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Review

Biosolids: The Trojan horse or the beautiful Helen for soil fertilization?

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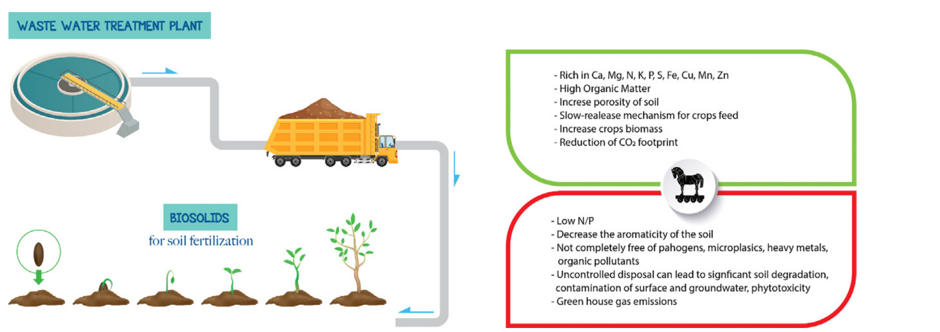
HIGHLIGHTS

- Emerging pollutants and microorganisms are the hidden enemies of Troy.
- Various treatment technologies can be the Achilles heel of biosolids' quality.
- Biosolids management policies and legislation need to be revised.
- Dielectric Barrier Discharge plasmas is a promising sanitary process for biosolids.
- Biosolids can be a valuable part of circular economy instead of unexploited waste.

GRAPHICAL ABSTRACT



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ABSTRACT

The simultaneous requirement to manage resources and wastes in more rational way has meant that many communities worldwide have begun to search for long-term alternative solutions. Reuse and recovery of biosolids is considered to be a constant solution of circular sustainability, as waste disposal without further reuse background like fertilizer is no longer an alternative to be promoted. There have been developed many treatment methods over the years for the stabilization and sanitization of biosolids. However, the literature concludes that none of them is fully integrated by meeting all the basic criteria. Each method has its Achilles heel, and the appropriateness of the method lies in what is the goal each time. There are conventional methods with positive reciprocity in terms of sustainability, reuse indicators and technological maturity, but have high risk of microorganisms' reappearance. New advanced sustainable technologies, such as cold plasma, need to be further studied to apply on a large scale. The reuse of biosolids as construction materials is also discussed in the context of circular economy. Biosolids reuse and management legislation frame need to be revised, as a directive adopted 30 years ago does not fully meet communities' current needs.

Contents

Abbreviations: N, nitrogen; P, phosphorus; K, potassium; CMCs, Component Material Categories; TN, Total Nitrogen; RP, Rock Phosphate; VS, Volatile Solids; EPS, Extracellular Polymeric Substances; TSS, Total Suspended Solids; TS, Total Solids; FE-DBD, Floating Electrode Dielectric Barrier Discharge; PFRP, Process to Further Reduce Pathogens; PSRP, Process to Significantly Reduce Pathogens; EPA, Environmental Protection Agency.

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1. Introduction

The growing global population, which is expected to reach 8.5 billion people by 2030, has straight-caused the generation of excessive amounts of wastewaters worldwide which makes apparent the need for sustainable wastewater management and treatment (Lahlou et al., 2021; Ungureanu et al., 2020). In a recent study, Jones et al. (2021) estimate that 359.4 billion m^3 of wastewater is generated annually, with a global average of 49.0 $m^3\ yr^{-1}$ per capita. Global wastewater collection and treatment is estimated to be 225.6 and 188.1 billion $m^3\ yr^{-1}$, respectively. These values show that about 63% and 52% of the wastewater produced worldwide is collected and treated, respectively, with about 84% of the collected wastewater undergoing a treatment process (Jones et al., 2021). Wastewater reuse is estimated at 40.7 billion $m^3\ yr^{-1}$, which is about 11% of the total wastewater generated. Specifically, only 22% of the treated wastewater is reused, the remaining 78% (a total of 147.4 billion $m^3\ yr^{-1}$) is discharged to the environment, and about 171.3 billion $m^3\ yr^{-1}$ of wastewater is discharged directly into the environment without being treated in any way (Jones et al., 2021).

Wastewater treatment processes lead to two main products: treated water and sludge. The treated water is finally disposed of to the natural water recipients (surface water, sea, deep wells, groundwater), while the produced sludge must be treated properly, before disposing of it mainly in landfills or using it as a soil conditioner (Al-Musharafi et al., 2012; Bisognin et al., 2020; Jasim et al., 2016). The sewage wastewater comes from domestic activities, containing wastewaters from households, public buildings, restaurants, and hospitals. Their composition is a complex mixture containing water together with organic, inorganic components and a large number of pathogenic bacteria as well as viruses and parasites (Y. Wang et al., 2018; Xavier and Varghese, 2020). It should be noted that the hospital wastewater is a unique category of sewage wastewater because of its major constituents (Xiong et al., 2020; Zamparas et al., 2019b). The hospital wastewater contains daily loads of high concentration in antibiotics, disinfectants, and antibiotic-resistant bacteria from the hospital and medical centers' activities (Kümmerer, 2001; Shokoohi et al., 2020; Voigt et al., 2020), a fact that makes it necessary to be pretreated before entering the Wastewater Treatment Plant. Industrial wastewater is non-sewage wastewater that comes from industrial operations such as chemical, electrochemical, electronic, petrochemical, and food-processing industries (Abdelbasir and Shalan, 2019; Rajesh Banu et al., 2020). These wastewaters are associated with high concentrations of dissolved metal salts (heavy metals) and may include some domestic sewage (Azimi et al., 2017; Yachigo and Sato, 2013).

Biosolids are the solid residues generated during the sewage treatment processes (Cieślik et al., 2015). This term has recently replaced the term "sewage sludge" and should not be used interchangeably (Sharma et al., 2017). However, the term "sludge" refers to a liquid or semi-liquid produced during the wastewater treatment, that is not submitted to further treatments (Ukwatta et al., 2015). Biosolids represent sewage sludge that has been treated by processes such as digestion, dewatering, disinfection, thermal and chemical conditioning (Chaudhary and Gough, 2021; Sharrer et al., 2009; X. Wang et al., 2018), advanced oxidation treatment processes, and has met standards required for beneficial use. The particular characteristics of the biosolids vary depending on their origin and the treatment process they have gone through (Collivignarelli et al., 2020).

The amounts of biosolids produced in the European Union testify to the size of municipal wastewater production in each country but also to the degree of treatment it undergoes. It is a fact that many wastewater treatment facilities throughout the world are unable to produce biosolids appropriate for land disposal, due to the lack of sanitation and stabilization treatment stages in their facilities. Therefore, the presence of several potentially toxic chemical elements (TCE) such as heavy metals, persistent organic pollutants and pathogenic agents (i.e. bacteria, protozoa, viruses) make them unsuitable for direct disposal. The use of biosolids as soil conditioner/fertilizer in agriculture is the most common practice in most Member States. Some countries reuse less than 20% of the biosolids produced in agriculture while in others the reuse in agriculture reaches 100% (Hudcová et al., 2019). It is estimated that 4×10^6 t of sludge has been used in agriculture, while 0.5×10^6 t is buried. Furthermore, relatively similar is the situation in the USA. More than 50% of the biosolids are used as organic fertilizer in crops, while 22% are intended for burial. Undoubtedly, the agricultural use of good quality sludge signifies a value-added route to ensure growth sustainability worldwide, where raw material availability, for example, phosphorus (phosphate rock is the major raw material for P-fertilizers), is insufficient to meet demand (Nedelciu et al., 2020; Zamparas, 2021; Zamparas et al., 2019a).

The world's current fertilizer production capacity is insufficient to satisfy rising demand, which has grown six-fold in the previous 50 years and continues to grow in synch with farmland (Usama and Khalid, 2018). As a result, the use of chemical fertilizer has reached its limit and a more sustainable approach is required. According to a recent publication by Margenot et al. (2019), using recovered P instead of chemical fertilizers will reduce the need for mineral-based phosphorus fertilizers and minimize the amount of P released into water bodies in the Midwest of the United States (Margenot et al., 2019). In Germany, Sewage Sludge Ordinance requires P recovery from sewage sludge, forcing Wastewater Treatment Plants to find a suitable technology for P recovery or to ensure P recovery after sludge incineration in order the environmental policy of the country to align with the sustainable development goals laid out in the 2030 Agenda, adopted by the United Nations in September 2015 (Günther et al., 2018).

Another alternative method to traditional fertilization is biosolids utilization in agriculture, for sustainable crop production, relieving pressure on nonrenewable resources. For this reason, use of biosolids for agricultural activities is one way to reduce the burden on conventional chemical fertilization. Based on science latest implementations and social, economic, and environmental needs, governments must employ alternative resources in the context of sustainable development and Circular Economy (Halden and Venkatesan, 2020).

The noticeable difference of the present review is the holistic approach of biosolids utilization in agriculture, focusing on multiple aspects of this always up-to-date topic. Thus, the article is believed to enhance the understanding around biosolids main physicochemical characteristics, emphasizing their pros, and cons as fertilizers. The legislation framing their use, disposal and reuse is revised and also conventional and emerging methods for treatment, focusing on sanitation and stabilization of biosolids have been extensively presented.

1.1. Bibliometric data

Literature overviews indicate that the use of biosolids that have been examined broadly can be categorized into the study of biosolids concerning (a) pathogens (Epstein, 2019; Oun et al., 2014; Sidhu and Toze, 2009; Viau and Peccia, 2009), (b) heavy metals (Bai et al., 2012; Hosseini Koupaie and Eskicioglu, 2015; Nikolaidis and Chheda, 2001), (c) pharmaceuticals (Bair et al., 2016; García-Santiago et al., 2016; K. Wang et al., 2018; Yadav et al., 2019), (d) the reuse in agriculture as a soil conditioner and (e) regulations to minimize sludge-related health issues and environmental risks (Eisenberg et al., 2008). The large number of published articles addressing biosolids (from 1987 – one year after the Sewage sludge directive 86/278/ECC – to 2021) represent the current growing attention being paid to resource recovery, circular economy, and environmental protection (Brown et al., 2020) (Fig. 1).

Moreover, the linear regression line represents a rising trend throughout the study period of the last 35 years, showing the interest of many scientists and researchers in the field of wastewater treatment (Fig. 1).

