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REVIEWS

Methods to alleviate the inhibition of sludge anaerobic digestion by emerging contaminants: a review

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Abstract

The rising occurrence of emerging contaminants in sludges both inhibits the anaerobic digestion of sludges and induces health issues when sludges are recycled in agriculture, calling for methods to remove contaminants. Here we review emerging pollutants in wastewater treatment plants, before and after anaerobic digestion. We present their inhibitory effects and remediation methods to alleviate inhibition. Pharmaceuticals have been detected in about 50% of the sludge samples. Sewage sludge contaminants include 19% of diuretics, 16–21% of lipid-modifying agents, hydrochlorothiazide, diclofenac, furosemide, clarithromycin, atorvastatin, and carbamazepine. Levels of antibiotics, azithromycin, ciprofloxacin, and estrone range from 500 to 600 ng/g in sludges from wastewater treatment plants. Remediation methods comprise electrooxidation, ultrasonication, thermal hydrolysis, ozonation, and bioaugmentation. Fermenting the sludges with acidogenic bacteria reduces the level of emerging pollutants in the supernatant. Nonetheless, liquid digestates still contains emerging pollutants such as sunscreen octocrylene at 147 ug/L and acetaminophen at 58.6 ug/L. As a result, pretreatment of sludge containing emerging pollutants is required.

Keywords Emerging pollutants · Sewage sludge · Anaerobic digestion · Valorization · Climate change

Introduction

Sewage sludge or biosolids are organic and inorganic compounds produced by the biological activity of microbes in wastewater treatment plants (Tawfik et al. 2012; Xie et al. 2022). The production of biosolids has grown in recent decades as a result of the use of aerobic methods in sewage treatment, such as the activated sludge process (Abdul et al. 2022b; Tawfik et al. 2022a). Anaerobic digestion has been increasingly accepted as a low-cost and sustainable treatment approach for sewage sludge stabilization, in which microorganisms break down the organic matter under anaerobic conditions (Zhao et al. 2021b; Abdul et al. 2022a). Sewage sludge is characterized by high organic matter and nutrient concentrations (Tawfik et al. 2006). Two byproducts of anaerobic digestion are produced: biogas and stabilized sewage sludge or biosolids, considered valuable resources under the circular economy concept (Tawfik et al. 2015).

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Biogas produced from the anaerobic digestion of sewage sludge is considered a promising sustainable clean energy, representing a commercial value as renewable energy (Tawfik et al. 2021b). Likewise, stabilized sewage sludge is high in organics and nutrients that could be reused as biofertilizers in agricultural applications (Rawoof et al. 2021; Kani et al. 2022).

However, the anaerobic digestion process is driven by the high level of emerging pollutants that has lately increased due to increased human activity (Morin-Crini et al. 2022). There is currently no legislation governing the reuse of biosolids for agricultural applications, particularly those containing emerging contaminants (Petrie et al. 2015). Emerging pollutants, including pharmaceuticals, hormones, pesticides, and household and industrial chemicals, pose potential environmental and human health risks (Saravanan et al. 2021; Ahmad et al. 2022). Furthermore, emerging pollutants are known to impede the anaerobic digestion of sewage sludge, resulting in a decrease in biogas productivity (Tawfik and ElBatrawy 2012).

Pharmaceuticals are the most abundant emerging pollutants in wastewater because pharmaceuticals are widely and intensively used by humans and veterinarians in medicine (Tijani et al. 2016). Furthermore, several pharmaceuticals are used to promote the growth of fish and livestock farms (El-Gohary et al. 2010). Pharmaceuticals are easily metabolized into polar and soluble forms in the human body (Nikolaou et al. 2007). Therefore, these pharmaceuticals and their metabolites are mostly discharged into wastewater treatment plants via urine and feces (Bassuney and Tawfik 2017). Other emerging pollutants such as herbicides, insecticides, food additives, fungicides, preservatives, protective coatings, plasticizers, flame retardants, corrosion inhibitors, sunscreen agents, textiles, and microplastics may also enter wastewater treatment plants as a result of the unregulated discharge of these wastes into the sewage system. Some of these compounds could be removed from wastewater treatment plants by chemicals and biodegraded by biological treatment processes (Allam et al. 2016). The removal of emerging pollutants from the aqueous phase occurs, and emerging pollutants are mainly adsorbed onto the biosolids (Jones et al. 2005).

The presence of emerging pollutants in the sludge causes serious problems, notably in agricultural sectors, and these emerging pollutants also have a detrimental impact on the efficacy of the anaerobic digestion process; thus, sludge treatment and removal of emerging pollutants are high priorities (Jelic et al. 2011; Ahmad et al. 2022). The sludge produced by wastewater treatment facilities mostly comprises emerging contaminants that need to be eliminated prior to agriculture uses (Sena et al. 2010). Morales et al. (2016) investigated the fate of emerging pollutants in an anaerobic digester fed with sewage sludge and operated at an organic loading rate of 1.1–1.4 kg/m^3·d and sludge residence time of 20 days. Sweeteners such as acesulfame, pesticides such as thiabendazole, and pharmaceuticals such as venlafaxine, carbamazepine, irbesartan, valsartan, diclofenac were detected in the aqueous and solid phase, while metabolites byproducts were also observed, i.e., salicylic acid, a metabolite of acetylsalicylic acid, and fenofibric acid. Moreover, metabolites of 4-formyl aminoantipyrine, fenofibrate, 4-acetyl aminoantipyrine, 4-aminoantipyrine, and metabolites of dipyrone were highly detected in the solid phase. The adsorption of the emerging pollutants was the removal mechanism due to the presence of the emerging pollutants in the solid phase during the anaerobic digestion process.

Therefore, this review aims to provide a complete overview of sources and types of emerging pollutants that enter wastewater treatment facilities. The contents of such contaminants before and after the anaerobic digestion process and their inhibitory effects and mechanisms are assessed. In addition, the recent trends of mitigation approaches to tackle the inhibition effects of emerging pollutants on the anaerobic digestion of sewage sludge are also thoroughly discussed. We expect our work to contribute to the advancement of value-added byproducts from sludge-containing emerging pollutants and their commercial viability.

Emerging pollutants in wastewater

Wastewater treatment plants are typically designed to remove nitrogen, phosphorus, and chemical oxygen demand from wastewater (Ismail and Tawfik 2016; Ismail et al. 2021). As a result, the presence of emerging pollutants in the influent wastewater may impact the treatment processes and the quality of the treated effluent (Tyagi et al. 2021). The types and concentrations of emerging pollutants that could reach wastewater treatment plants vary according to the source, dose management, and persistent nature of the emerging pollutants (Tawfik et al. 2022b). Figure 1 depicts the path emerging contaminants take from their origins all the way to the environment.

Industrial wastewater

Industrialization has resulted in a significant deterioration in the integrity of the water resources as a result of unmanaged wastewater disposal from industrial operations such as mining, pharmaceuticals, and textile (Ali et al. 2017; Xie et al. 2022). Industrial activities contribute a range of inorganic and organic contaminants to water systems, altering the water quality upon which biological life relies (Azzam and Tawfik 2015). The major pollutants in industrial water include pesticides, herbicides, petroleum, refinery, heavy metals, pharmaceuticals and personal care products due

