactants in the Environment, Biochimica et Biophysica Acta (BBA)-Biomembranes 1508 (1–2) (2000) 235–251, https://doi.org/10.1016/50304-4157(00)00013-7.
[148] Guang-Guo Ying, Fate, Behaviour, and Effects of Surfactants and their Degradation Products in the Environment, Environ. Int. 32 (2006) (2006) 417–431.
[149] Qing Zhang, Weiping Mei, Longfei Jiang, Qian Zheng, Chunling Luo, Gan Zhang, Environmental Fate and Effects of Organo-phosphate Flame Retardants in the Soil-Plant System, 2021, Soil Ecology Letters 3 (3) (2021) 178–188, https://doi.org/10.1007/s42832-021-0084-4.
[150] NIH, Endocrine Disruptors (Online), National Institute of Environmental Health Sciences, 2023. Available at: https://www.niehs.nih.gov/health/topics/agents/endocrine/index.cfm. (Accessed 26 March 2023).
[151] Manoj Kumar, Devojit Kumar Sarma, Swasti Shubham, Manoj Kumawat, Vinod Verma, Anil Prakash, Rajnarayan Tiwari, Environmental Endocrine-Disrupting Chemical Exposure: Role in Non-Communicable Diseases, Frontier Public Health, Sectional Environmental Health, and Exposome 8 (2020), https://doi.org/10.3389/fpubh.2020.553850.
[152] FAO, Status of the World's Soil Resources. Food and Agricultural Organization of the United Nations Main Report (2015). Available at: https://www.fao.org. (Accessed 7 March 2023).
[153] Hana Hudcova, Vymazal Jan, Milos Rozkosny, Present Restrictions of Sewage Sludge Application in Agriculture within the European Union, Soil Water Res. 14 (2) (2019) 104–120.
[154] Christina Tsiarta, Steven Watson, Joe Hudson, Final Implementation Report for Directive 86/278/EEC on Sewage, Report for Director General on the Environment, European Commission. The European Commission (2018). ENV.C.2/FRA/2013/0023.
[155] EUREAU, Sewage Sludge Directive. 86/278/EEC- Stakeholder Questionnaire: EUREAU Responses, European Federation of National Associations of Water Resources, 2013, 2013.
[156] The European Commission, Legislation of Sewage Sludge: Directive 86/278/EEC on Sewage Sludge. European Commission Director General on the Environment (2006). Available at: https://www.europe.eu.int/comm/environment/waste/index.htm. (Accessed 13 March 2023).
[157] MACROTRENDS, World Population between 1950 and 2100. Macro-Trends LLC, 2023. Available at: https://www.macrotrends.net/countries/WLD/world/population. (Accessed 9 March 2023).
[158] USEPA, Biosolid Laws and Regulations. United States Environmental Protection Agency (2002). Available at: https://www.epa.gov/biosolids/biosolids-laws-and-regulations. (Accessed 13 March 2023).
[159] USEPA, A Plain English Guide to the EPA Part 503 Biosolid Rule, Office of Water Management, United States Environmental Protection Agency, 1994. EPA/832/R-93/003.
[160] G.K. Evanylo, Agricultural Land Application of Biosolids in Virginia: the Regulations, Virginia Co-operation Extension Publication. Department of Crop and Soil Environmental Sciences, Virginia Tech, 2009, pp. 302–452.
[161] C.P. Gerba, I.L. Pepper, L.F. Whitehead, A Risk Assessment of Emerging Pathogens of Concern in the Land Application of Biosolids, Water Sci. Technol. 46 (1) (2002) 225–230.
[162] Robert Brobst, Biosolid Regulations. Biosolid Management Handbook, EPA Region VIII, 18 Str., Denver Colorado 80202, 2001.
[163] R. Iranpour, H.H.J. Cox, R.J. Kearney, J.H. Clerk, A.B. Prince, G.T. Daigger, Regulations for Biosolid Land Application in U.S. and European Union, Journal of Residual Science and Technology 1 (4) (2014).