Further to that, it was also counted that all the papers were from 104 countries, among which the USA published the highest number of 5471

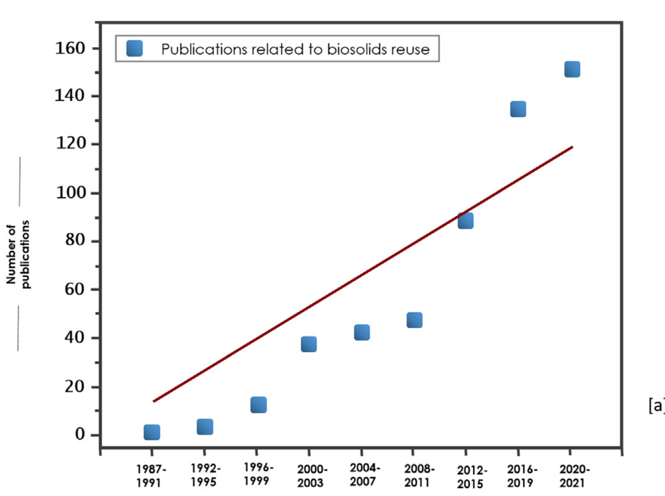

Fig. 1. Publications related to biosolids reuse from 1987 to 2020. The linear regression line shows a rising trend of publications related to the reuse of biosolids during the study period of the last 35 years (authors' own study, data from Scopus).

papers, accounting for 45.1% (2467 papers), followed by Canada (622 papers, 11.36%), Australia (488 papers, 8.91%), China (273 papers, 4.98%) and the United Kingdom (263 papers, 4.8%).

To estimate the most common keywords used, all papers were entered into VOS Viewer, which is a software tool for constructing and visualizing bibliometric networks. So, the top keywords from the 5471 biosolids studies are presented in Fig. 2. Colors refer to different clusters and related topics. The size of the circle demonstrates how frequently the term was used as keyword in the different publications. The distance between terms indicates the frequency of two terms occurring together in either the abstract title or keyword listing of publications (Fig. 2).

Around 240 papers, reviews, books, and conference proceedings were examined extensively and from which the most substantial conclusions were drawn for the creation of this review.

2. Main characteristics of biosolids

2.1. Biosolids composition

Biosolids contain high concentration in water at about 70%–80%, and also high amounts of organic matter (humic and fulvic substances) ranging from 50% to 70% (Fischer et al., 2020) and C/N ratio of 9:1 (Sakellariou-Makrantonaki and Dimakas, 2013). So, they can be considered as carbon-based materials, with biopolymeric compounds, while their composition varies depending on their origin, their production process, the storage, and environmental conditions. It has been reported that fatty acids constitute the predominant polar fraction, thereby representing 51% of the organic compounds in biosolids, whereas steroids and aliphatic compounds contribute to 13% and 14%, respectively (Torri and Alberti, 2012). The aromaticity of the biosolids is probably due to the alkyl carbon from the fatty acids, and paraffinic structures (Chiu and Tian, 2011). Total solids (TS) and volatile solids (VS) are crucial parameters describing the organic load and solid contents of biosolids. Specifically, liquid, dewatered, dried, or compost biosolids contain 2–12%, 12–30%, and 50% TS, respectively. VS indicates the availability of readily decomposable organic matter in biosolids (Wijesekara et al., 2016). The VS fraction has a strong correlation with odor emissions in biosolids, and hence VS is a critical determinant when applying biosolids to land (Wijesekara et al., 2016).

About 80% of the biosolids mass can be the extracellular polymeric substances (EPS), which are a complex mixture of high molecular weight biopolymers, consisting mainly of proteins and carbohydrates and to a lesser extent of humic substances, DNA and RNA, creating a protective layer for bacterial cells with hydrophilic and hydrophobic structures, on which xenobiotic molecules and water are bounded (Rajesh Banu et al., 2021). This complex net structure full of water reduces dewatering capacity of biosolids, which directly affects the anaerobic biodegradability. Therefore, removing EPS prior to this process will be more effective in enhancing the anaerobic biodegradability and biogas yield (Sowmya Packyam et al., 2015). The extraction of EPS is cost and energy efficient, as it significantly reduces pretreatment input energy and accelerates the biosolids liquefaction, which indirectly supports the profitable output yield.

Biosolids are rich in macronutrients such as Ca, Mg, N, K, P and S (Fischer et al., 2020) that are embedded in their organic matrix. They are also known as a good source of a variety of micronutrients, such as Fe, Cu, Mn, and Zn, which are not supplied by conventional fertilizers (Sun et al., 2016). As shown in Table 1, biosolids are composed of micro and macro nutrients, as well as include a number of other heavy metals, which, even in small amounts of ppb can be considered toxic in the context of landfilling. In addition, there is a rate gap between selected countries, which may be due to various factors such as seasonality, population or cultural criteria and dissimilar wastewater treatment technologies or variation of type of wastes (Sharma et al., 2017). For this reason, depending on the chosen reuse method biosolids require to be chemically analyzed further.

In Greece, the percentage of the organic matter in biosolids were found to be above 50% and their nutrient content were found to be in the range of 3.26–5.61% for N, 0.15–0.18% for K and 1.68–1.89% for P (Sakellariou-

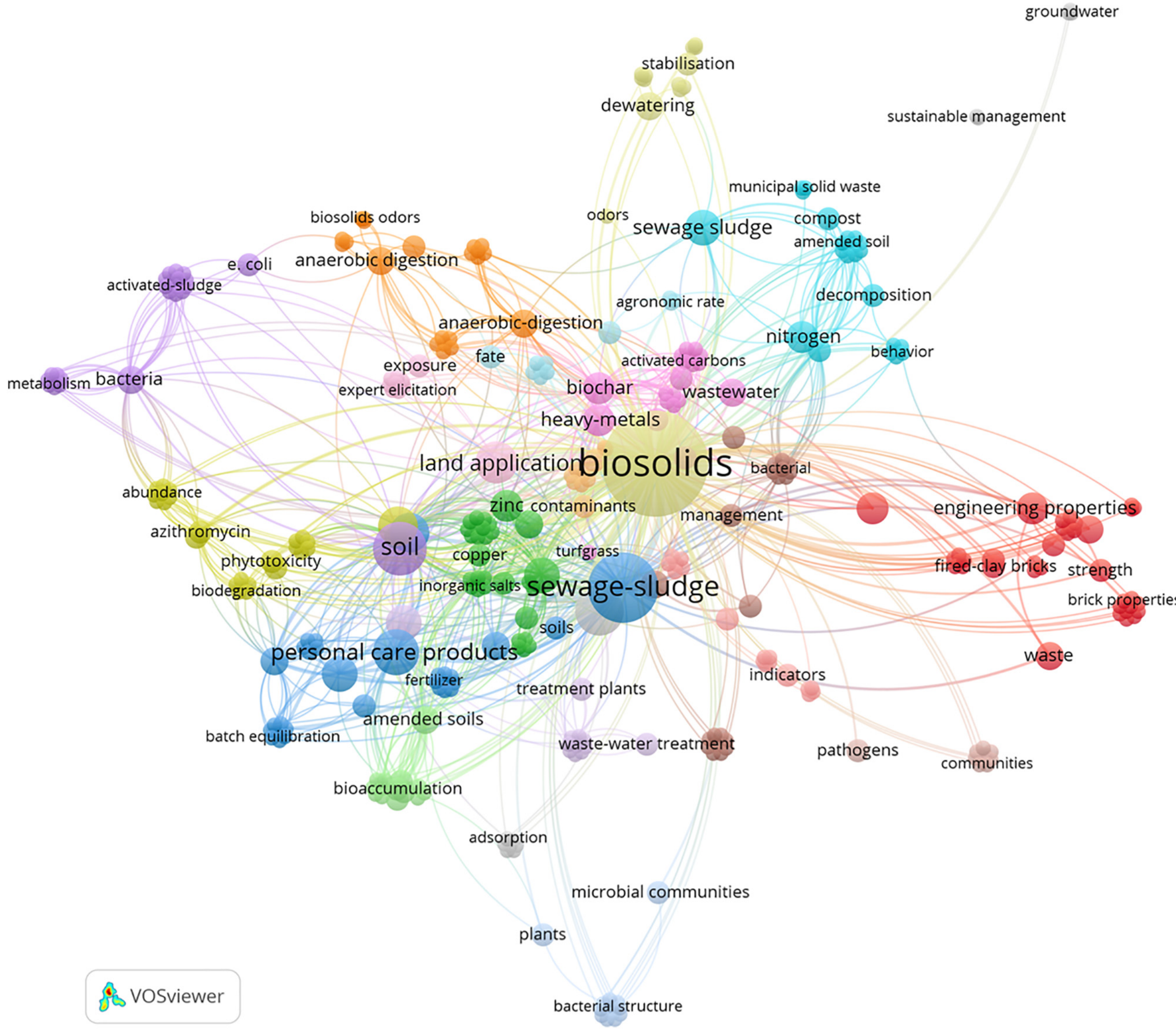

Fig. 2. Network visualization map of co-occurrence keywords for biosolids research. The colors represent different clusters.

Table 1
Physicochemical properties and composition of biosolids in different countries (adapted from (Antolín et al., 2005; Collivignarelli et al., 2019; Saha et al., 2017)).

Properties	India	China	Australia	Spain
pH	6.16–7.5	6.86–8.73	4.4–8.3	7.1–8.1
EC (ms cm^{-1})	2.28–2.7	0.667–5.01	1.6–7.9	1.2–3.9
Org.C (%)	5.52–12.6	–	–	–
Total N (%)	1.6–1.73	2.23–6.50	0.60–2.5	3–4.1
Total P (%)	0.49–1.3	1.06–2.18	0.28–0.83	2–3.6
Av. P (mg kg^{-1})	132–716.7	–	–	13,900
Total K (%)	0.8–1.26	0.46–0.62	0.18–0.45	0.24–0.47
Ex. K (mg kg^{-1})	208.9	593	–	–
Ex. Na (mg kg^{-1})	483	–	–	–
Ex. Ca (mg kg^{-1})	154.1	–	–	–
Total metals				
Fe (mg kg^{-1})	6059–14,390	0.46–2.40	13,824–18,026	31,200
Ni (mg kg^{-1})	47.17–60	52.5–202	166	<25–71
Mn (mg kg^{-1})	186.2–260	0.35–537	173	165–233
Zn (mg kg^{-1})	161–2050	0.21–1350	210–3060	560–1100
Pb (mg kg^{-1})	28.5–240	49.1–186	323	43–219
Cr (mg kg^{-1})	35.5–60	52.8–288	308	1–210
Cd (mg kg^{-1})	32.3–154.5	2.23–7.61	0.70–13.6	<0.2–3
Cu (mg kg^{-1})	186–330	0.27–975	92–1996	149–230

Makrantonaki and Dimakas, 2013). Phosphorus in biosolids is found in both soluble and insoluble organic or inorganic form (mainly in inorganic form), as iron phosphate and calcium phosphate (Torri et al., 2017). The inorganic P form makes up 70–90% of the total phosphorus.