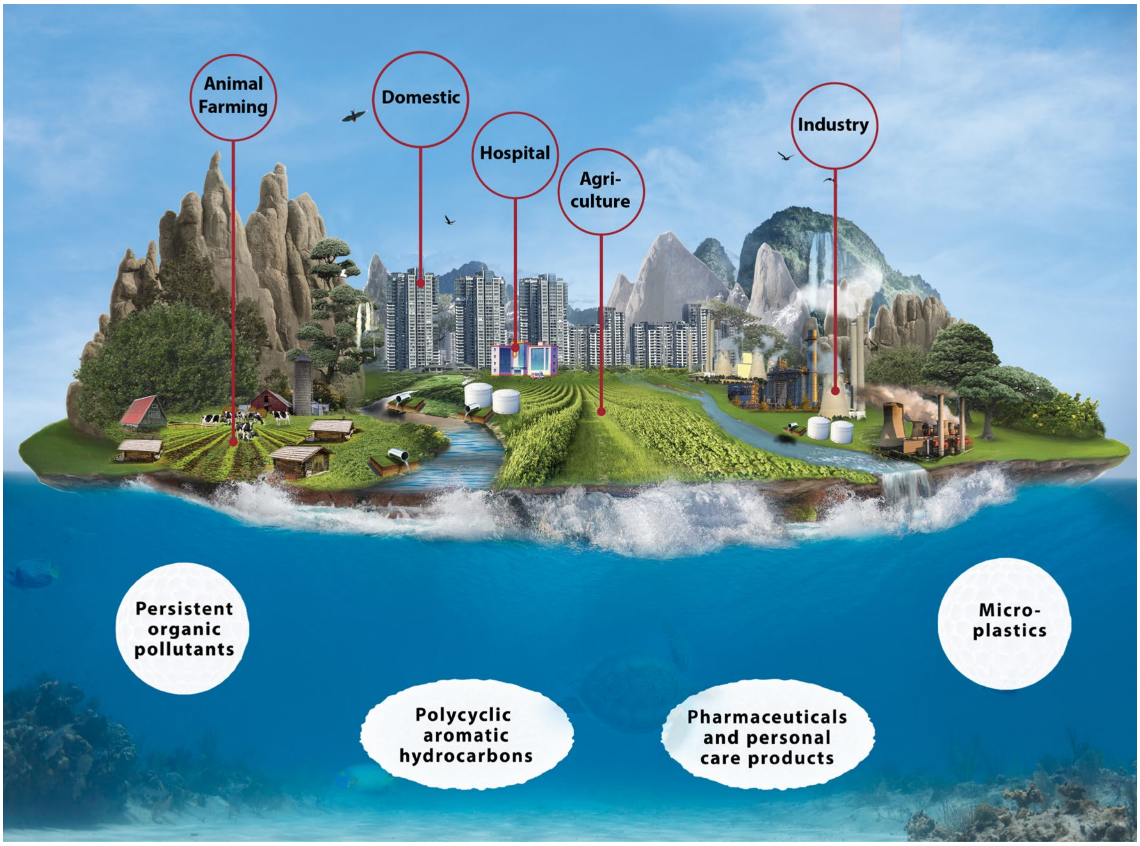

Fig. 1 Fate of emerging pollutants from their sources to the environment. Hospitals, residences, industries, agriculture, and animal farming are potential sources of emerging contaminants. Pharmaceuticals and personal care products, persistent organic pollutants, polycyclic aromatic hydrocarbons, and microplastics are amongst the most significant emerging pollutants. The major portions of emerging pollutants are produced from the industry. The presence of emerging pollutants in the influent wastewater would impact the treatment processes and the quality of the treated effluent

to their toxicity and the presence of refractory compounds (Gar Alalm et al. 2017). Pharmaceuticals and personal care products are chemical substances utilized in personal care, medicinal items, and cosmetics (Gar Alalm et al. 2016). Pharmaceuticals include antibiotics, anticancer, antidiabetics, antiseptics, antimicrobials, antianxiety, anti-inflammatory and anticonvulsant medications, barbiturates, analgesics and lipid regulators (Nikolaou et al. 2007; Okuda et al. 2008; Bulloch et al. 2015; Chen et al. 2016; Archer et al. 2017). The major emerging contaminants that are released from pharmaceuticals are sulfamethazine, carbamazepine, diclofenac, caffeine, clofibric acid diclofenac, ibuprofen, ciprofloxacin, bisphenol A, metronidazole, metalaxyl, dimetridazole, diatrizoate, atenolol, tricyclazole, fludioxonil, carbofuran, bentazon, and iopamidol (Tijani et al. 2013; Zenker et al. 2014; Lin et al. 2015). Personal care industries produce an end-off pipe effluent rich in emerging contaminants, such as triclosan, benzophenones, ethylparaben, and methyl-dihydrojasmonate (Daughton and Ternes 1999; Snyder et al. 2003; Matamoros et al. 2007; Kasprzyk-Hordern et al. 2009).

Power plants generate the most toxic emerging pollutants, such as nitrogen and carbon, oxides of sulfur, formaldehyde, particulate matter, dioxins, furans, and heavy metals, e.g., mercury, copper, cadmium, lead, zinc, and chromium (Demirak et al. 2005; Rahman et al. 2020). Also, petroleum refineries industries cause severe pollution since these industries discharge effluent rich in benzene, acetone, phenol, nitrogen, and petroleum byproducts (Damian 2013). Electroplating factories and printed circuit boards discharge wastewater containing heavy metals, e.g., zinc, magnesium, calcium, potassium, and sodium (Sage and Schnitzer 1994; Gerić et al. 2017; Xiu et al. 2019; Chen et al. 2021). Furthermore, pesticide industry effluent contains high chemical oxygen demand and compounds such as phenols, halomethanes, and volatile aromatics (Bachmann Pinto et al. 2018; Lin et al. 2020), causing the wastewater to be highly contaminated and toxic (Zeyad et al. 2019). For paper mill and pulp industries, various toxic substances are generated, such as diterpene alcohols, resin acids, juvaniones, unsaturated fatty acids and chlorinated resin acids (Pokhrel and Viraraghavan 2004; Kamali and Khodaparast 2015), which has the potential to cause adverse effects to aquatic organisms (Ali and Sreekrishnan 2001).

Moreover, the textile industry generates wastewater high in colorants, salts, pH, temperature, dissolved solids, metals, and chemical oxygen demand (Sharma et al. 2007; Sekomo et al. 2012; Afanga et al. 2020). Additionally, the printing industry effluent containing nickel, lead, copper, and chromium as major contaminates lead to their accumulation in aquatic organisms (Kiurski et al. 2012; Papadopoulos et al. 2019; Safwat 2020). Both dyeing and textile printing wastewater contain a high amount of microfibers (Xu et al. 2018; Zhou et al. 2020b), defined as particles less than 5 mm in length. Such microfibers can be ingested by aquatic organisms and cause adverse biological effects (Mohsen et al. 2020; Rebelein et al. 2021). Microfibers are a type of microplastics that have been extensively detected in industrial wastewater. Microplastic abundance in industrial and municipal wastewater treatment plants ranged from 16,000 to 31,400 particles (Liu et al. 2021a). Table 1 summarizes the emerging pollutants contained in industrial effluents.

Domestic wastewater

Domestic wastewater is released from human activities in households, industries, offices, institutions, and premises (Xu et al. 2022). This sewage contains organic and inorganic pollutants in a complex mixture composed primarily of around 99% water (Tawfik et al. 2008). Domestic wastewater is mainly the source of emerging pollutants, where most of the pharmaceuticals and personal care products are consumed by humans and reach the environment via urinating and defecation processes (Ismail and Tawfik 2017). According to metropolitan Melbourne's water utilities, the major contaminants in sewage include color, total dissolved solids, mercury, cadmium, copper, arsenic, nickel, zinc, lead, and boron. Also, caffeine, acetaminophen, and paraxanthine are the most emerging contaminants in sewage (Rosal et al. 2010; Sophia and Lima 2018). Furthermore, naproxen, diclofenac, and ketoprofen were highly detected in sewage at concentrations of 4.2–7.2, 0.4–1.5, and 1.1–2.3 mg/L, respectively (Jelic et al. 2011).

Moreover, lipid modifying agents were detected at considerable amounts in the influent wastewater (i.e., 7–12% for fibrates, 8–10% for statins, and 5–9% for diuretics). The amounts of furosemide, atenolol, carbamazepine, and bezafibrate in sewage ranged from 0.4 to 1.4 mg/L. There were 35–44% non-steroidal anti-inflammatory drugs, 8–29% lipid-modifying compounds, and 17–30% psychiatric pharmaceuticals, e.g., benzodiazepine and antiepileptic derivative drugs, in the influent wastewater treatment facility in Spain (Jelic et al. 2011). The highest quantities of naproxen, carbamazepine, and diclofenac were found in wastewater at 0.4–1.0 mg/L (Table 2).

The discovery of microplastics as pervasive contaminants in the environment has sparked worldwide concern and led to extensive research on this topic. Numerous products in personal care and cosmetics include microplastics in their ingredients, such as toothpaste, scrubbers, and facial cleanser. For instance, polyethylene microbeads are applied in facial cleansers. An exfoliant might emit between 4,594 and 94,500 microbeads with a single use (Napper et al. 2015; Anderson et al. 2016; Hu et al. 2019). Urban areas generate a large amount of microplastics into wastewater treatment plants, which enter the environment due to the huge quantities of microplastics discharged from wastewater treatment plants {Formatting Citation}(Talvitie et al. 2017). Recent research estimated that a single and medium-sized wastewater treatment plant with a flow rate of 30–50,000 m^3/day might emit up to $1.83*10^{10}$ microplastics/day (Leslie et al. 2017; Ben-David et al. 2021). Table 2 lists the most significant emerging pollutants found in the household or domestic wastewater.

Animal farming waste

Animal waste dumping in water pollutes the receiving water bodies and contributes to the spread of water-borne diseases, resulting in a scarcity of safe drinking water (Tawfik et al. 2021a). For instance, the pollution of water by cattle excrement caused several deaths in the Canadian community of Walkerton (Singh and Rashed 2017). The runoff of animal wastes is a major potential source of decomposable organic matter, nutrients, antibiotics, and hormones (Al Salah et al. 2019). Antibiotics, which are used to prevent or cure animal illnesses, and steroid hormones, which are used to fatten farmed animals, are considered emerging pollutants (Senarathna et al. 2021). Furthermore, the excessive quantity of animal waste fertilizing could leach high nitrogen content and runoff into the surface and groundwater, contaminating both animal and human drinking water sources with excessive concentrations of nitrates (Sahoo et al. 2016). As shown in Table 3, the principal components of emerging pollutants in animal farming wastewater are hormones and antibiotics. Additionally, Fig. 2 illustrates the sources of emerging pollutants in the environment and their associated kinds. In summary, the separation of animal farming waste for treatment prior to reuse in agriculture is needed to avoid the accumulation of hormones and antibiotics.

