



## Review



# Occurrence, spatiotemporal trends, fate, and treatment technologies for microplastics and organic contaminants in biosolids: A review

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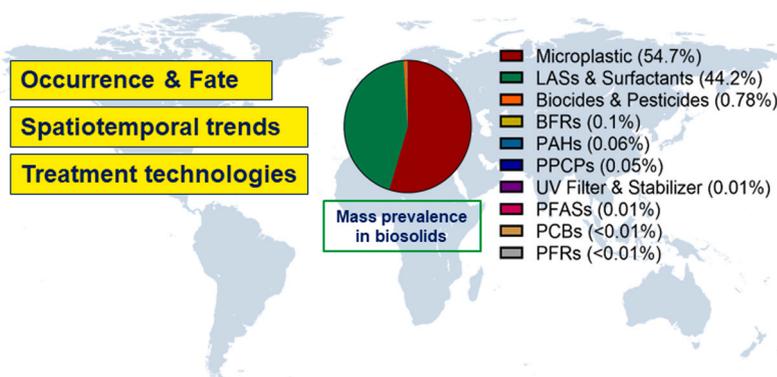
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## HIGHLIGHTS

- LAS (44.2%) and microplastic (54.7%) account for the highest prevalence in biosolids.
- Occurrence of several OCs in Europe is higher than in Asia and America.
- Australia has microplastic in biosolids 10 times higher than other continents.
- Anaerobic digestion is the most mature and practical for OCs treatment in biosolids.
- Thermal treatment is a viable option but still requires additional improvements.

## GRAPHICAL ABSTRACT



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## ABSTRACT

This review provides a comprehensive overview of the occurrence, fate, treatment and multi-criteria analysis of microplastics (MPs) and organic contaminants (OCs) in biosolids. A meta-analysis was complementarily analysed through the literature to map out the occurrence and fate of MPs and 10 different groups of OCs. The data

Fate  
Occurrence  
Spatiotemporal trend  
Treatment  
Multicriteria analysis

demonstrate that MPs (54.7% occurrence rate) and linear alkylbenzene sulfonate surfactants (44.2% occurrence rate) account for the highest prevalence of contaminants in biosolids. In turn, dioxin, polychlorinated biphenyls (PCBs) and phosphorus flame retardants (PFRs) have the lowest rates (<0.01%). The occurrence of several OCs (e.g., dioxin, per- and polyfluoroalkyl substances, polycyclic aromatic hydrocarbons, pharmaceutical and personal care products, ultraviolet filters, phosphate flame retardants) in Europe appear at higher rates than in Asia and the Americas. However, MP concentrations in biosolids from Australia are reported to be 10 times higher than in America and Europe, which required more measurement data for in-depth analysis. Amongst the OC groups, brominated flame retardants exhibited exceptional sorption to biosolids with partitioning coefficients ( $\log K_d$ ) higher than 4. To remove these contaminants from biosolids, a wide range of technologies have been developed. Our multicriteria analysis shows that anaerobic digestion is the most mature and practical. Thermal treatment is a viable option; however, it still requires additional improvements in infrastructure, legislation, and public acceptance.

## 1. Introduction

Biosolids are by-products of the wastewater treatment process. Wastewater sludge, once subjected to physical and chemical treatments to reduce volume, eliminate pathogens, and/or stabilize its organic content, is then referred as biosolids. Biosolids are rich in nutrients such as nitrogen, phosphorus, and organic matter, hence they have been considered a resource for end-use applications in agriculture and other land-based applications. Reusing biosolids is a worldwide sustainable practice to manage the large quantity of biosolids produced on a global scale. Each country has its own pattern of end-use applications (Fig. 1). In Australia, an estimated 300–400 kilotons dry weight (dw) of biosolids are produced annually, and approximately two-thirds of the biosolids have been used for agricultural and non-agricultural (forestry, landscaping, construction) end-use applications [12]. The corresponding percentages of biosolids for reuse in the EU and the USA are 37% and 60%, respectively [47].

Although it may seem desirable to increase the reuse of biosolids globally as a means to capture their high nutrient content and improve soil quality and productivity, biosolids also harbour various organic contaminants (OCs) and microplastics (MPs) that can be released into the environment [148,211]. The volume of anthropogenic compounds being introduced into the market continues to increase, and these materials end up in biosolids in either their original or transformation by-product forms [139,188]. Concentrations of OCs in biosolid-amended soil were found to be several orders of magnitudes higher than a reference soil, given that part of these contaminants migrate into the surrounding plants, animals, and humans via the food cycle, which can cause severe health and environment problems [67, 167].