The global supply shortage is insufficient to meet fertilizer demand. This fact indicates the need to find P alternative resources. In this frame, there have been efforts to extract P from biosolids, which are rich in organic phosphorus in the form of orthophosphate monoesters, diesters, phosphonates, and phospholipids (Torri et al., 2017). A fundamental study of P, N, and K levels in biosolids carried out by Stehouwer et al. (Stehouwer et al., 2000) using 240 samples analyzed from 12 Publicly Owned Treatment Works in Pennsylvania, exhibited average P levels of 4.74%. The studied biosolids were aerobically digested, anaerobically digested, or alkali-treated (Stehouwer et al., 2000). While Ajiboye states that the most preferred method for P extraction occurs mainly in the form of bioavailable inorganic P. For instance, 72% of the total P in the biosolids was HCl extractable, 33% $NaHCO_3$ extractable, 23% NaOH extractable, and 18% was water-soluble. The P extracted with hydrochloric acid was mainly associated with Ca, while the P extracted with $NaHCO_3$ and NaOH was mainly associated with Al and Fe (Ajiboye et al., 2004). The comparatively low amount of H_2O-extractable P could be due to the Fe, Al, and Ca in the biosolids, which are introduced as metal salts and lime during the treatment process (Ajiboye et al., 2004). It turns out that, stabilization procedures considerably alter the nutrient concentrations of biosolids. The type of treatment also impacts the rate of nutrient release or mineralization.

It seemed that the use of chemical fertilizer has reached its limit and that a more sustainable approach was required. The use of biosolids for agricultural activities is one way to reduce the burden on conventional chemical fertilization, as N and P are some of the hidden treasures. Germany is one of the first countries which requires sewage plant operators to recycle phosphates based on its new German Sewage Sludge Regulation.

Table 2 presents the main properties of the sewage sludge and its chemical composition in treated and untreated samples.

According to the above, organic matter and nutrients, as the main components of biosolids make them suitable for application in the soil as fertilizers or as organic soil conditioner. Therefore, their use can be, under certain conditions, an alternative to the use of synthetic fertilizers, while they can also be used in soils that are vulnerable to erosion (Moffet et al., 2005). Furthermore, biosolids with a high organic matter content have the advantage that they can be used to remediate metal-polluted regions, by converting metals into less soluble components (Mohapatra et al., 2016). Possible decrease in the aromaticity of the soil can be caused by the biosolids application due to their aliphatic structure as the ^{13}C NMR spectroscopy revealed.

2.2. Types of biosolids

The processes that take place in a Wastewater Treatment Plant, as well as the various ways in which the sewage sludge is treated and stabilized, determine the physicochemical properties of the biosolids. The quality and the quantity of biosolids depends on several parameters such as: (a) the origin of sewage sludge, (b) the age of sewage sludge, (c) the management conditions (pH, temperature, microbiological competition, and residence time), (d) the storage of sewage sludge. The existing parameters, help ensure that biosolids are processed, handled and land-applied in a manner that minimizing potential risk to human health.

Biosolids are classified into four categories based on their post-treatment processes. Their details are mentioned below (Management, 2006):

a. Biosolids cake

Raw sludge is biologically stabilized (most typically by anaerobic digestion), and the liquid biosolids are dewatered to make biosolids "cake". The separation of the water from biosolids is done to obtain a semi-solid or solid product by using dewatering technologies (centrifuges, belt filter presses, plate and frame filter presses, and drying beds and lagoons) (Phillips and Kobylinski, 2012).

b. Biosolids pellets

Raw sludge or liquid biosolids are dehydrated to around ~20% total solids, subsequently heated and dried to 95% total solids to pelletization.

Table 2
Typical chemical composition and properties of untreated/digested sludge (Fytili and Zabaniotou, 2008).

Item	Untreated	Digested
Total dry solids (TS), %	2.0–8.0	6.0–12.0
Volatile solids (% of TS)	60–80	30–60
Ether soluble	6–30	5–20
Ether extract	7–35	–
Protein (% of TS)	20–30	15–20
Nitrogen (N, % of TS)	1.5–4	1.6–6.0
Phosphorous (P_2O_5, % of TS)	0.8–2.8	1.5–4.0
Potash (K_2O, % of TS)	0–1	0–3.0
Cellulose (% of TS)	8.0–15.0	8.0–15.0
Iron (not as sulfide)	2.0–4.0	3.0–8.0
Silica (SiO_2, % of TS)	15.0–20.0	10.0–20.0

c. Lime amended biosolids

The sludge is stabilized by mixing alkaline materials such as lime, cement, or incinerator fly ash and maintained at pH above 12 for 24 h or at temperature 70 °C for 30 min.

d. Composted biosolids

Dried biosolids are composted, to generate a high-quality product appropriate for land use. This is a biological process where organic matter decomposes to produce humus after the addition of a quantity of dry bulking material such as sawdust, wood chips, or shredded yard waste under controlled aerobic conditions.

Beyond the aforementioned classification, biosolids are also categorized into two groups depending on the sanitization they have undergone, the Class A and Class B biosolids. These two different classes have specified treatment requirements for pathogens and vector attraction reduction, as well as general requirements and management practices. More specifically, Class A biosolids are considered as "excellent quality" because they are free of pathogenic microorganisms such as fecal coliforms and *Salmonella* sp. and can be obtained through their thermophilic aerobic digestion at 55 °C. Class A biosolids can also be bagged and sold to the public. On the other hand, mesophilic aerobic digestion at 33 °C leads to biosolids which are classified as Class B, which have with limited applicability to the soil, as they are not completely free of microbiological load compared to Class A biosolids (Gerba and Pepper, 2009). Class B biosolids are unregulated for use, so they cannot be sold or given away in bags or other containers or disposed of at sites used by the public. In the US, the classification of biosolids under these two classes come from the regulations found in 40 CFR Part 503 of EPA for the use and disposal of sewage sludge (US-EPA, 1994). The 1986 Sewage Sludge Directive refers to this classification for EU member states respectively.

Biosolids, depending on their abovementioned classification tend to be used as fertilizers in agriculture, or be stored in lagoons, or generally are used for landfilling, which is the method with the lowest-cost and easiest practice of disposal, but less preferable as is considered as an obsolete method.

3. The beneficial effects of biosolids in agriculture

The organic and inorganic constituents of biosolids are essential for soil and plants (Ahmad et al., 2019; Sharma et al., 2017). When applied to land at the appropriate agronomic rate, biosolids provide several benefits including nutrient addition and improved soil structure while achieving water reuse. Agriculture application of biosolids also can have economic and waste management benefits (e.g., conservation of landfill space; reduced demand of synthetic fertilizers) (Randall et al., 2019). The use of biosolids in agriculture provides the soil with organic matter, which is necessary after continuous harvests, improving the structure of the soil. The benefits of carbon sequestration have also been explored (Torri et al., 2014). More specific, the application of biosolids in the soil improves the physical properties of the soil, increases the porosity, facilitating the aeration and penetration of the root system to a greater depth (Al-Gheethi et al., 2018). Furthermore, the use of biosolids prevents the formation of surface crust, enhances water retention between soil particles (two-phase flow) by reducing evaporation and filtration (Kalavrouziotis, 2016; Lu et al., 2012; Mohapatra et al., 2016). In this way, soil moisture is maintained in the upper layers of the soil, due to their water holding capacity can reduce the need for irrigation water (Laidlaw et al., 2015).

In addition, biosolids enrich the soil with bioavailable nutrients such as N, K, and P, as well as bioavailable S to avoid the shortage often observed after successive harvests (Case and Jensen, 2019; Zamparas, 2021; Zamparas et al., 2019a). It is worth mentioned that since nutrients in biosolids are not as soluble as those in conventional fertilizers, they follow the slow-release mechanism. Thus, biosolids can feed crops more slowly over a longer period, resulting in higher use efficiency and less likelihood of groundwater contamination if the application rate is appropriate (Mohapatra et al., 2016). According to Brown et al. in comparison to conventional chemical fertilizers, biosolids enhanced total N concentration and bioavailable P in soil, with the added benefit that a portion of the N supplied was retained via partitioning to soil organic matter (Malara and Oleszczuk, 2013).

In case of lime-treated sewage sludge, soil pH can be improved, as well as cation exchange capacity, and nutrient content, which is beneficial to crop productivity (Reszel et al., 2007). Biosolids treated by lime are particularly beneficial for soils with low Ca concentration.

As reported by Sakellariou-Makrantonaki and Dimakas for specific types of crops, the use of biosolids for fertilization can increase crop biomass by 13–15% more compared to inorganic fertilizers. The increase of biomass of crops (e.g., sweet sorghum) that are either intended for the production of biofuels such as biodiesel and bioethanol or are alternative feedstock for the production of heat and electricity is an indirect positive consequence of the use of biosolids compared to conventional chemical fertilizers (Sakellariou-Makrantonaki and Dimakas, 2013). Replacing inorganic fertilizers with biosolids further leads to a reduction in the CO_2 footprint concerning the production of synthetic fertilizers, a sector that contributes worldwide to the production and emission of significant amounts of CO_2 (Sharma et al., 2017). A typical example is that despite the reduction that has taken place in recent years, the inorganic fertilizers sector in the EU produced 17.712 kt CO_2 in 2017 (Jiang et al., 2019). CO_2 is known to be the key greenhouse gas associated with climate change (Cassia et al., 2018; Malhi et al., 2021). Relevant studies have shown that between the various methods of disposal of sewage sludge, its reuse in agricultural areas usually has the lowest effect on global warming potential (GWP) (Samolada and Zabaniotou, 2014; Willén et al., 2017). Only in cases where different factors are combined, such as the nature of the sludge or the local climatic conditions, the agricultural disposal may not be considered as the best management option (Samolada and Zabaniotou, 2014).