Removal of emerging contaminants in wastewater treatment plants

Emergent contaminants, such as dissolved organics, colloidal and suspended particulates, pathogens, and nutrients, are removed with 20–50% efficiency during primary treatment. Those contaminants are further removed by 30–70%

Table 1 Majority of emerging pollutants presented in industrial wastewater. Pesticides and herbicides are the most emerging contaminants. The insecticides are the lowest. Food additives, fungicides, preservatives, protective coatings, plasticizers, flame retardants, corrosion inhibitors, sunscreen agents, and textile are detected in the end-off pipe effluent. Microfibers are presented in textile wastewater

Industry	Emerging contaminants	References
Pesticides	2,4-dichloroaniline	Birch et al. (2015), Thompson et al. (2021)
	3,4 dichloroaniline	
	Aldrin	Hirooka et al. (2006), Buttiglieri and Knepper (2008)
	Atrazine	
	Bentazone	
	Carbaryl	Häggblom et al. (2003), Birch et al. (2015)
	Chlorpyrifos-methyl	Baun et al. (2004), Smital et al. (2004)
	Desethylatrazine	Buttiglieri and Knepper (2008)
	Diazinon	Smital et al. (2004)
	Dichlobenil	Buttiglieri and Knepper (2008)
	Dichlorvos	Smital et al. (2004)
	Dieldrin	Buttiglieri and Knepper (2008)
	Dimethoate	Smital et al. (2004)
	Diuron	Birch et al. (2015)
	Endosulfan	Smital et al. (2004)
	Endrin	Buttiglieri and Knepper (2008)
	Fenoxycarb	Smital et al. (2004)
	Glifosphate	
	Heptachlor	Buttiglieri and Knepper (2008), Vilar et al. (2012)
	Hexachlorobenzene	
	Hexachlorobutadine	
	Isobenzan	
	Isodrin	
	Isoproturon	
	Malathion	Smital et al. (2004), Gar Alalm et al. (2015)
	Malic hidrazid	
	2-methyl-4-chlorophenoxyacetic acid	Birch et al. (2015)
	Metamitron	Smital et al. (2004)
	Methomil	
	Phosalone	
	Pirimicarb	
	Propiconasole	
	Quintozene	Buttiglieri and Knepper (2008)
	Simazine	
	Simazine	Birch et al. (2015)
	Terbutylazine	Buttiglieri and Knepper (2008)
	α-endosulfan	

Table 1 (continued)

Industry	Emerging contaminants	References
Herbicides	2,4-dinitrophenol	John et al. (1982), Hirooka et al. (2006), Birch et al. (2015), Palatucci et al. (2019)
	2,3- and 3,4-dichloronitrobenzene	
	3,4-dichloro aniline	
	Ametryn	
	Asulam	
	Atrazine	
	Bromacil	
	Bromoxynil	
	Chlorpyriphos	
	Dalapon	
	Dicamba	
	Desethyl atrazine	
	Desisopropyl atrazine	
	Diuron	
	Flumeturon	
	Fluroxypyr	
Insecticide	Aldrin	Singh and Walker (2006), Daneshvar et al. (2007), Navarro et al. (2009), Murray et al. (2010)
	Dichlorodiphenyltrichloroethane	
	Dieldrin	
	Endrin	
	Lindane	
Food additive	Anti-oxidants	
Fungicide	Benomyl	
	Carbendazim	
Preservatives	Butylparaben	Kasprzyk-Hordern et al. (2009), Sophia and Lima (2018)
	Propylparaben	
Protective coatings	Perfluorate	Murray et al. (2010), Stasinakis (2012), Mailler et al. (2017)
Plasticizer	Phthalates	
Flame retardant	Polybrominated diphenylether	
Corrosion inhibitors	Triazole	
Sunscreen agents	Benzophenone-1	Kasprzyk-Hordern et al. (2009), Badia-Fabregat et al. (2012)
	Benzophenone-3	
	Benzophenone-2	
	Benzophenone-4	
Textile	Microfibers	Xu et al. (2018), Zhou et al. (2020b)

in the secondary treatment process (Rout et al. 2021). Jelic et al. (2011) analyzed pharmaceuticals in 43 sludge samples that resulted from conventional wastewater treatment plants. Their findings revealed that 50% of the sludge samples had significant levels of pharmaceuticals. 19% of diuretics and 16–21% of lipid-modifying agents were detected in the sludge. Hydrochlorothiazide, diclofenac, furosemide, clarithromycin, atorvastatin, and carbamazepine were detected in the sludge samples of the wastewater treatments at the levels of 30–60 ug/g total solids. In sludge samples, low concentrations of beta-blockers, histamine H_2-receptor antagonists, and beta-agonists were found. Otherwise, the sludge from the sewage treatment plant included high concentrations of pharmaceuticals (Castiglioni et al. 2006; Zorita et al. 2009).

Removal of about 80% of naproxen, ketoprofen, and antihypertensive enalapril was achieved with no accumulation in the sludge (Lishman et al. 2006; Sim et al.

Table 2 Emerging pollutants found in domestic wastewater. Endocrine disruptors and surfactants are the most contaminants. Pharmaceuticals are abundant. Naproxen, diclofenac, and ketoprofen are highly detected in sewage. Caffeine, acetaminophen, and paraxanthine are the most pharmaceuticals in wastewater

Sources	Emerging contaminants	References
Endocrine disruptors	4-tert-octylphenol	Kasprzyk-Hordern et al. (2009)
	Bisphenol A	
Pharmaceuticals—analgesics and anti-inflammatories	Acetaminophen	Rivera-Utrilla et al. (2013), Pietrini et al. (2015)
	Ibuprofen	
Pharmaceuticals—b-blockers	Atenolol	
Pharmaceuticals—antibiotics	Azithromycin	
	Metronidazole	
	Trimethoprim	
Pharmaceuticals	Benzafibrate	Petrović et al. (2003), Larsen et al. (2019)
	Carbamazepine	
	Clofibric acid	
	Diclofenac	
	Gemfibrozil	
	Microplastics	Leslie et al. (2017), Ben-David et al. (2021)
Pharmaceuticals—antiepileptics	Carbamazepine	Rivera-Utrilla et al. (2013), Harris and Logan (2014)
	Ibuprofen	Petrović et al. (2003), Gottschall et al. (2012)
	Ketoprofen	
	Naproxen	
	Triclosan	
Pharmaceuticals—lipid-lowering drugs	Bezafibrate	Rivera-Utrilla et al. (2013), Ding et al. (2017)
	Clofibrate	
	Gemfibrozil	
Pharmaceuticals—analgesics and anti-inflammatories	Diclofenac	
	Ketoprofen	
	Naproxen	
Surfactant	Phenol	Murray et al. (2010), Okada et al. (2013)
Pharmaceuticals—antacids	Ranitidine	Rivera-Utrilla et al. (2013), Reddy et al. (2021)
Pharmaceuticals—b-blockers	Sotalol	

2010). Also, anticonvulsant carbamazepine was removed only by lower than 25% in conventional wastewater treatment plants (Joss et al. 2005; Pérez and Barceló 2007; Radjenović et al. 2009). Additionally, partial removal of lower than 30% of antibiotics, including trimethoprim and benzodiazepine, lorazepam and metronidazole, was taken place by aerobic bacteria (Bendz et al. 2005; Göbel et al. 2007; Kasprzyk-Hordern et al. 2009). On the other hand, no removal of salbutamol, bezafibrate, and furosemide was observed during the aerobic treatment process (Castiglioni et al. 2006). However, over 85% of 55 different pharmaceuticals and personal care products were removed by an activated sludge treatment plant (Kasprzyk-Hordern et al. 2009). Furthermore, histamine H_2-receptor antagonist removal efficiency was quite low in the wastewater treatment plant (Radjenović et al. 2009), but the removal performance can reach as high as 86% (Kasprzyk-Hordern et al. 2009; Petrie et al. 2015). Moreover, the activated sludge plant removed diclofenac up to 24–60% (Kimura et al. 2007; Cirja et al. 2008).

Microplastic removal from wastewater is a critical contribution. Microplastics are removed via the primary treatment stage, with an average removal efficiency of 72–93% (Ateia et al. 2022). Also, the removal of biodegradable microplastics during the secondary treatment stage occurs via microorganisms. Furthermore, numerous technologies have been proposed for microplastic removals, such as ozonation and membrane bioreactor (Bui et al. 2020).

In conclusion, the primary treatment of emergent pollutants is necessary before discharge into wastewater treatment plants. The pretreatment process will undoubtedly improve the efficiency of existing wastewater treatment plants.