To minimize the risk of contaminant exposure to humans and the environment, legislation regulating contaminants in biosolids have been in place since the 1980 s. In 1993, the United States issued “The

Standards for the Use or Disposal of Sewage Sludge”, which regulate different usages of biosolids, and which set an upper limit for heavy metals and pathogens. Later, when organic contaminants emerged, and to address the concern relevant to OCs, nationwide studies were conducted in the US, EU, Australia, and China [139,148,22].

To mitigate the risk caused by biosolids to human health, it is important to understand their spatiotemporal trends and the fate of MPs and OCs in biosolids. In the last decades, extensive monitoring and archiving programs for that purpose have been conducted. For example, the United States Environmental Protection Agency (USEPA), Swedish Environmental Protection Agency, Germany, Switzerland, and China have been involved in global monitoring studies [141,206]. USEPA pioneered studies on the occurrence of OCs in biosolids in the 1980 s. From 1982 to 2007, USEPA developed a national monitoring program for contaminants in biosolids, looking at 72 pharmaceuticals, 25 steroids and hormones, and several other OCs across 35 states [206].

Biosolids in China are particularly concerned due to significant levels of MPs and OCs detected in biosolids; for example, 54% of the detected OCs are organo-halogen contaminants [139]. In 2013, 6.2 million tons of biosolids were produced in China, and up to 80% of biosolids were not disposed of safely (directly disposed to surface water or open fields) [139]. Despite many nationwide surveys, to our knowledge, no systematic meta-data studies of the global fate and spatiotemporal trend of an extensive set of MPs and OCs in biosolids have been reported. Similarly, there is still a limited understanding of the effect of contaminant characteristics and socio-demographic factors which lead to contaminants in biosolids which is also worthy of systemic investigation. Those factors are vital as they compromise the end-use application of biosolids.

The occurrence of MPs and OCs in biosolids is strongly driven by a set of chemical factors such as characteristics of the chemical themselves, basic operating conditions of the sewage treatment process, such as pH, sludge retention time, temperature, and social-demographic factors influencing their upstream water quality [127,223,239]. Sludge

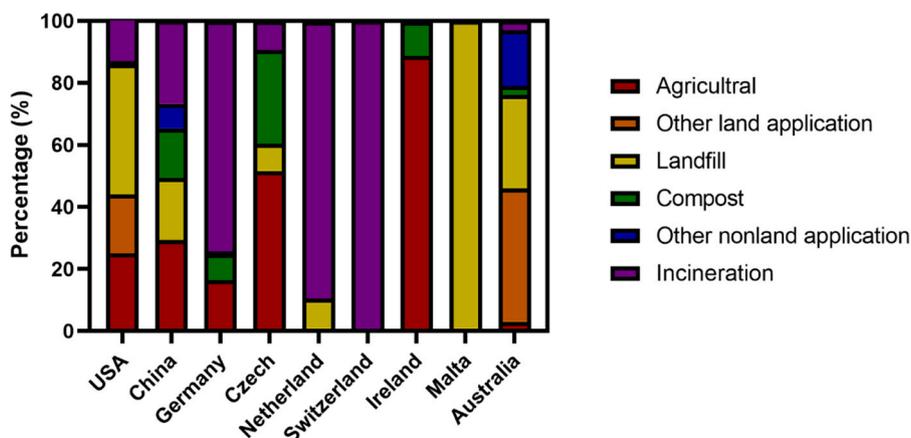


Fig. 1. Pattern of biosolids applications in different countries [12,204,220,57].

treatment technologies also significantly influence the degree of treatment of MPs and OCs in biosolids. Most traditional sludge stabilization technologies include aerobic digestion, anaerobic digestion, alkaline stabilization, advanced oxidation processes, composting, and thermal treatment [63,175]. Recently, a few novel technologies have been introduced for recalcitrant MPs and OCs, such as hydrothermal alkaline treatment and super critical water oxidation [105]. Given the range of technologies available for biosolids treatment, there has yet to be a systematic review of the technologies' performance, operating conditions, strengths, and weaknesses to establish guidelines for a broader range of users.

This study presents a systematic review and a meta-analysis of MPs and 10 selected OCs in biosolids collected from 249 publications (Table S1), based on the past 30 years of scientific literature. A wide range of OCs are present in biosolids; however, this study focused on 10 prevalent OCs groups, with each group comprised of the chemicals listed in Appendix 1. The chart string development and data collection method were provided in Appendix 2. These groups were selected for review based on their high usage and release worldwide, frequent detection in biosolids, potential risks to human health and ecosystems, and data availability in the literature. The objectives are to (i) study the occurrence and spatiotemporal trends of the selected MPs and OCs in biosolids, (ii) assess the factors influencing the fate of MPs and OCs in biosolids, (iii) discuss the progress of various treatment technologies, and (iv) perform a multicriteria analysis of the technologies.