4. Negative effects of biosolids in the environment

4.1. Heavy metals

Different sludge management methods and techniques are used worldwide and hence the concentrations of heavy metals in sludge vary (Buta et al., 2021; Fijalkowski et al., 2017; Mortvedt, 1995). The problem of biosolids application in agriculture is caused by the high concentrations of heavy metals, which accumulate after years of application and become toxic to plants and consequently to humans and animals due to transport through the food chain (Fijalkowski et al., 2017). The effects of heavy metals on plants depend on several factors, such as plant species, the bioavailability of metals, which is determined by physicochemical factors such as soil type and pH, total concentration of heavy metals in biosolids and soil, organic matter content of biosolids that binds metals, and possible interactions between metals (synergistic or antagonistic) (Kalavrouziotis et al., 2012; Ntzala et al., 2013; Shomar et al., 2013).

Yang et al. determined a linear correlation of common heavy metals (e.g., Zn, Cu, Hg, Pb, Cd), added to soil over a long period, identifying the years of safe use in soil (Yang et al., 2018). Moreover, Yang et al. in 2018 found that the bioaccumulation of heavy metals differs between plants and between shoots and seeds in the same plant because they are up taken in a specific order (Zn > Cu > Cd > Hg > Cr = Ni > Pb > As), as expressed by the values of bioaccumulation factors (BCFs) (Yang et al., 2018). Mossa et al. reported that the benefits of fertilization to soil are inhibited when heavy metal concentrations in it are exceeded (e.g., Zn > 1000 mg kg^{-1}), reducing the biodiversity of soil microorganisms (Mossa et al., 2017). Gomes et al. studied the impact of heavy metals on cardamom (*Lepidium sativum*) plant after the application of sewage sludge, and concluded that the heavy metal content was not higher than the official limits and restrictions. Therefore, the concentrations were not prohibitive for the application of biosolids on at least 90% of the Portuguese territory. Furthermore, no adverse effects on the environment and humans were found (Gomes et al., 2019).

The relation of heavy metals to their uptake by vegetables (cabbage, broccoli) and perennial ryegrass *Lolium perenne* L., under the application of biosolids, was studied in greenhouse experiments, in the Laboratory of Sustainable Waste Management Technologies of Hellenic Open University School of Science and Technology based in Patras, Western Greece (Ntzala et al., 2013). It was found that this relation varied with the plant species and the concentration changes in heavy metals of the plant dry matter and soil, depending on the source of biosolid and the rate of application (Kalavrouziotis, 2017; Ntzala et al., 2013).

The application of biosolids in low doses in agriculture, as expected, does not significantly increase the concentration of heavy metals in soil (Singh and Agrawal, 2008). Literature also shows that the concentration of potentially toxic heavy metals steadily decreases over time after years of repeated sludge application. In Germany, there was a reduction of Cu from 378 mg kg^{-1} to 300 mg kg^{-1} and Ni from 131 mg kg^{-1} to 25 mg kg^{-1}. A reduction of 90% has also been observed in Sweden (Gomes et al., 2019), which can be attributed to the implementation of environmental programs and the introduction of limits for heavy metals in sludge and soil (Kirchmann et al., 2017).

So, it turns out that the uncontrolled disposal of biosolids in soil - instead of improving soil properties - can lead to significant soil degradation, in terms of contamination of surface and groundwater, uptake of plant pollutants with the potential for phytotoxicity, and transport of hazardous toxins through various pathways, mainly through the food chain with the mechanism of bioaccumulation. According to Mossa et al. (Mossa et al., 2017), in addition to the positive effect on soil fertility, biosolids are often the “Trojan horse” for the transport of heavy metals (e.g., Fe, Cr, Mn, Zn, Hg, Pb, Ni, Cd, Cu), nanoparticles, organic micropollutants and pathogenic microorganisms in the soil with a significant impact on the balance of the biosystems (Mossa et al., 2017).

4.2. Organic contaminants

A significant number of studies on this field of organic contaminants has been conducted during the last thirty years. Priority types of persistent organic pollutants have received special attention, including chlorinated dioxins/furans (PCDD/Fs) (Bright and Healey, 2003), polychlorinated biphenyls (Leiva et al., 2010), and Polycyclic Aromatic Hydrocarbons (Taha et al., 2018). More recently, estrogens (Langdon et al., 2014), Chlorinated Napthalenes (Clarke and Smith, 2011), Polychlorinated Alkanes and Nonylphenol (Buyuksonmez and Sekeroglu, 2005) have all been studied in biosolids. Because of their low solubility, these substances tend to precipitate from wastewater and concentrate in sludge (Abad et al., 2005). These organic contaminants can be retained in the biosolids matrix because they have the ability to sorb onto organic matter and bind at the EPS functional groups, due to their lipophilic and hydrophobic properties. In comparison with other organic compounds, organic contaminants are high environmental persistent pollutants, due to their chemical stability, relatively low volatility and high dielectric constant (Torri and Alberti, 2012). Contamination of agricultural lands with persistent organic pollutants has consequences for both plants and soil. In particular, impurities alter plant metabolism, often causing a reduction in crop yields. This in turn, affects the conservation of the soil, as wilting crops cannot protect the soil from erosion.

4.3. Pathogens

Pathogens can survive in the soil environment depending on various factors. These factors are related to the environmental conditions such as temperature and sunlight, the quality of the biosolids i.e., pathogen concentration, content of organic matter, competing microorganisms, concentration of toxic substances, and microbial composition (Dumontet et al., 1999; Sidhu and Toze, 2009). After application to the soil, pathogens are concentrated on the soil surface, in plants, or at a very shallow depth into the soil. Almost 90 to 95% of the microorganisms are concentrated in the upper 5 cm of the soil (Delibacak et al., 2020). Based on their pathogen

Table 3
Pathogen density limits of Class A and Class B biosolids, and restrictions on the application of Class B biosolids (Gerba and Pepper, 2009).

a/a	Standard density limits (dry weight)	Class A biosolids	Class B biosolids
		Salmonella < 3 MPN per 4 g total solids or fecal coliforms < 1000 MPN g^{-1} and enteric viruses < 1 PFU per 4 g total solids and viable helminth ova < 1 per 4 g total solids	Fecal coliform density < 2×10^6 MPN g^{-1} total solids
1	Food crops that do not touch the soil surface (eg apples, oranges, etc.)	Immediate harvest	No harvest for 30 days
2	Food crops that are above the soil but come into contact with it (eg tomatoes, strawberries, etc.)	Immediate harvest	No harvest for 14 months
3	Food crops that are below the soil surface and biosolids have been applied for less than 4 months.	Immediate harvest	No harvest for 20 months
4	Food crops that are below the soil surface and biosolids have been applied for over 4 months.	Immediate harvest	No harvest for 38 days
5	Pastures	Immediate grazing	No grazing for 30 days

content, biosolids are separated into the Classes A or B (Table 3). Class A biosolids generally require a reduction in fecal coliform to less than 1000 MPN g^{-1} or a reduction in *Salmonella* sp. at undetectable levels (Table 3). It is noted that only one of the two conditions of reduction pathogens must be satisfied to be considered as Class A biosolids. Regulations for Class B biosolids require a significant reduction in the concentrations of pathogenic microorganisms until environmental factors such as temperature, sunlight, or desiccation have additional reduced pathogen load (Gerba and Pepper, 2009). Thus, while the application of Class A biosolids is permitted in areas e.g., agricultural and forestry, the application of Class B biosolids is limited, as summarized in Table 3.

4.4. Microplastics

Microplastics, which end up in Wastewater Treatment Plants, are categorized into six basic forms: synthetic fibers, granules, pellets, membranes, foam, and fragments (Fig. 3). Fragments and synthetic fibers are the predominant forms of microplastics in municipal wastewater. The most common polymers that end up in municipal wastewater treatment plants in the form of microspheres, are polyethylene and polypropylene, which are found in 11% to 42% and 3% to 32%, respectively, and they originate from household and personal care products, such as facial scrubs, water bottles, containers, plastic bags, and food packaging films (Lassen et al., 2015; Ngo et al., 2019). Moreover, microplastics in wastewater from the textile industry, appear in the form of microfibers, which are mainly polyethylene terephthalate (up to 42.26%), polyester (79.1%), and polyamide (61.2%), and are widely used in synthetic clothing (Ziajahromi et al., 2017).

When microplastics come into contact with water, their surface is weakened so that they break down into nanoplastics (Karimi Estahbanati et al., 2021). This process depends mainly on the degree of elasticity of the material and the applied shear stress forces (Enfrin et al., 2020). The use of additives in plastics and the adsorption of chemicals alters the chemistry of the surface of the particles and affects the processes of accumulation and

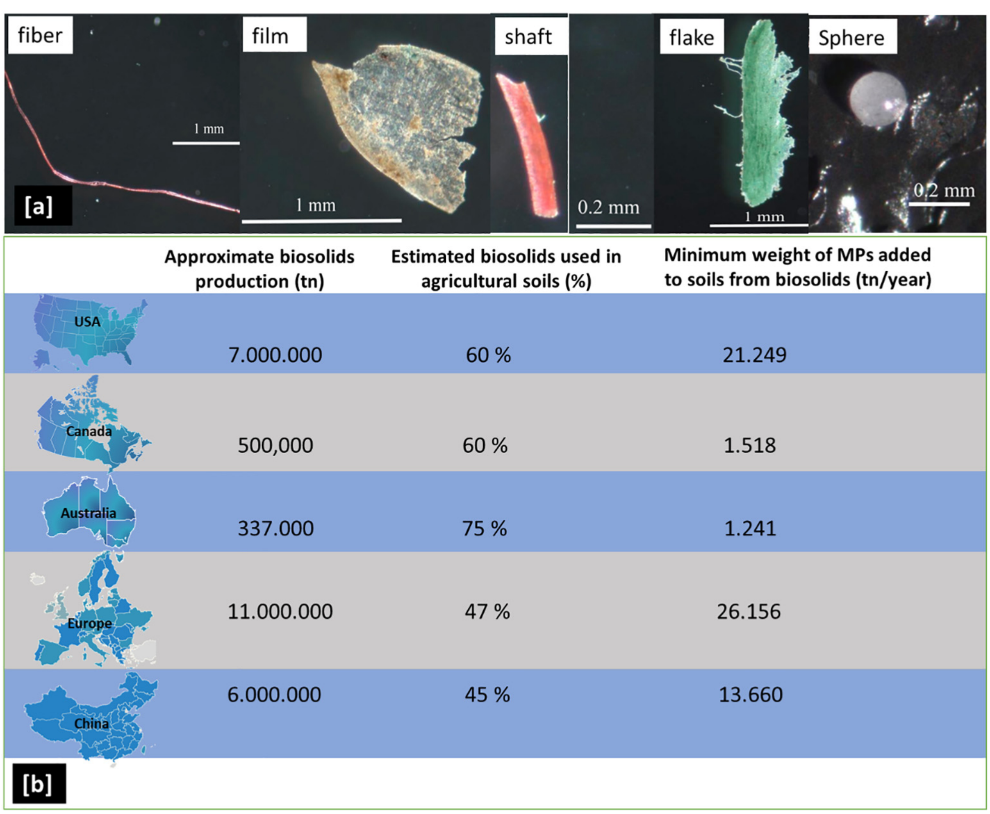

Fig. 3. (a) Stereomicrograph of representative microplastics extracted from the sewage sludge (adapted and modified from Li et al., 2018 with permission from Elsevier), (b) Estimated weight of microplastics added to agricultural soils in five countries (redrawn from Mohajerani and Karabatak, 2020).

dissolution (Hahladakis et al., 2018). The uptake of persistent organic pollutants due to the lipophilicity of plastics leads to the formation of new functional groups on the surface of microplastics and nanoplastics and the change of their surface density (Enfrin et al., 2019). Microplastics that enter Wastewater Treatment Plants can fragment, accumulate microorganisms on their surface, which become more durable and thus prevent Wastewater Treatment Plants processes such as disinfection and filtration (Xu et al., 2021).