Table 3 Animal farming wastewater is rich with emerging pollutants. Dumping the runoff of animal wastes into the environment causes severe pollution. The leachate of animal waste after utilization in agriculture contaminates the groundwater. The animal farming waste is rich in nitrogen species. Antibiotics and hormones are the major contaminants in animal farming wastewater

Source	Emerging contaminants	References
Hormone	4-androstene-3,17-dione	Blair et al. (2013), Sami and Fatma (2019), Wang et al. (2020)
	17-alpha-estradiol	
	17-beta-estradiol	
	17,20-dihydroxyprogesterone	
	Estriol	
	Estrone	
	Lincomycin	
	Progesterone	
	Testosterone	
Antibiotic	Azithromycin	Blair et al. (2013), Zhang et al. (2019), Leng et al. (2020)
	Ciprofloxacin	
	Clarithromycin	
	Lomefloxacin	
	Miconazole	
	Norfloxacin	
	Ofloxacin	
	Sarafloxacin	
	Sulfachloropyridazine	
	Sulfadiazine	
	Trimethoprim	

Emerging contaminants in sludges

The recovery of sludge and biosolids from wastewater treatment plants has recently received great attention for the circular economy (Meky et al. 2019). Biosolids are mainly produced due to biological activities or chemical processes (Tawfik and Elsamadony 2017). The biosolids' characteristics have various chemical, biological and physical compositions. The recovery and re-utilization of biosolids are feasible; however, the presence of emerging contaminants in the municipal, hazardous sludge from industry, and biosolids from treating industrial wastewater makes a big barrier to the re-utilization of such natural resources. In 2001, the United States Environmental Protection Agency identified 38 out of 72 emerging pollutants in 110 biosolid samples at mg/kg level in a countrywide survey and reported environmental contaminants with concentrations up to several hundred parts per million in 74 wastewater treatment plants. Additionally, by 2006, sewage sludge worldwide had been shown to include 516 different chemicals belonging to 15 different emerging contaminants classes (Dubey et al. 2021). Benedetti et al. (2020) found that azithromycin, antibiotics, ciprofloxacin, and estrone were abundant at about 500–600 ng/g in the sludge generated from wastewater treatment plants.

Polycyclic aromatic hydrocarbons of 24 compounds are detected and identified in Table 2. This list of emerging pollutants includes 16 polycyclic aromatic hydrocarbons. For the petroleum fuel and oil industry, 2,6-dimethyl naphthalene, 2-methyl naphthalene, and 2,3,6-trimethyl naphthalene were the majority abundant chemicals. Additionally, lighter polycyclic aromatic hydrocarbon compounds such as fluorene and phenanthrene were abundant. Those contaminants

Fig. 2 Main discharge route of emerging contaminants. Industrial wastewater should be treated prior to reaching the wastewater treatment plant. Hospital sewage, agricultural effluent, domestic wastewater, and animal farming wastewater are the main sources of emerging contaminants in the environment. Treatment plants receive wastewater-rich emerging contaminants. The effluent quality of wastewater treatment plants depends on the loading rate of the emerging contaminants

are rapidly biodegraded and/or volatilized from the soil, of which 2,6-dimethyl naphthalene and 2-methyl naphthalene are particularly 20–30% dominant. The summation of the polycyclic aromatic hydrocarbons including benzo[bjk] fluoranthene, acenaphthene, fluoranthene, phenanthrene, fluorene, pyrene, benzo[ghi]perylene, indeno[1,2,3-cd]pyrene and benzo[a]pyrene should not exceed 6 mg/kg dry matter, according to European Union regulations.

Table 4 shows that total polycyclic aromatic hydrocarbons concentrations varied from 18 to 50 mg/kg dry weight in the sludge, complying with the standards (Moreda et al. 1998; Manoli and Samara 1999; Duan et al. 2021). Polychlorinated biphenyl congeners of 46 were detected in the sludge samples. Fortunately, the congeners of 54, 104, 114, 105, 155, and 188 were not quantified. Total polychlorinated biphenyl concentration was 220 ug/kg dry weight. Polychlorinated naphthalenes had 35 congeners, 17 of which were found in the sludge samples, and their mean concentration was 83 ug/kg dry weight.

Anaerobic digestion of sludges containing emerging contaminants

Food waste is almost free from emerging contaminants; however, biogas productivity from anaerobic digestion of food waste as a standalone substrate is quite low (Chew et al. 2021; Liu et al. 2021b). The bioenergy productivity is highly increased by anaerobic digestion of food waste with co-substrate, i.e., agricultural, biosolids or sludge, and manures. The latter contains emerging contaminants that make a barrier to the reuse of the digestate that resulted from the anaerobic co-digestion process (Zhang et al. 2021). Acidogenic fermentation of the aluminum-sludge and iron-sludge resulted from chemically enhanced primary sedimentation of sewage provided removal efficiencies of 50% and 58% of retinoids, and 50% and 47% of endocrine-disrupting chemicals (4-nonylphenol, estrone, diethylstilbestrol, triclosan, triclocarban, and bisphenol A), respectively, in bulk liquid (Zhou et al. 2020a). However, the concentrations of retinoids and endocrine-disrupting chemicals increased after fermentation in the solid phase, indicating that these chemicals were adsorbed in the sludge. After the acidogenic fermentation process, the retinoid 13-cis-RA concentrations in aluminum-sludge and iron-sludge increased from 19 to 140 ng/g dry weight and 25 to 97 ng/g dry weight, respectively.

This was not the case for endocrine-disrupting chemicals, which slightly increased in the sludge after acidogenic fermentation. The anaerobic microbial community greatly contributed to removing antibiotic resistance genes from sludge (Jang et al. 2017). The presence of endocrine-disrupting chemicals, such as 4-nonylphenol and triclocarban, improved the activities and diversities of the microbial community and subsequently enhanced the solubilization and acidogenesis process of sludge (Duan et al. 2016; Wang et al. 2017). However, 4-nonylphenol in the activated sludge could be biodegraded by acidogenic microorganisms (Duan et al. 2016, 2018). On the contrary, the microbial activities were quite low for removing antibacterial agents, i.e., triclocarban and triclosan (Wang et al. 2017; Yan et al. 2019a). A high concentration of both triclocarban and triclosan was found in the fermented sludge.

Likely, Wang et al. (2017) found that triclocarban was slightly removed from sludge during the anaerobic digestion of activated sludge. This indicates that the risks of triclocarban and triclosan in the fermented sludge are still causing severe pollution. Triclocarban and triclosan should be removed from fermented sludge before disposal. Triclocarban and triclosan were mainly accumulated in the sludge during the anaerobic digestion process due to the sorption of solid materials (Zhou et al. 2019). The fermentation of the sludge by acidogenesis only minimized the levels of retinoids by 3.3% and for endocrine-disrupting chemicals by 1.4% in the supernatant, confirming a low contribution of anaerobic digestion to remove emerging contaminants from sludge (Zhou et al. 2020a).

Lin et al. (2017) reported that the sludge fermentation with acidogenic bacteria could successfully lower the level of emerging pollutants in the supernatant, which is suitable for resource recovery. Additional research is needed to optimize the removal of endocrine-disrupting chemicals from the solid sludge phase to reduce their health hazard and environmental impact. The supernatant (liquid) digestate that resulted from anaerobic digestion of sludge containing emerging pollutants had the highest concentration of 147 ug/L for sunscreen octocrylene and 58.6 ug/L for acetaminophen (Ali et al. 2019).

However, lower fractions of octocrylene (more than 600 ng/g wet weight) and tris-1-chloro-2-propyl-phosphate (more than 500 ng/g wet weight) were observed in solid digestates. The total contaminants of emerging concern were 1411 ng/g in solid digestates and 354 ng/g in liquid digestates. The behavior of irbesartan, benzoylecgonine, and venlafaxine in the sludge after mesophilic and thermophilic anaerobic digestion was reported by Morales et al. (2016). Apparently, benzoylecgonine and irbesartan in the sludge were probably biodegraded by fermentation. Venlafaxine concentrations were substantially lower in the aqueous stage and significantly higher in the solid stage. Irbesartan, benzoylecgonine, and venlafaxine concentrations in the solid stage were significantly greater than in the aqueous stage, indicating that irbesartan, benzoylecgonine, and venlafaxine were substantially adsorbed onto the biosolids. The emerging pollutants found in the treated sludge are listed in Table 5.