The accumulation of microplastics in agricultural soils to which sewage sludge was applied was examined by Corradini et al. (Corradini et al., 2019). Thirty-one plots with similar soil and climatic conditions were sampled, on which sewage sludge had been applied at a rate of 40 t ha^{-1} over ten years. It was found that the content of microplastics in sewage sludge ranged from 18 to 41 parts gr^{-1}, with an average value of 34 parts gr^{-1}. The majority of microplastics were fibers, 90% in sewage sludge and 97% in soil. The study concluded that the long-term application of sewage sludge to agricultural soils leads to the presence of microplastics in large concentrations, which eventually lead to its contamination (Corradini et al., 2019). In a recent study, microplastics were detected in biosolids from two different vendors as well as soils from three agricultural areas in Ontario, Canada, from April to November 2017 (Crossman et al., 2020). High microplastics concentrations ranging from 8.7×10^3 MP kg^{-1} to 1.4×10^4 microplastics kg^{-1} were found in biosolid samples. Lower microplastics concentrations observed in 2nd's Supplier's biosolids which may be due to storage, settling, and supernatant removal before application. Following biosolids application, two fields showed a significant increase in soil microplastics concentrations, with preferential retention of MP fibers over fragments observed, while a decrease in soil microplastics concentrations was observed in the third field. Surprisingly, only one field showed a net increase in microplastics. In all three fields, 99% of the microplastics applied with sewage sludge in 2017 were not detected (Crossman et al., 2020).

Biosolids can be an important route for the release of microplastics from domestic and industrial origin into the natural environment. (Xu et al., 2021) Now, new research has shown that microplastics can penetrate the roots of crops, traveling up the plant into the parts we eat. Two peer-reviewed studies published today highlight the presence of microplastics in our food and call for more research into the relationship between plastic and our health.

4.5. Environmental issues

Major environmental issues related to the use of biosolids in the soil are greenhouse gas emissions (Alvarez-Gaitan et al., 2016; Majumder et al., 2015) and the risk of leaching nutrients through surface runoff to adjacent water systems causing eutrophication (Hanief et al., 2015; Withers et al., 2016). The greenhouse gas that are most related to the treatment, disposal, and reuse of biosolids are CO_2, CH_4, N_2O (Brown et al., 2010). Biosolids contain 30 to 40% carbon, most of which is converted to CO_2 during processing and disposal or use. For example, Archer et al. (Archer et al., 2020) examined the carbon emissions of sludge in small Wastewater Treatment Plants. Carbon emissions ranged from -119 to 299 kg CO_2 equivalents per dry tone of raw sludge. Six small Wastewater Treatment Plants that used land application displayed negative greenhouse gas emissions; the carbon credits gained from fertilizer production avoidance outweighed emissions associated with sludge treatment and transportation. The results encourage that this practice is sustainable from a greenhouse gas emissions perspective (Archer et al., 2020).

High amounts of CH_4 are produced during the treatment, storage, and disposal or use of the biosolids. However, significantly higher emissions occur when sewage sludge ends up in landfills, a management option that in many countries has almost completely stopped (Majumder et al., 2015). In addition, when biosolids is used in agricultural soils, N_2O emissions emerge, as N decomposes and oxidizes. Direct N gas emissions occur in warm and humid soil conditions due to denitrification (Li and He, 2013) and indirect emissions are mainly from NH_3 as N evaporates and is deposited back into the ground leading to further N_2O emissions.

Ammonia is converted to nitrate NO_3^- in the soil, the drains of which are transported through the surface runoff to adjacent surface waters and even to groundwater. Leaching of nutrients could lead to eutrophication of aquatic systems, especially in cases of non-renewable surface waters. However, Withers et al. (Withers et al., 2016) noted that eutrophication risk associated with biosolids application has been overestimated, and they support the more widespread use of this valuable and renewable nutrient resource on some UK soils (Withers et al., 2016). Moreover, Hanief et al. in 2015 assuming equivalent load of TN and TP in biosolids-amended soils and inorganic fertilizers evaluated their loss of N and P to the aquatic systems., concluding that the load of N and P from biosolids may contribute relatively less to eutrophication than inorganic fertilizers (Hanief et al., 2015).

5. Current legislation for biosolids management in Europe

The management of sewage sludge is becoming an issue of growing importance. In all countries of the European Union, directives are introduced based on which each member state has to create relevantly. According to European regulations management methods including storage (Smith, 2009) are now being replaced by approaches leading to waste stabilization and safe recycling (Management, 2006) (Fig. 4).

The gradual application of urban wastewater treatment technologies laid down in Directive 91/271/EU for all EU Member States has led to increasing the amounts of sludge requiring management (Collivignarelli et al., 2019). This increase is mainly due to the practical implementation of the Directive as well as the slow but steady raise in the number of households linked to the sewerage network as well as escalating the degree of treatment (Silva et al., 2014).

In the last years, the main option for the reuse of biosolids is the application on agricultural land. This practice is controlled in different ways at the European level, due to the implementation by the Member States of Directive 86/278/EU, which allows the reuse on land only for biosolids of good quality. However, Directive 86/278/EU does not provide specific information about pathogens, or relevant guidance for appropriate processing methods for removing them. It only refers either to cases where requirements have not been adequately applied or to users who did not comply with hygiene rules (Hudcová et al., 2019). For this reason, every country with respect to Directive 86/278/EU provides different requirements for heavy metals, pathogens, and organic micropollutants both in biosolids and soils. Significant differences are also clear in other aspects of the regulation among Member States for the maximum amount of biosolids spread on land, about the type of soil where the use of biosolids is prohibited, or the treatment requirements. Specifically, from the twenty-seven (27) EU Member States, sixteen (16) incorporated stricter criteria for the concentrations of heavy metals in sludge, compared to those of Directive 86/278/EU, while ten (10) out of the twenty-seven (27) Member States set stricter criteria for the concentrations of heavy metals in soil. For example, in France and Spain, it is necessary to analyze agronomic parameters of biosolids such as concentrations of organic matter, phosphorus, and nitrogen, while in Italy values are set at Organic Matter >20%, Total Nitrogen >1.5%, and Total Phosphorus >0.4% of the dry substance. In Sweden and Latvia, a maximum annual limit of phosphorus and nitrogen that can be dispersed on agricultural soils is also defined (Lamastra et al., 2018). Germany, Denmark, Sweden, and Austria have set limits for the presence of organic pollutants in biosolids (Lamastra et al., 2018). The UK has adopted regulatory requirements to impose treatment processes, in order to achieve the appropriate biosolids quality for land application. The UK has adopted a set of restrictions based on the study of the decomposition of pathogens in treatment and recycling processes, the so-called Safe Sludge Matrix (Tyson, 2002). In this manual, two levels of treatment are necessary to achieve specific *Salmonella* spp. and *E. coli* serotypes in sludge. The improved quality of the processed sludge is achieved as a result of a treatment comprising at least one pasteurization cycle, usually including a

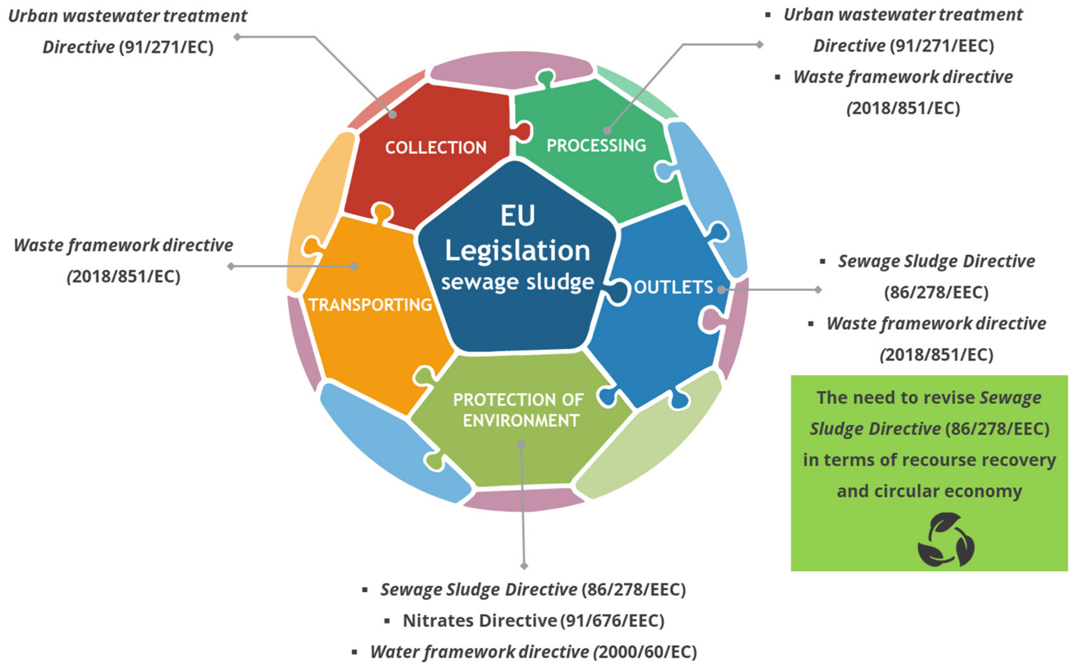

Fig. 4. Graphical overview of EU legislation concerning sewage sludge.

thermophilic step to ensure the removal required. The sanitized sludge can be used beneficially to agricultural land by limiting public health risks (Pascual et al., 2010; Pascual et al., 2008).