Table 4 Emerging contaminants detected in sewage sludges (Stevens et al. (2003; Mailler et al. 2017). The emerging contaminants include polycyclic aromatic hydrocarbons. The polychlorinated biphenyls and polychlorinated naphthalene are detected in sewage sludge. Azithromycin, antibiotics, ciprofloxacin, and estrone are abundant in the sludge generated from wastewater treatment plants. The petroleum fuel and oil industry contain 2,6-dimethyl naphthalene, 2-methyl naphthalene, and 2,3,6-trimethyl naphthalene, nd: non-detected

Polycyclic aromatic hydrocarbons (mg/kg dry weight)	Min	Max	Mean	Median
Naphthalene	0.15	19	3.7	1.4
2-methylnaphthalene	5.9	93	24	13
1-methylnaphthalene	2.4	39	9.9	5.0
Biphenyl	1.7	28	6.3	4.0
2,6-dimethylnaphthalene	5.0	110	30	18
Acenaphthylene	0.030	0.10	0.060	0.050
Acenaphthene	1.7	6.6	4.0	3.9
2,3,6-trimethylnaphthalene	0.96	15	6.9	5.7
Fluorene	3.6	8.1	5.7	5.7
Phenanthrene	1.4	7.4	4.9	5.4
Anthracene	0.38	1.8	0.72	0.65
1-methylphenanthrene	0.46	8.1	3.9	3.5
Fluoranthene	1.4	7.4	4.9	5.4
Pyrene	2.1	5.6	4.2	4.5
Benz[a]anthracene	0.6	2.8	1.8	1.8
Chrysene	1.0	6.0	2.6	2.3
Benzo[b]fluoranthene	1.1	7.2	3.0	2.9
Benzo[jk]fluoranthene	0.7	4.5	2.2	1.9
Benzo[e]pyrene	0.82	4.4	2.2	2.0
Benzo[a]pyrene	0.69	4.0	2.1	2.1
Perylene	0.12	0.61	0.36	0.35
Indeno[1,2,3-cd]pyrene	0.39	2.7	1.3	1.1
Dibenz[ah]anthracene	0.060	0.38	0.19	0.19
Benzo[ghi]perylene	0.47	2.3	1.3	1.1
Total	67	370	130	93

Polychlorinated biphenyls concentrations (ug/kg dry weight)	Min	Max	Mean	Median
18	1.5	14	5.7	5.0
22	1.7	43	9.3	6.0
28	5.1	26	12	11
31	3.5	56	13	8.1
44	1.0	6.5	3.1	2.8
41/64	1.3	7.3	3.4	3.1
49	1.7	13	4.6	3.8
52	3.1	28	12	8.7
60/56	0.4	4.8	1.8	1.9
70	2.7	33	8.3	6.1
74	1.7	8.7	3.5	3.0
87	0.9	5.3	2.6	2.1
90/101	3.8	74	13	8.2
95	2.3	22	6.4	4.4
99	1.1	4.9	2.6	2.1
110	1.5	10	4.6	4.0
118	1.6	20	6.1	5.2
123	0.3	8.4	4.0	3.4
132	10	39	20	19
138	6.9	23	13	12

Table 4 (continued)

Polychlorinated biphenyls concentrations (ug/kg dry weight)	Min	Max	Mean	Median
141	1.3	5.7	2.8	2.3
149	5.7	20	11	8.9
151	2.1	7.6	3.8	2.9
153	7.3	27	14	13
156	0.5	2.1	1.1	0.97
157	0.1	0.49	0.31	0.29
158	0.2	2.3	1.2	1.0
167	0.2	1.1	0.49	0.40
170	1.3	8.6	3.3	2.3
174	1.6	9.7	3.9	2.9
180	4.7	23	10	8.5
183	1.2	5.7	2.6	2.1
187	2.6	12	5.8	4.8
189	0.010	0.35	0.17	0.17
194	0.1	7.5	2.6	2.0
199	0.090	1.3	0.35	0.26
203	1.4	11	3.1	2.5
Total	110	440	220	190
Polychlorinated naphthalene (ug/kg dry weight)	Min	Max	Mean	Median
19	nd	1.8	0.50	0.20
23	nd	20	10	9.7
15	12	78	27	23
16	13	97	31	26
42	0.3	0.8	0.5	0.5
Polychlorinated naphthalene 4–11	nd	0.4	0.2	0.2
38 (40)	1.5	3.9	2.4	2.2
46	nd	1.5	0.9	0.9
33/34/37	1.9	4.4	3.0	2.9
47	0.6	3.2	1.1	0.9
36/35	0.2	1.1	0.6	0.6
52/60	nd	0.9	0.3	0.3
59	nd	1.9	0.4	nd
Total	50	190	83	76
Synthetic musks	Min	Max	Mean	Median
Celestolide	0.010	0.26	0.071	0.035
Phantolide	0.032	1.1	0.41	0.39
Traseolide	0.044	1.1	0.45	0.45
Galaxolide	1.9	81	27	26
Tonalide	0.12	16	4.7	4.0
Total	2.1	99	32	31
Polychlorinated (chain length)	Min	Max	Mean	Median
ΣC10	0.99	21	3.8	1.80
ΣC11	1.6	60	12	4.7
ΣC12	2.5	62	14	5.7
ΣC13	1.8	69	13	4.2
Total short-chain	6.9	200	42	16
ΣC14	19	6000	1000	290

Table 4 (continued)

Polychlorinated (chain length)	Min	Max	Mean	Median
Σ C15	7.3	2500	490	150
ΣC16	3.0	1100	210	56
ΣC17	0.92	310	50	18
Total medium-chain	30	9700	1800	540
Total short and medium-chain	45	9900	1800	560

Inhibition effects of the emerging pollutants on anaerobic digestion of sludges

Methanogenesis and methane productivity

Fluoxetine inhibited the anaerobic digestion of excess sludge (Zhao et al. 2021a) at concentrations exceeding 2.0 mg/kg. However, methane productivity was unaffected by a fluoxetine dose of 0.1 mg/kg. Nevertheless, a fluoxetine dose of 2.0 mg/kg resulted in a 91.2 ± 4.3 mL/g reduction in methane productivity of volatile suspended solids. This reduction in methane productivity was equivalent to 59.9 ± 3.4% of the control. The fluoxetine declined hydrolysis process, acidification, and methanogenesis activities due to inhibiting enzyme activities.

Sludge reduction and solubilization

Volatile solids reduction mainly occurs due to the hydrolysis and anaerobic metabolism activities where a portion of organic matter is solubilized during sludge fermentation. However, the presence of emerging pollutants in the sludge would affect volatile solids reduction (Zhang et al. 2012; Fang et al. 2020). The volatile solids reduction of sludge free emerging pollutants was optimized at a level of 26.9 ± 1.1% during anaerobic digestion (Zhao et al. 2021a), which was reduced to 26.5 ± 1.1, 20.3 ± 0.9, and 16.9 ± 0.8% for sludge containing fluoxetine of 0.1, 0.5, and 2.0 mg/kg, respectively. This indicates that the presence of fluoxetine in the sludge reduced the hydrolysis and conversion of organics by anaerobes (Zhao et al. 2017). Fluoxetine strongly destroys the metabolism activities of anaerobes, resulting in a low volatile solids reduction in the digested sludge. Brooks et al. (2003) reported a significant inhibition of fluoxetine on the anaerobic digestion of sludge, which inhibited bacterial growth and cell malformation.

Hydrolysis, acidification and methanogenesis process

Hydrolysis of sludge under anaerobic digestion is the rate-limiting step due to the limited secretion of hydrolytic enzymes in the presence of emerging pollutants. The acidification process is also negatively affected due to the toxicity inhibition of emerging pollutants (Zhao et al. 2017). The organics hydrolysis increased the solubilization of coarse particles, increasing soluble chemical oxygen demand in the fermentation medium. However, the presence of fluoxetine would inhibit the solubilization process and, subsequently, the acidification activities. Zhao et al. (2021a, b) found that the highest solubilization was 695 ± 12.3 mg soluble chemical oxygen demand/L at a fluoxetine concentration of 0.1 mg/kg and decreased to 659 ± 20.6 mg/L at a fluoxetine dose of 2.0 mg/kg. The authors also found that fluoxetine in the sludge inhibited the solubilization of proteins and polysaccharides of the sludge under anaerobic conditions, which negatively affects methane productivity.

Volatile fatty acid productivity and accumulation

Acidification of sludge by acidogenesis produces volatile fatty acids and hydrogen gas. This biological activity resulted in a drop in pH value and alkalinity. Moreover, the anaerobes are suffered from buffering capacity (Li et al. 2019). The volatile fatty acid variations during sludge fermentation are highly affected by emerging pollutants such as fluoxetine. The volatile fatty acids production of 456 ± 26 mg/L was quite high for fermentation of sludge free emerging pollutants, which was dropped to 425 ± 16 mg/L, 406 ± 21 mg/L, and 358 ± 19 mg/L for sludge containing fluoxetine of 0.1, 0.5, and 1.0 mg fluoxetine/kg, respectively, and volatile fatty acids concentration remained at the same level at a higher dose of 2.0 mg fluoxetine/kg. The inhibition of volatile fatty acids productivity occurred at concentrations exceeding 1.0 mg fluoxetine/kg.

Enzyme activities

Anaerobic degradation of biosolids is mainly taken place by the presence of enzymes secreted by the microorganisms. However, emerging pollutants would negatively affect enzyme activities and, subsequently, bioenergy productivity. Proteins and polysaccharides hydrolysis by protease and cellulase enzymes occur under anaerobic conditions. α-glucosidase, acetate kinase, coenzyme F420, and butyrate kinase are responsible for methane productivity (Li et al. 2016; Zhao et al. 2017; Zhu et al. 2019). The presence of fluoxetine in the sludge inhibited the enzyme activities of

Table 5 Micropollutants in treated sludges. The concentrations of various types of micropollutants discovered in several types of sludge are quantified. The total contaminants of emerging concern are 1411 ng/g in solid digestates and 354 ng/g in liquid digestates. The micropollutants type in the sludge depends on the treatment process. Adsorption is followed by biodegradation which is the main removal mechanism of micropollutants

Micropollutants	Type of sludge	Mean (mg/kg dry matter)	Range (mg/kg dry matter)	References
Organotins				
Triphenyltin	Various	0.63	Less than 0.02–9	Clarke and Smith (2011)
Tributyltin	Various	0.86	0.02–6.0	Clarke and Smith (2011)
	Digested	0.004		Olofsson et al. (2012)
	Digested	1.1±0.4		Fent (1996)
Dibutyltin	Various	1.28	0.41–7.5	Clarke and Smith (2011)
	Digested	1.5±0.5		Fent (1996)
Monobutyltin		0.93	0.1–6.0	Clarke and Smith (2011)
	Digested	0.074		Olofsson et al. (2012)
	Digested	0.5±0.2		Fent (1996)
Phthalates				
Di2-(ethylhexyl)phthalate	Digested	126	91–179	Marttinen et al. (2003)
Di2-(ethylhexyl)phthalate	Various	58	Less than 0.02–3,514	Clarke and Smith (2011)
	Digested	159	13–345	Aparicio et al. (2009)
	Thermally dried	148.8	1.5–3,514	Abad et al. (2005)
Σ8 Polybrominated diphenyl ethers		1.360	0.005–4.690	Clarke and Smith (2011)
Polycyclic aromatic hydrocarbons				
Σ11 Polycyclic aromatic hydrocarbons	Thermally dried	1.89	0.13–7.35	Abad et al. (2005)
Polychlorinated biphenyls				
Σ7 Polychlorinated biphenyls	Thermally dried	0.041	Less than 0.006–0.131	Abad et al. (2005
Alkylphenols				
Nonylphenol	Digested	0.17	Less than 0.04–0.45	Stasinakis et al. (2008), Stasinakis (2012)
	Digested	102.1	Less than 0.19–358.2	González et al. (2010)
	Thermally dried	61.7	16.5–124.9	Stasinakis et al. (2008), Bergé et al. (2012)
	Various	128	0.02–2,530	Bergé et al. (2012)
Nonylphenol mono-ethoxylate	Digested	12.3	1.01–41.3	Stasinakis et al. (2008)
	Digested	53.2	Less than 0.75–287.8	González et al. (2010)
	Various	40.2	0.15–850	Bergé et al. (2012)
Various				
Tributylphosphate	Various		Less than 0.020–2.400	
	Digested	0.011		Olofsson et al. (2012)
Polybrominated diphenyl ethers				
Brominated diphenyl ether 209	Various	0.120	0.006–1.000	Law et al. (2006)
	Digested	0.443	0.133–1.339	Knoth et al. (2007)
	Various	1.039	0.003–18.632	Clarke and Smith (2011)
Σ6 Polybrominated diphenyl ethers	Various	0.250	0.024–1.260	Law et al. (2006))
Σ8 Polybrominated diphenyl ethers	Digested	0.577	0.186–1.627	Knoth et al. (2007
Polycyclic aromatic hydrocarbons	Various	1.360	0.005–4.690	Clarke and Smith (2011)
Σ6 Polycyclic aromatic hydrocarbons	Digested	14.8	4.75–28.1	Stevens et al. (2003), Stevens-Garmon et al. (2011)
	Dewatered	1.68	0.52–3.36	Blanchard et al. (2004)
Σ11 Polycyclic aromatic hydrocarbons	Thermally dried	1.89	0.13–7.35	Abad et al. (2005)
Polychlorinated biphenyls				
Σ7 Polychlorinated biphenyls	Digested	0.080	0.033–0.221	Stevens-Garmon et al. (2011)
	Thermally dried	0.041	Less than 0.006–0.131	Abad et al. (2005)
	Dewatered	0.617	0.12–1.93	Blanchard et al. (2004)

α-glucosidase, acetate kinase, coenzyme F420, and butyrate kinase at concentrations exceeding 0.5 mg/kg and, subsequently, affected the methane productivity (Zhao et al. 2021a). Fluoxetine significantly inhibited cytochrome P450 activity in the liver of fish (Laville et al. 2004; Zhang et al. 2014). Zhang et al. (2013) found that fluoxetine caused inhibition pathways to convert the enzyme of P-glycoprotein and glutathione and suppress the microorganism's metabolic activity, thereby reducing the methanogenesis process.

Mitigation of the inhibition effect of emerging pollutants on the anaerobic digestion of sludges

Sludge solubilization could be highly achieved by the pretreatment process (Eq. 1), which might rupture the microbial cells, solubilize the coarse suspended solids, and increase the soluble chemical oxygen demand, resulting in a high degradation efficiency of refractory compounds, mineralization of organics and biodegradability, thereby improving the methanogenesis process in the presence of the emerging pollutants in the sludge (Mohapatra et al. 2011, 2012; Samaras et al. 2014). Pretreatment of the sludge reduces the required hydrolysis time (Carballa et al. 2007). The pretreatment process will increase the solubility of emerging pollutants in the sludge and subsequently enhance the biodegradation and biomethanization process (Zhang et al. 2021). Sonication and ozonation were effective for sludge solubilization, resulting in a high removal of emerging pollutants from sludge (Mohapatra et al. 2012). Different pretreatment techniques were reported to mitigate anaerobic digestion inhibition by emerging pollutants.

$$\text{Sludge} \overset{\text{pretreatment}}{\rightarrow} \text{Solubilizedsludge} \quad (1)$$

Electro-oxidation

Electrochemical conversion and combustion of organics, such as phenol and hormones, occur due to the generation of free radicals, particularly hydroxyl radicals (▪OH), by which a constant direct current is applied (Rivera-Utrilla et al. 2013). A boron-doped diamond electrode was reported to be the most active anode for the oxidation and mineralization of emerging pollutants (Chen 2004). The electro-oxidation using boron-doped diamond electrodes oxidizing sludge achieved 4,4-(propane-2,2-diyl)diphenol, nonylphenol, 5-chloro-2-(2,4-dichlorophenoxy)phenol removal of 89, 73, and 82% at a pH value of 3.0 and current density of 40 mA/cm^2 for 1 h. The volatile solids of the sludge were mineralized by 23%, and chemical oxygen demand was removed by a value of 27% (Barrios et al. 2015). 42% mineralization of emerging pollutants was achieved at a pH of 3.0 and decreased to 25% and 14% for pH values of 5 and 7, respectively (Barrios et al. 2015).

Moreover, zeta potential decreased from -17 to -10 mV due to mineralization, destroying and destabilizing of negatively colloidal particles and eventually improving sludge quality. Chemical oxygen demand removal was maximized at a level of 31% at the current density of 40 mA/cm and highly dropped at 10 mA/cm. Titanium (Ti) anodes removed pathogens and enhanced sludge dewatering (Drogui et al. 2012).

Emerging pollutants are destabilized and destroyed by hydroxyl radical (▪OH) released by electrodes. The ▪OH radical reacts with emerging pollutants (Eq. 2),

$$\mathrm{RH} +^{\circ} \mathrm{OH} \rightarrow^{\circ} \mathrm{R} + \mathrm{H_2O} \quad (2)$$

The electrochemical cell produces hydroxyl radical, which reacts with emerging pollutants in the sludge. The extracellular polymeric substances further react with hydroxyl radical or oxygen to generate oxidative byproducts (Eqs. 3 and 4).

$$^{\circ}R + O_2 \rightarrow \text{Products} \quad (3)$$

$$^{\circ}R +^{\circ} OH \rightarrow \text{Products} \quad (4)$$

The oxidation of 4,4-(propane-2,2-diyl)diphenol by ▪OH radicals breaks the adjacent bonds, i.e., methyl bridge, into phenol and iso-propylene alcohol, as shown in Fig. 3. The iso-propylene alcohol molecule is further oxidized by ▪OH producing short-chain carboxylic acid, while catechol, hydroquinone, and resorcinol compounds might be produced from the hydroxylation of the phenolic ring. Hydroquinone was the main byproduct in earlier studies (Gözmen et al. 2003). Subsequently, quinone is formed by a dehydrogenation reaction with ▪OH radicals. Phenolic ring molecules are mainly oxidized and converted into small fragmented byproducts, resulting in short-chain aliphatic acids. Maleic, oxalic, fumaric, formic, and acetic acid are the main aliphatic acids. Those byproducts could be efficiently utilized by anaerobes for bioenergy production. However, extending the reaction time will allow the attack of ▪OH on those acids, resulting in carbon dioxide of complete degradation of bisphenol A (Gözmen et al. 2003).