Collivignarelli et al. in 2019, pointed out that Directive 2018/851/EU provided a waste hierarchy that shall apply as a priority order in waste prevention and management legislation and policy: prevention (e.g., minimization techniques), preparing for reuse (e.g., chemical or biological stabilization), recycling (e.g., resource recovery), other recovery (e.g., energy recovery), and disposal (e.g., landfilling) (Collivignarelli et al., 2019).

However, the new Directives of 2018 included Directive 851 do not plan a revision of the old sludge directive 86/278. According to the Commission, this directive does not require any update. In the document "ex-post evaluation of certain waste stream directives", the sludge directive was considered to be effective, efficient, relevant, and coherent with other EU legislation. Nevertheless, it also appeared that a directive adopted 30 years ago does not fully meet current needs. It should therefore be re-examined taking into account EU action plans on the Circular Economy, the reduction of fertilizers, and the biodiversity strategy for 2030. The Action Plan on the new Circular Economy adopted in March 2020 emphasizes the need for this revise (Domenech and Bahn-Walkowiak, 2019).

To promote Circular Economy, legislation regarding the recovery of materials from sludge may be required (EurEau, 2021). The Fertiliser Regulation is being revised to allow certain products recovered from sewage sludge to be authorized on the EU market. The JRC report, proposing criteria for "STRUBIAS" materials (STRUvite/recovered phosphate salts, BIochars/pyrolysis materials, ASh-based products), was published in 2019 and is the basis for new Component Material Categories (CMCs). These new CMCs approved in 2021 and added into the Fertiliser Regulation annexes of EU Regulation 2019/1009. Struvite and ash-based products will be authorized as ingredients for EU fertilizers, except for biochar which is not permitted from sewage sludge. This will allow Wastewater Treatment Plant operators to potentially enter this market through phosphorus recovery technologies (EurEau, 2021).

Finally, the challenge of ensuring food systems is an additional critical factor for the necessity of reusing biosolids. Global food security for a growing population will depend on finding new sources of phosphorous to improve crop yields. In the framework of the new Green Deal (European Commission, 2020), the EU's "Farm-to-Fork" policy poses ambitious objectives for agro-food system sustainability, including a reduction of 50% in nutrient losses and 20% for fertilizer use before 2030, to improve nutrient stewardship (European Commission, 2020; "The New Circular Economy Action Plan", 2020).

Along with the application of the EU directives, aiming at the most effective and safe reuse management of biosolids, the use of a Decision Support Systems can be very useful practical tools, towards accomplishing rational and safe reuse. The School of Science and Technology of Hellenic Open University in Patras, Greece, has recently released an online Decision Support System, which is a clever software, that helps to ensure biosolids and wastewater reuse in agriculture, in order to reduce the application of fertilizers, to minimize the toxicity problems from the accumulation of heavy metals on the growing plants, and to protect the environment from pollution (Koukoulakis et al., 2020).

6. Conventional and emerging methods for treatment

6.1. The obsolete methods of landfilling and storage

Until recently, landfilling was a panacea in the management of sewage sludge. Landfilling is currently restricted in the EU by Directive 1999/31/EC, which aims to limit the quantity of biosolids to 35% compared to the amount landfilled in 1995. In practice, it has been significantly reduced or abandoned altogether by the institutional framework of the Member States themselves. It is classified as the worst option of Life Cycle Assessment, which should be the basic principle for waste management (Krogmann et al., 1999; Paz-Ferreiro et al., 2018).

Storage of sewage sludge may be temporary or permanent (Aarab et al., 2006). The storage method was formerly used as the only method of treating sewage sludge, but in hot climates, it required long periods of stabilization and was gradually abandoned as inefficient (Carrington, 2001). Cases of bacterial regrowth after long storage periods have been reported not only in untreated sludge but also in stored disinfected biosolids, (Arthurson, 2008) adding another deterrent factor of this obsolete method.

6.2. Conventional methods

Nowadays, the elimination of pathogens requires sanitization and stabilization treatments, which are divided into three broad categories:

biological, chemical, and thermal. The most common are aerobic and anaerobic (mesophilic or thermophilic) digestion, composting (biological), drying and heat treatment (thermal), and stabilization with lime or its variants (chemical). Different treatment technologies achieve varying levels of pathogen removal or inactivation, ranging from the relatively moderate ability of mesophilic anaerobic digestion to reduce *E. coli* concentrations to the high degrees of metabolic cell inactivation by heat treatment.

6.2.1. Aerobic and anaerobic digestion

Biological treatment with aerobic and anaerobic digestion is the most common practice in all European Member States, with the second method being preferred due to biogas production (Kelessidis and Stasinakis, 2012). Anaerobic digestion involves the decomposition and biodegradation of organic matter under controlled conditions, to produce CH_4. The principle of anaerobic digestion is to reduce the fermentation capacity of biodegradable waste, increase biogas production and ensure that the digested product can be used for compost. Anaerobic digestion takes place at mesophilic temperatures of 30–35 °C, with an average residence period of 15 d, or at thermophilic temperatures of 45–65 °C for 20 h (Kim, 2020). Since anaerobic digestion in mesophilic temperatures is disadvantaged in terms of removing pathogens, requires longer residence times. However, treatment is still not sufficient to ensure the desired removal of the various species of pathogens (Kim, 2020). Anaerobic digestion presents the advantage of biomass energy production, which along with the reduction of the processing costs also leads to a reduction in CO_2 emissions. Aerobic digestion relies on endogenous respiration, i.e., cell degradation observed at small ratio of F/M (food/microorganisms). Mesophilic aerobic digestion takes place between 30 and 35 °C, while thermophilic usually between 45 and 65 °C. Liu et al. (2012) suggest a temperature in a range of 45–55 °C for thermophilic aerobic digestion, leading to class A biosolids in terms of fecal coliform content and *Salmonella* serotypes (Liu et al., 2012). Digestive methods, excluding thermophilic aerobic digestion, are included in the Processes to Significantly Reduce Pathogens as they lead to biosolids which are classified as class B in the USA, with limited applicable on the ground (US-EPA, 1994). According to Arthurson (Arthurson, 2011), optimization of the methods used requires an additional step for the qualitative upgrade of class A biosolids. Thermophilic aerobic digestion belongs to the Processes to Further Reduce Pathogens and leads to class A biosolids (US-EPA, 1994).

6.2.2. Composting

Composting is the biological degradation and stabilization of organic matter under conditions that allow the development of temperatures in the range of 50–60 °C, which is ensured from biologically produced heat giving a final product satisfactorily stabilized for storage and use as a soil conditioner without environmental effects (Shilpa and Girija, 2021; Stephens et al., 2017). The process requires special conditions of aeration and humidity. The efficiency of composting in the conversion of fermentable organic matter contained in sewage sludge into a stabilized odor-free product is related to biological activities that take place mainly in the solid rather than the liquid phase (Angelova and Shilev, 2021). The stabilization phase in the composting process is related to the period in which significant amounts of cellulose and progressively lignin decompose leading to a reduction in the biologically required oxygen of the composting material (Viau and Peccia, 2009). Temperatures in this phase normally range between 35 and 55 °C. For most composting systems the stabilization phase takes place after the sanitization phase. The three main systems of composting that are found in literature are the series system, the system of ventilated static piles and the closed bioreactors (Fournel et al., 2019). The composting process is preceded by dehumidification to produce biosolid cake. The composted material can be sludge mixed with bulk materials such as green waste, small pieces of wood, etc. (Kamal et al., 2017). This method can lead to Class A biosolids with unrestricted application to the soil (US-EPA, 1994). However, the success of the method requires a considerable land area and maturation time as well as maintaining constant temperature levels (≥55 °C) at all points of the piles (Kamal et al., 2017). The efficiency of composting is based on the heat generated during the process and on the fact that during the mesophilic and then thermophilic steps, the composting mass is transformed into a substrate unsuitable for the survival of most pathogens. Removal of pathogens is also associated with factors such as ventilation, humidity, pH, nutrient background, competition with native microorganisms (Matiz et al., 2015). It has been shown that the reduction of pathogens observed during composting depends on the quality of the initial sewage sludge, the additional green waste, as well as on the conditions of the process (temperature, aeration, and residence time). For *Salmonella* spp. serotypes the reduction is significant when the temperature is higher than 65 °C. It is also reported in the literature that sludge becomes safe when a compaction period of two weeks has elapsed at a temperature of ≥55 °C or 1 week at ≥65 °C. Composting has better results in reducing *E. coli* and *Salmonella* spp. than in enterococci. It has also been shown to be effective in reducing helminths when applied conditions are: 4 d at 60–76 °C or 8 d at 60–70 °C. Based on the above results, composting is a satisfactory method of hygiene in the removal of pathogens and reuse of agriculture by-products for soil-conditioners production.

6.2.3. Lime

Treatment of biosolids with lime (CaO or $Ca(OH)_2$) is a stabilization method that achieves reduction of pathogenic microorganisms, inhibition of their regeneration, and reduction or even elimination of odors (Krach et al., 2008). On the other hand, the addition of lime reduces the solubility of P and leads to lower concentrations of total P (Krach et al., 2008). The temperature rises to 55–70 °C in the lime-mud mixture and the pH value goes above 12 destructing the pathogens. It is usually recommended to add 30% lime to the dry matter of the sludge, otherwise, fermentation will occur. Stabilization with lime achieves sanitization, increases the dry matter content, and thus enables better management. According to Arthurson in 2011, alkaline methods result in Class A biosolids (Arthurson, 2011). However, the method using $Ca(OH)_2$ is not included in the EU recommended treatments for pathogen-free sewage sludge. Moreover, the method is not suitable for calcareous soils and at the same time increases the volume of the products (Venglovsky et al., 2006). However, it has an advantage over other stabilization methods that use chemical disinfectants, because they have less impact on the environment. According to Zrubková in 2017, the lime stabilization method is superior to methods such as anaerobic digestion and pasteurization in eliminating pathogens. This kind of stabilization has a low risk of pathogen reoccurrence. The disadvantage of the method is the reduction of total P, as well as reduction of N concentration in biosolids due to NH_3 emissions, leading to a low quality soil conditioner in terms of nutrients (Zrubková, 2017).