Carboxylic acids are the main byproducts of the degradation of chlorophenols, as shown in Fig. 4. 5-chloro-2-(2,4-dichlorophenoxy)phenol, known as triclosan, molecules are completely oxidized with conductive-diamond electrochemical (Gözmen et al. 2003). Electro-oxidation cell was initially operated at pH of 3.0, current density of 40 mA/cm^2 and reaction time of 60 min, and achieved removal efficiency of 73% for 4,4-(propane-2,2-diyl) diphenol, known as bisphenol A; 89% for nonylphenol;

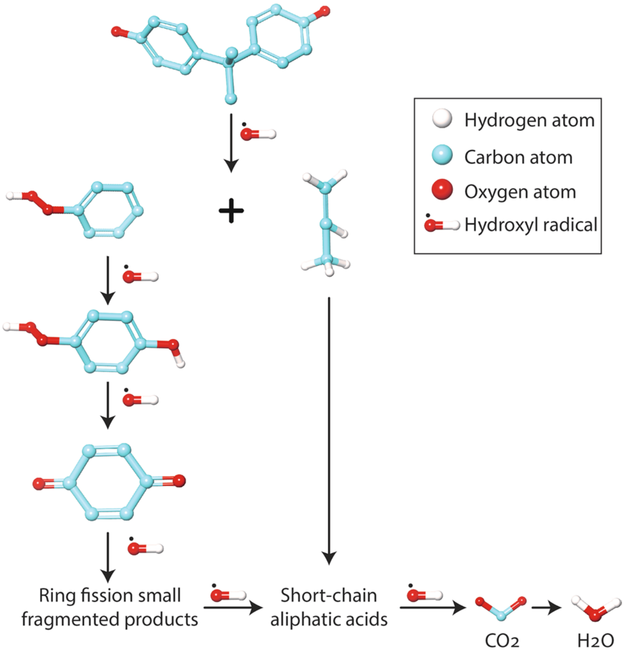

Fig. 3 Anodic oxidation reaction mechanism of 4,4-(propane-2,2-diyl)diphenol, known as bisphenol A, with a conductive diamond electrode (Martín de Vidales et al. 2013). The oxidation of 4,4-(propane-2,2-diyl)diphenol by ▪OH radicals breaks the adjacent bonds, i.e., methyl bridge, into phenol and iso-propylene alcohol. The iso-propylene alcohol molecule is further oxidized by ▪OH producing short-chain carboxylic acid. Those byproducts could be efficiently utilized by anaerobes for bioenergy production. The attack of.▪OH on acids results in carbon dioxide

and 82% for 5-chloro-2-(2,4-dichlorophenoxy)phenol (Barrios et al. 2015).

Ultrasonication

Ultrasonication is an efficient mechanical pretreatment process of the sludge to enhance biodegradability (Pilli et al. 2011). Ultrasonication is regarded as digesting the sludge easier by changing sludge's physical, biological, and chemical properties. The degree of sludge breakdown is mainly affected by the sonication parameters and sludge characteristics. The full-scale applications of the ultrasonication system efficiently demonstrated a 50% increase in biogas productivity. Moreover, a net energy gain to electricity consumption ratio of 2.5 was found in the energy balance by the ultrasonic system. According to the frequency, the ultrasound range diagram could be separated into three sections: power ultrasound (20–100 kHz), high-frequency ultrasound (100 kHz–1 MHz), and diagnostic ultrasound (1–500 MHz).

Thermal hydrolysis

Thermal hydrolysis is a viable method of making sewage sludge more susceptible to anaerobic digestion. Thermal hydrolysis is a procedure that involves heating sludge to the desired temperature, typically using saturated steam injection, to facilitate subsequent anaerobic digestion (Díaz et al. 2020). The best treatment temperature for thermal hydrolysis is between 160 and 180 °C with a pressure ranging from 600 to 2500 kPa in several experiments (Stuckey and McCarty 1984; Neyens and Baeyens 2003; Bougrier et al. 2008; Carrère et al. 2010; Barber 2016). Furthermore, some thermal hydrolysis investigations have been conducted at temperatures ranging from 60 to 220 °C and for durations ranging from 1 to 4320 min (Gavala et al. 2003; Climent et al. 2007; Ferrer et al. 2008; Wilson and Novak 2009). There are many advantages of raising the temperature of thermal hydrolysis to an ideal range, including reducing the average particle size and apparent viscosity, improving sludge digestibility, and enhancing biopolymers solubilities, such as proteins and carbohydrates, while lipids are not largely affected. Also, there is a potential for refractory compounds reproduction, such as color, chemical oxygen demand, and nitrogen (Barber 2016; Kor-Bicakci and Eskicioglu 2019).

Ozonation

The aim of ozonation during sludge pretreatment is to induce organic matter partial oxidation and hydrolyzation. Ozonation destroys pathogens and volatile solids during the anaerobic digestion of sludge. Ozone damages bacteria's cell walls at low doses, resulting in their lysis. At higher levels, ozone destroys flocks by directly attacking the bacterial extracellular polymeric matrices critical for bacterium colonies in the flocks (Chiellini et al. 2014). Furthermore, many studies have also demonstrated that ozonation may help eliminate or increase the biodegradation of emerging pollutants. Ozone doses from 0.1 to 30 mg/L were used to remove the contaminants by 60–99%, such as antibiotics, pesticides, natural and synthetic estrogens, anti-epileptics, and anti-inflammatories (Carballa et al. 2007).

Moreover, organic solids reduction and methane generation have both improved significantly during ozonation. Anaerobic biodegradability and biomethanation were increased with sludge pretreatment aided by ozone dosage. An ozone dosage of 0.06 g O_3/g total suspended solids was used to achieve the greatest daily elimination efficiency of 32% total solids, 69% volatile suspended solids, 42% volatile solids, 35% chemical oxygen demand, and 60% total suspended solids. Ozone also resulted in a 48% increase in methane-enriched biogas generation via the acetotrophic route (Tuncay et al. 2022).

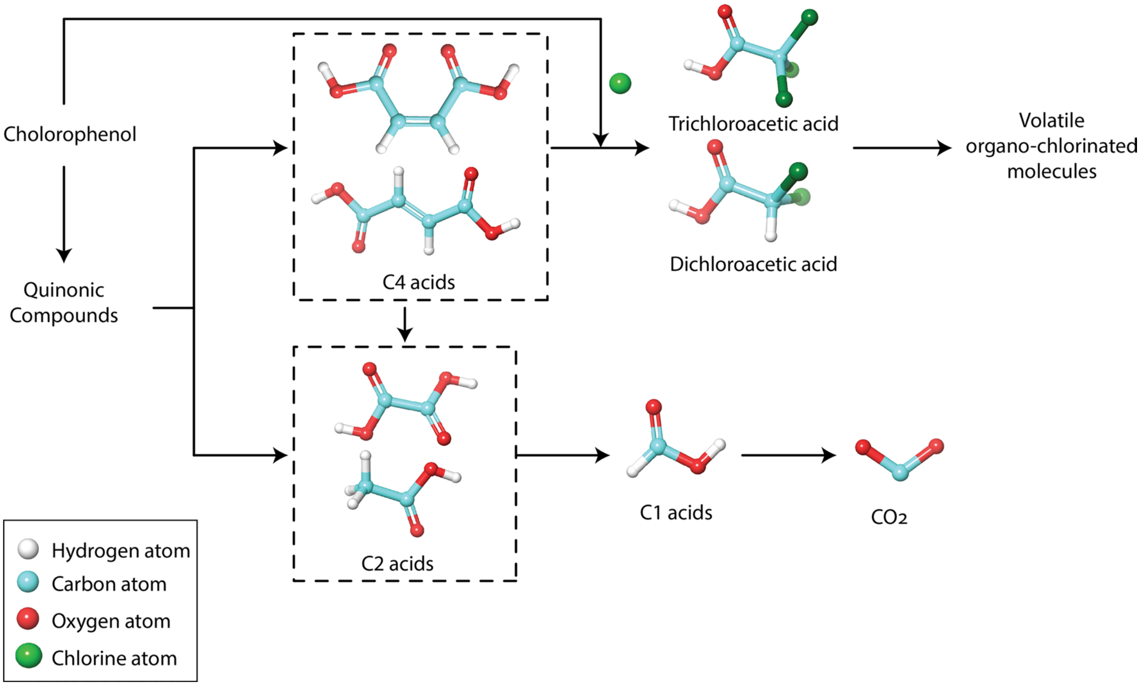

Fig. 4 Anodic oxidation reaction mechanism of 5-chloro-2-(2,4-dichlorophenoxy)phenol, known as triclosan, with a conductive diamond electrode (Martín de Vidales et al. 2013). Carboxylic acids are the main byproducts. 5-chloro-2-(2,4-dichlorophenoxy) phenol molecules are completely oxidized with conductive-diamond electrochemical. The optimum operating conditions for the electro-oxidation process are acidic pH of 3.0, a current density of 40 mA/cm.^{2} and a reaction time of 60 min

Degradation and transformation of emergent contaminants during sludge pretreatment

Including transformation products during studying emergent contaminants removal is critical as the transformed products might be more toxic than the original compound. For instance, carbamazepine is converted into acridone and acridine, which are more poisonous to algae, bacteria, and daphnia magna than the original compound (Donner et al. 2013). During wastewater biological treatment, nonylphenol ethoxylates are bio-transformed into various metabolites, including nonylphenol diethoxylate, nonylphenol, and nonylphenol monoethoxylate (Chang et al. 2005; Patureau et al. 2008). Also, estrogen E1 is transformed into E2 during anaerobic conditions with various kinds of sludge (des Mes et al. 2008; Paterakis et al. 2012). Furthermore, the antimicrobial triclocarban can be converted partly or entirely into a variety of products through a variety of abiotic and biotic mechanisms. Triclocarban is biotransformed into similar compounds with decreased chlorine content in anaerobic and anoxic environments through the reductive dechlorination process (Kor-Bicakci et al. 2020).

Moreover, naproxen, a non-steroidal anti-inflammatory drug, is biotransformed into 6-O-desmethylnaproxen, which persists in the anaerobic therapy for up to 161 days (Lahti and Oikari 2011; Azizan et al. 2021). Additionally the antibiotic spiramycin I was degraded into three molecules during the anaerobic digestion through the hydrolysis of mycaminose-mycarose and the hydrogenation of the aldehyde group (Zhu et al. 2014). The retransformation of N4-acetylsulfamethoxazole to sulfamethoxazole was strongly indicated during activated sludge treatment (Göbel et al. 2005).

Bioaugmentation

The addition of certain microorganisms is a potential technique for improving the performance of degrading specific contaminant compounds for suboptimal anaerobic digestion systems. This process aims to transform hazardous contaminants into less harmful ones to reduce the contaminant load. Also, adding nutrients or electron acceptors may stimulate the local microbiota, which might help improve the microbial breakdown of pollutants. However, some strains may not develop due to a lack of adaptation to environmental factors, predation, or competition with microbes. Studies showed that microorganisms that degrade pharmaceuticals and personal care products accelerated their breakdown in the activated sludge (Dubey et al. 2021).

Using *Caseobacter* sp. can get rid of oil and grease by 66.15% in bakery wastewater (Keenan and Sabelnikov 2000). Also, *Paracoccus* sp. LZ-G1 was shown to adsorb cadmium (Cd^{2+}) on the cell surface and reduce Cd^{2+} content in the microbial community, thereby improving anaerobic digestion, hydrolysis efficiency, and methane production (Guo et al. 2021). Furthermore, the addition of *Coprothermobacter proteolyticus* increased the hydrolysis and fermentation of proteins and polysaccharides (Lü et al. 2014). Additionally, mono-oxygenase enzymes, which have a broad variety of substrates, have been shown to break down pharmaceuticals and personal care products in several microorganisms (Zhou et al. 2014). Moreover, Roh and Chu (2011) revealed that starting with identical biomass of the 17-estradiol-degrading bacteria, *Sphingomonas* KC8, removing 1 mg/L of 17-estradiol in lab-scale sequencing batch reactors resulted in substantial amounts of elimination (Roh and Chu 2011).

Algae treatment has been shown to remove heavy metals and excess nutrients from wastewater more effectively compared to chemical treatment (Maryjoseph and Ketheesan 2020; Singh et al. 2021). For instance, *Chlorella pyrenoidosa* mixotrophic was cultured in an anaerobic digestate of sludge, resulting in 95% of orthophosphate, 99% of ammonium, and 62% of total organic carbon removal (Tan et al. 2020). Microalgae strains of *Scenedesmus* sp., *Chlorella*, and *Chlamydomonas* are highly adaptable to severe environmental circumstances. Microalgae species have been shown to remove pharmaceuticals. For instance, *Chlamydomonas* sp. Tai-03 was reported to remove 54.53% of ciprofloxacin and 100% of sulfadiazine (Xie et al. 2020). Also, *Haematococcus pluvialis* removed 84% of sulfamerazine, 74% of sulfamethoxazole, and 75% of sulfamonomethoxine (Kiki et al. 2020). Also, a range of microalgae species has been shown to contribute to the removal of personal care products, such as methylisothiazolinone, bisphenol A, climbazole, and triclosan (Wang et al. 2013; Bai and Acharya 2016; Pan et al. 2018; Xie et al. 2020); hormones, such as progesterone, estrone, and estriol (Peng et al. 2014; Maes et al. 2014); pesticides, such as propamocarb, trichlorfon, and isoproturon (Dosnon-Olette et al. 2010; Ardal 2014; Wan et al. 2020); and surfactants, such as nonylphenol (He et al. 2016).

Fungal bioremediation has emerged as a more cost-effective and long-term solution. Studies have shown that fungi can digest a broad range of chemicals, including emergent contaminants created by human activities. In biopiles systems, the sludge being treated is combined with a bulking substance used as a co-substrate by the fungi and enhances aeration (Khan et al. 2004; Llorens-Blanch et al. 2018). Furthermore, the whit-rot fungus *Tinea versicolor* is capable of degrading a broad variety of organic contaminants through the intracellular system, such as cytochrome P450, and extracellular ligninolytic highly oxidative enzymes (Asgher et al. 2008; Yang et al. 2013; Rodríguez-Rodríguez et al. 2014).

The role of extracellular polymeric substances in the removal of emerging contaminant

Extracellular polymeric substances are a complex combination of polymers released by microorganisms with a high molecular weight. Extracellular polymeric substances have a strong affinity for binding with organic pollutants, such as triclosan (Yan et al. 2019a), sulfamethazine (Xu et al. 2013), sulfonamides (Xu and Sheng 2020), phenanthrene (Bai et al. 2016), and heavy metals (Wei et al. 2017). Sorption of bisphenol A during the fermentation process takes place by sludge via strong binding to extracellular polymeric substances (Yan et al. 2019b), and subsequently biodegradation mechanism removal (Zhao et al. 2008). As Zhou et al. (2019) reported, triclocarban was partitioned onto the sludge. The removal of triclocarban mainly occurred due to the binding of the contaminant in sludge by extracellular polymeric enzymes (Yan et al. 2019b). Kindly check the section headings are correctly identified. yes, it is correctly identified

Anaerobic digestion and the circular economy

The goal of a circular economy is to reduce waste and increase the amount of recycling and reusing. Anaerobic digestion provides an appropriate scenario for the circular economy. Nutrient recycling and sustainable biosolids management difficulties may be solved through anaerobic digestion, which is considered environmentally friendly, protecting the environment and reusing the materials more wisely. Sustainable-renewable resources and fuels can be produced through anaerobic digestion. For instance, in 2016, 40% of food waste was processed to recover nutrients, and 32% was treated to recover both nutrients and energy (Fagerström et al. 2018). For a gas-powered automobile, 1200 kWh of biogas from one ton of food waste is needed for 1900 km. Also, a gas bus powered by food waste of 3000 families can run for a full year (Fagerström et al. 2018).

Additionally, anaerobic digestion is regarded as the most cost-effective biowaste treatment approach. Anaerobic digestion allows for the recovery of energy and the production of digestate rich in nutrients while reducing the natural consequences of waste transportation. Because of the high concentration of nutrients in the digestate derived from the raw materials, digestate is suitable for use as a fertilizer or organic amendment in agricultural operations (Wainaina et al. 2020).

Conclusion

The detection of emerging contaminants in nature has increased as detection technology has improved. Numerous emerging contaminants, such as personal care compounds, endocrine-disrupting chemicals, medicines, and converted products, whose presence at trace levels in treated wastewater poses a threat to human health and aquatic ecosystems. Despite this, little is known about the fate of these pollutants, how emerging pollutants interact with the environment, and the most effective methods for removing them from the environment. Anaerobic digestion has been studied as a potential method for removing these contaminants. The effect of extracellular polymeric enzymes on anaerobic digestion and the associated inhibitory mechanisms were thoroughly discussed. However, following treatment of these highly toxic and damaging pollutants, the concentration of their persistent transformation products is still limited, which needs to be a major focus in future studies.A funding declaration is mandatory for publication in this journal. Please confirm that this declaration is accurate, or provide an alternative.

Furthermore, combining multiple treatments to remove emerging pollutants may be more effective than single traditional technology. As a result, pretreatment procedures such as electro-oxidation, ultrasonication, thermal hydrolysis, and ozonation have been used to mitigate anaerobic digestion inhibition caused by emerging contaminants. Finally, future research should summarize all important factors influencing sewage sludge treatment with emerging pollutants.Kindly check the affiliations are correctly identified.

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