6.2.4. UV irradiation

UV disinfection has been described as a suitable technology for the inactivation of coliforms and *Salmmonella* spp. The principle of a UV disinfection system is to destroy the genetic material of the bacterial cell retarding its ability to reproduce. The effectiveness of a UV disinfection system depends on the characteristics of the wastewater, including the concentration of the pathogens, the intensity of the UV and the treatment time (Safoniuk, 2004). The major drawback of the method is that the high irradiation doses required are not yet economically feasible (Safoniuk, 2004). On the contrary, Asgari-Lajayer et al. in 2019 estimated the dose of γ-irradiation to produce class A biosolids, but concluded that it is not possible to completely remove the pathogens. They also found that coliform germs were more resistant to γ-radiation than total bacteria (Asgari Lajayer et al., 2019). There is some evidence that the operating costs of UV irradiation may be lower than incineration. However, the process is still under investigation.

6.2.5. Pyrolysis

Pyrolysis has a lower carbon footprint than other various treatments, making it a promising method for biosolids management (Paz-Ferreiro et al., 2018). Compared to incineration, pyrolysis produces less exhaust that must be controlled. At the same time, the amount of acidic gases and dioxins formed is considerably reduced. Pyrolysis is divided into two types: slow pyrolysis and fast pyrolysis. Slow pyrolysis maximizes the solid fraction operating at long residence times and slow heating rates (Domínguez et al., 2008). Fast pyrolysis performs thermo-chemical conversion at a high rate (around 100 °C min^{-1}), maximizing the liquid and gaseous fractions. The conversion of wet biosolids to pyrolysis can result in a higher yield of Pyrolysis gasoline or Pygas and a higher hydrogen content than dry biosolids (Domínguez et al., 2008).

6.3. The emerging process of Cold Atmospheric Pressure Plasma

To further ensure the respect of the abovementioned requirements for treatment and stabilization of biosolids, novel, plasma-based methods have been proposed (Pasolari et al., 2020; Svarnas et al., 2020). Briefly, plasmas of electrical discharges are gaseous ionized phases containing free electrons, positive and negative ions (atomic or molecular), UV-NIR photons, excited atoms and molecules, feedstock gas neutrals, free radicals, metastables, ozone, etc., being all together quasi-neutral since charges balance on a macroscopic scale. While the charge particles can respond to external electromagnetic energy fields and transport energy, the fluid properties of plasmas are enhanced by particles setting up internal self-consistent electric and magnetic fields, resulting in collective effects like flows, waves, instabilities, and self-organization. Each species may have independent energy distribution, not necessarily in equilibrium with other species. The internal energy is composed of thermal, electric, magnetic and radiation fields, whose relative magnitudes allow the plasma state to exist in an extended, multi-dimensional parameter space (John, 2005). Thirty years ago, US National Council report (NRC: National Research Council, 1991) indicated a number of important uses of plasmas including laminations, sterilization of medical products, gas discharges for lighting and lasers, high-definition television, isotope separation, electrical power switching, cutting and welding technology, plasma-based propulsion systems, and environmental clean-up (Van Veldhuizen, 2000).

Plasmas are produced by the introduction of energy (usually supplied by an external electromagnetic field) to a gas being neutral initially (e.g., air, argon, helium, oxygen etc., or mixtures of them). Depending on the operating gas or mixtures, gas pressure, features of the driving electromagnetic field, electrode configuration, plasma reactor design etc., a wide variety of different plasmas may be produced for tailored applications. A main categorization of plasmas may be realized in terms of electron density n_e, and electron and ion temperature T_e and T_i, respectively. Indicatively, n_e varies over 28 orders of magnitude from 10^6 to 10^{34} m^{-3}, and kT_e can vary over seven orders from 0.1 to 10^6 eV (Chen, 2016). Roughly, plasmas may be distinguished to cold ($T_i \approx T_{gas} \approx 300$ K; $T_i << T_e \leq 10^5$ K), thermal ($T_e \approx T_i \approx T_{gas} < 2 \times 10^4$ K), and hot ($T_i \approx T_e > 10^6$ K) ones (Harry, 2010). Most of the laboratory made plasmas lie in the first category, making plasma an entire "laboratory of dry, low-temperature chemistry". The continuously growing interdisciplinary nature of the low-temperature cold plasma field and its equally broad range of applications are making it increasingly difficult to identify major challenges that encompass all the sub-fields and applications (Adamovich et al., 2017).

A great part of the increasing interest in cold plasma applications is related to the dielectric-barrier discharges (DBDs). DBDs gave the possibility of sustaining stable, engineerable, homogeneous, out of thermal equilibrium plasmas at atmospheric pressure (vacuum free), leading thus to an enviable track record of atmospheric pressure cold plasma technology. The DBD gap usually includes one or more dielectric layers located in the current path between two metal electrodes. This approach prevents formation of sparks and current growth in the channels formed by streamers, and thus overheating, generation of local shock waves, and noise (Fridman and Kennedy, 2004). Important contributions in fundamentals and industrial applications of DBD were made by ABB, although this discharge was first introduced by Siemens. It has been shown that this discharge occurs in a number of individual tiny breakdown channels (current filaments), which are nowadays referred to as microdischarges. From a physical point of view, these microdischarges are actually streamers that are self-organized taking into account charge accumulation on the dielectric surface (Fridman and Kennedy, 2004).

In terms of environmental applications, DBD has extensively been used for soil (Aggelopoulos et al., 2016) water (Ramdani et al., 2015) and gas (Holzer et al., 2002) cleaning from organic, inorganic, and biological pollutants. Although thermal plasmas have also been tested for industrial and wastewater sludge treatment (Ali et al., 2016), the exploitation of DBD-based cold plasmas for biosolid stabilization is limited. On the top of that, a very specific type of DBD, i.e., the so-called FE-DBD, can be introduced for such a stabilization process. FE-DBD stands for the term "Floating Electrode Dielectric Barrier Discharge" and is distinguished from the classic DBD since, the one of the electrodes is the dielectric-protected powered electrode and the second active electrode is the specimen under treatment which is not grounded and remains at a floating potential – without the specimen the discharge does not ignite (Fridman, 2008). FE-DBD plasmas have been proven to be highly reactive media and our group has demonstrated their efficiency to disrupt bio-membranes (Svarnas et al., 2017). In the case of recently developed biosolid treatment reactors (Pasolari et al., 2020; Svarnas et al., 2020), this direct treatment forces the discharge current to pass through the biosolid and at the same time exposes it to the various plasma induced chemically reactive species (radicals, photons, metastables, ozone, thermal fields, etc.). Thus, unlike to previously established treatment methods, this dry chemical environment contains many synergetic factors for biosolid sanitization and stabilization.

6.4. The Achilles heel of treatment processes

All the above-mentioned treatment methods have been summarized by the authors into Table 4, where all the methods of biosolids sanitation collected and divided into categories based on their processing technologies, emphasizing their main drawbacks. From the table it is clear that over the years many methods have been developed for the stabilization and sanitization of biosolids, using each one of them different processing technologies. The table also includes the main parameters of each method and information regarding the recognized body that proposes them, if any. Processes have furtherly categorized into biological, chemical, thermal, radiation, combinational and advanced methods. Further to that, three indicators of positive evaluation of each method were added in terms of economic scalability, sustainability and technical maturity and industrial scalability, which are highlighted in shades of green going from low positive to fairly positive and then highly positive.

Each method has its Achilles heel, and the appropriateness of the method lies in what the goal each time is. Literature, also concludes that none of them is fully integrated by meeting all the basic criteria, such as the elimination and non-reappearance of pathogens, the fast processing with low operational and energy costs and the final by-product to be class A.

Conclusively, biological methods have the most positive reciprocity in terms of sustainability, reuse indicators, technological maturity, and industrial scalability. The methods with the lowest positive indices are those of radiation. The chemical methods are low positive in the index of sustainability and reuse, while quicklime and hydrated lime are the only two in the whole table that have a highly positive economy index on a large scale, among other disadvantages. New advanced sustainable technologies, such as cold plasma, need to be further studied to apply on a large scale, as it's fast, energy efficient and has excellent rate of sanitation according to the results so far.

Sanitation is practiced worldwide with conventional biological, chemical and thermal methods and certain combinations thereof. Biological methods show limited inactivation of coliforms (mesophilic digestion), egg parasites (mesophilic and thermophilic digestion) (Czerska and Smith, 2008; Vochozka et al., 2016) and risk of recurrence of pathogens after sanitization (composting) (Arthurson, 2008; US-EPA, 1994). At the same time, they present increased demands in time and space, while their disadvantage is the smells (aerobic processes) (Czerska and Smith, 2008; Liu et al., 2012).

Table 4
The Achilles heel of approaches on various biosolids treatment technologies.

PROCESSING TECHNOLOGY	TREATMENT METHOD	DRAWBACKS	PROPOSED BY	INDICATORS OF EVALUATION: Economic Scalability	Technical Maturity & Industrial Scalability	Sustainability	REFERENCES
BIOLOGICAL METHODS							
Anaerobic digestion	Thermophilic (50-55°C)[1] Mesophilic (30-35°C)[2]	Mesophilic: insufficient reduction of coliforms. Extended stay. Both types are ineffective in pest eggs	EU, US EPA (PSRP)[1], US EPA (PSRP)[2]				(Czerska & Smith, 2008; Vochozka et al., 2016)
Aerobic digestion	Thermophilic (45-55°C)[1] Mesophilic (30-35°C)[2]	Factors e.g., ammonium production, microbial exozymes affect efficacy. No reduction of pests. Low reduction of enteroviruses	EU, US EPA (PFRP)[1], US EPA (PSRP)[2]				(Czerska & Smith, 2008; Liu et al., 2012)
Composting	Series / Vent Piles / Reactors (50-60°C)	Significant costs, odors, space and time requirements. Demanding procedure (stable temp.). Risk of bacteria reappearing	EU, US EPA (PFRP)				(Arthurson, 2008; US EPA, 1999)
Drying	>80°C	Significant energy requirements, risk of explosions, reduction of susceptibility to pathogens	EU, US EPA (PFRP)				(US EPA, 2006; Carrington, 2001)
Air Drying	Sand beds, basins / lagoons / artificial wetlands	Reduction of E. coli between 81.48 - 99.13%. Recurrence of Salmonella serotypes 3 weeks after treatment	EU, US EPA (PSRP)				(Czerska & Smith, 2008)
CHEMICAL METHODS							
Quicklime (CaO)	Mixed with sludge	Possibility of recurrence of pathogens (pH <9.5). Decrease of N, volume increase.	Proposed by: EU, US EPA (PSRP)				(US EPA, 1994; Venglovsky et al., 2006)
Hydrated lime [Ca(OH)2]	Mixed with sludge	Inhibits microbial activity - not accepted by the EU for pathogen sanitization	US EPA (PSRP)				(Carrington, 2001; US EPA, 1994)
Cement Kiln Dust + unaltered lime /Fly ash	Under investigation	Big sedimentation rate - volume increase (pH 8.0 - 9.0)	-				(Carrington, 2001; Elbaz et al., 2019; Smith & Campbell, 2000)
Urea/ NH4OH/ NH3	Under investigation	Inactivation delay. No effect of NH3 on sporogenic bacteria, odors	-				(Vinneras et al., 2003)
Paracetic acid (PAA)	Under investigation	Low reduction (10%) of pests from storage. Bacterial regrowth 24h after treatment. Reduced biogas production	-				(Koivunen & Heinonen-Tanski, 2005)
THERMAL METHODS							
Thermal process	≥180°C	Energy requirements, affect the structure of the biosolids	US EPA (PFRP)				(US EPA, 1994)
Pasteurization	100 °C < T < 65 °C (usually 70 °C)	It fails to eliminate bacterial endospores. Significant costs	US EPA (PFRP)				(Arthurson, 2008; US EPA 1994)
RADIATION METHODS							
Electron beam accelerator	Radiation β	Demand for high doses for class A biosolids. Not economically viable. Unsuccessful removal of pathogens.	US EPA (PFRP)				(Engohang-Ndong et al., 2015; Hossain et al., 2018)
^{60}Co/ ^{137}Cs ion source	Radiation γ	Demand for high doses for class A biosolids. Not economically viable. Unsuccessful removal of pathogens.	US EPA (PFRP)				(Asgari Lajayer et al., 2019)
COMBINATIONAL METHODS							
▲ Heat treatment with water vapor/ Pasteurization + Mesophilic (at least) anaerobic digestion		Significant requirements in space and time of maturation	EU				(Carrington, 2001)
2 pasteurization cycles or Pasteurization + digestion/ composting/ mixing with lime	Under Investigation	Delays between stages favor the regrowth of endospores	-				(Arthurson, 2008)
ADVANCED METHODS							
Cold plasma	Atmospheric pressure discharge (FE-DBD), 380K	Not yet implemented on a large scale	-				(Svarnas et al., 2020)
Thermal hydrolysis + digestion	130-180 °C	Reduces energy production when digested, by 20%	PFRP-equal				(Sahu et al., 2022 ; Kepp et al., 2000; Pickworth et al., 2006)

Processing technology
▲ Method
Proposed by
↓ Drawbacks

PFRP: Process to Further Reduce Pathogens
PSRP: Process to Significantly Reduce Pathogens
EPA: Environmental Protection Agency

Economic Scalability
Technical Maturity & Industrial Scalability
Sustainability

Chemical methods have significant disadvantages such as sedimentation (Carrington, 2001; Elbaz et al., 2019; Smith and Campbell, 2000) - deposition which increases the volume of the final product and reappearance of bacteria when the alkaline pH drops below 9.5 (mixing with lime) (US-EPA, 1994; Venglovsky et al., 2006), the additive chemicals have increased cost and impact on the environment, while in the case of ammonia disinfectant action is slow to start, the efficiency in removing pests is low and there is a nuisance of odors (Vinnerås et al., 2003). In addition, a recurrence of bacteria is recorded 24 h after hygiene with PAA and the risk of outbreaks (Koivunen and Heinonen-Tanski, 2005).

Thermal methods show significant operating costs to maintain the temperature at the required residence time, inconsistency in efficiency which depends on the structure of biosolids and the way the particles approach heat (US-EPA, 1994). There is a risk of re-growth of bacteria (pasteurization) and their combination with other methods (biological and/or chemical) and a short period of time between successive processing steps is required.

The β and γ rays have not yet achieved complete inactivation of the bacteria and the required absorption doses make them economically unviable (Asgari Lajayer et al., 2019; Engohang-Ndong et al., 2015; Hossain et al., 2018). The Advanced technology of Thermal hydrolysis in the treatment of biosolids is usually combined with digestion, the so-called Campi THP hydrolysis, which takes place at 130-180 ° C and is considered an equivalent method to PFRPs. It does, however, cause a 20% reduction in energy production when digested (Sahu et al., 2022; Kepp et al., 2000; Pickworth et al., 2006).

From the existing literature and the findings of researchers until today, it appears that cold plasma technology results in the removal of bacteria at levels below the detection limit for most species. This emerging technology which is still being investigated, especially in terms of the chemical properties of biosolids, is environmentally friendly, the energy consumed is low and the processing time is short. It could therefore be an economically feasible and sustainable option for integration into urban wastewater treatment plants.

7. Environmental and economic benefits

In order to promote the reuse, recycling and recovery of wastes, the European Commission adopted an ambitious Circular Economy Package (EC, 2018). The aim of Circular Economy is closing the loop of product lifecycles keeping their added value for as long as possible and eliminate waste with obvious benefits for the environment and the economy. The urban Wastewater Treatment Plants can be an important part of circular sustainability thanks to integration with the concept of reuse of biosolids (Neczaj and Grosser, 2018).

According to Calkins (2008), each year more than three billion metric tons of raw materials are used to make building materials and products worldwide. In addition, the construction industry faces the problem of depletion of natural materials such as pumice, slag, crushed stone and clay. Eco-friendly fired clay building bricks can be made by biosolids. The biosolid-bricks production is less energy consuming manufacturing procedure, reducing the need of mine clay for brick making purposes. Biosolids can be also the construction materials for lightweight artificial aggregates, and cement-like materials, which can be considered as a good practice because the waste is converted into useful materials while resolving the very important problem of disposal and raw resource reduction (Ukwatta et al., 2015). Beyond that, the economic aspect has become important in recent years. Researchers noted that the cost of sewage sludge management is approximately 50% of the total running costs of the Wastewater Treatment Plants (Bertanza et al., 2015; Collivignarelli et al., 2015).

In 2017, the American Department of Energy, on its report (U.S. Department of Energy, 2017) mentioned that across the country, municipal Wastewater Treatment Plants are estimated to consume more than 30 terawatt hours per year of electricity, which equates to about $2 billion in annual electric costs. This operating cost could be greatly reduced by using biosolids as a raw material for gas production for electricity generation.

8. Conclusions & discussion

The simultaneous requirement to manage resources and wastes in a more rational way has meant that many communities worldwide have begun to search for long-term alternative solutions instead methods employed to dispose of their waste and to produce energy.

Biosolids have the potential to meet many needs of society and become a part of circular economy. They can be used as soil conditioners and in agriculture, to reduce or replace chemical fertilizers with small environmental fingerprint. Biosolids can be used as an energy source at a time of energy crisis. Also, they can be an important source of phosphorus, providing a solution to the global shortage of stocks.

Emerging pollutants, microorganisms, microplastics, greenhouse gases and nutrition overload of water bodies are the hidden risks of biosolids usage, like another Trojan Horse. For example, soil carbon sequestration, improvement of soil quality in terms of physical, chemical and biological fertility are important remarks from the beneficial use associated with the use of biosolids in soil, although the presence of contaminants may impact soil microbial communities on the long term. However, the benefits from biosolids application have to be weighed against potential deleterious effects, where the Trojan Horse can be the beautiful Helen, as long as the state and the scientific community, deal biosolids in an epochal paradigm shift: as an opportunity and not as a problem.

For this reason, it is absolutely necessary to strictly control the treatment plants for the technology and the methods they use in practice, as well as the quality of biosolids that they produce, in order to prevent easy practices that, in addition to being obsolete, pose risks.

Our comprehensive review discovered that a strategy for revising the existing legislation for sustainable biosolids management and emerging technologies for sanitation and stabilization of biosolids, is of great importance and will lead society in a new era. The exculpation of their use will come through their proper management at the Wastewater Treatment Plants according to the demands of society.

This comprehensive review article is believed to enhance the understanding of the various treatments biosolids undergone on the Waste Water Treatment Plants and that every method hides disadvantages. There is a need of introducing technologies that do not pose risks and nuisances to the biotic and abiotic environment, show high efficiency in a short processing time and operate with low energy and space requirements. Thus, would be useful to the readers of the relevant communities and various stakeholders to investigate potential technologies to eliminate these drawbacks.

In closing, we adopted the scoping literature review method and endeavored to incorporate relevant literature from different research fields. But there is a possibility that some factors or topics are missing in our framework which might be discovered in future research.

In Greek mythology, the Trojan horse enters the city of Troy and the Greeks win the war. After a fruitless 10-year siege, the Greeks at the behest of Odysseus constructed a huge wooden horse and hid a select force of men inside, to gift it to Trojans as a sign of goodwill and peace. But, having achieved to enter the well-kept gates of Troy, Greeks entered and destroyed the city. Metaphorically, a “Trojan horse” has come to mean any method that hide true intentions or can become dangerous despite looking good at the outside.

Concluding this comprehensive review article, biosolids can be paralyzed as a modern Trojan horse, as they are quite likely to pose risks if they have not previously been properly treated, stabilized and sanitized for disposal or reuse. Having neutralized the dangerous soldiers from the organic matter of biosolids such as pathogenic microorganisms, heavy metals, odors etc. then, this rich in nutrients by-product is a coveted bride for the soil like another Beautiful Helen and is very likely that in the near future, with the new emerging technologies being applied on a large scale, the management and disposal of solids will no longer be a difficult matter to handle for the stakeholders, but an object of desire offering satisfaction to whoever wins the trophy.

CRediT authorship contribution statement

A.K. conceived of the presented idea. E.I. set the contents and defined the search keywords of the review. I.K. encouraged A.K. to investigate on negative effects of biosolids and supervised this manuscript. P.S. developed the theoretical formalism of Cold Atmospheric Pressure Plasma treatment emerging process. A.K. wrote the manuscript with support from E.I.. All authors discussed the review and contributed to the final manuscript.

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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