## 2. Meta-data analysis on the occurrence, spatiotemporal trends, and fate of OCs in biosolids

### 2.1. Occurrence of OCs

Fig. 2 presents the reported occurrence of MPs and the 10 selected OCs and their constituent compounds in biosolids, and Fig. 3 presents their reported concentration, as found in our literature review of wastewater treatment plants (WWTPs). The biosolids in this review are from WWTPs which might receiving solely or both domestic and industrial wastewaters from various source at different degrees. Among the contaminants, MPs and linear alkylbenzene sulfonates (LAS) and other surfactants are by far the most prevalent contaminants in biosolids, with almost 98% detection rate and reaching levels as high as  $10^5$  µg/kg. While microplastic leakage into biosolids was limited before the 1990s, it has increased substantially since then [148]. LAS are a commonly used class of surfactants found in household and industrial cleaners and detergents, and their annual global demand has increased from 1.8 million metric tonnes in 1980 to 2.4 million metric tonnes by 2000 [93]. However, LAS and other surfactants are not readily biodegradable in anaerobic conditions, resulting in high concentrations in primary and secondary effluents and biosolids [13]. High levels of LAS and other surfactants are also attributable to chemicals used in the wastewater treatment process, such as polymers used as dewatering

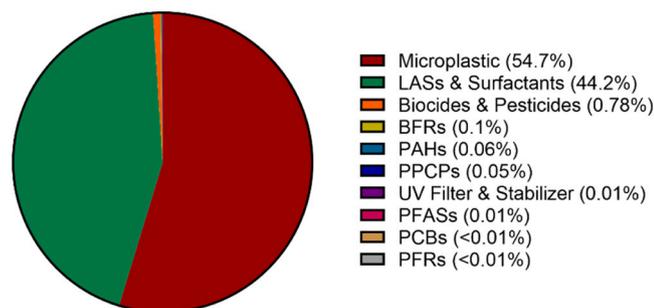


Fig. 2. Occurrence (%) of OCs groups in biosolids. The prevalence of dioxins is insignificant as shown in the figure. The calculation method for the occurrence of OCs is provided in Appendix 3.

aids.

Brominated flame retardants (BFRs) are another group of chemicals of concern as they have the third-highest concentration in biosolids at a reported range of approximately  $10^1$  µg/kg. BFRs are frequently used in plastics and in the textile industry, and have a high affinity to solids, contributing to their accumulation in biosolids [186].

Dioxins were rarely found in biosolids and do not even appear in Fig. 2. The concentration of dioxins in biosolids was found to be the lowest of all OC groups, with reported levels in the range of  $10^{-2}$  or  $10^{-3}$  µg/kg. Dioxins are not intentionally produced, but they are released through incineration and combustion processes and can accumulate in biosolids through various indirect pathways.

To gain further insight into the occurrence of MPs and OCs in biosolids, we have focused to analyse the concentration of MPs and the 10 OC groups, and their main constituent chemicals, as shown in Fig. 3. Each OC group is illustrated with several chemicals with the highest concentration. The following sections discuss the primary contaminants (by concentration) within each of the 10 OC groups.

#### 2.1.1. Microplastics

Microplastic are known to be widely distributed in soils, food, freshwater, rainwater, snow, and sea ice; hence, it also appeared substantially in biosolids. Microplastics, including polyethylene terephthalate (PET), polyethylene (PE), polystyrene (PS), and polyvinyl chloride (PVC), have been found to have the occurrence levels from  $10^5$  to  $10^6$  µg/kg dw. PET has been detected at levels ranging from µg/g to mg/g in biosolids across reviewed studies (Müller et al., 2020; [240]), while mg/g levels of PET, PE, PS and PVC have been reported in biosolids [52,148]. Recent studies suggest that PE is the most prevalent microplastic in the biosolids (accounting for 54% of the total sampled microplastics), followed by PVC (26%), PET (11%), and PS (3%) [148, 149]. Occurrence and composition of microplastics in biosolids mainly depend on the wastewater (WW) catchment, i.e., prevalence of microplastic fibre from the textile and laundry process and packaging [77].

#### 2.1.2. Linear alkylbenzene sulfonates and surfactants

Concentrations of LAS and surfactants are present in a much greater magnitude than the others (apart from MPs), i.e., 200 to 1500 mg/kg of LAS are found in sewage sludge [13], and 2300 to 3600 µg/kg of anionic surfactants are found in stabilized sludge [3]. Anionic surfactants are among the most abundant compounds, with 23 different compounds being identified in sludge at various stages of stabilization treatment [3]. The levels of these surfactants in biosolids can be influenced by treatment technology and the physicochemical properties of the surfactants. For example, Abril et al. [3] reported that aerobic digestion treatment technology tends to increase biosolids' sorption of anionic surfactants. After anaerobic digestion, the concentrations of four anionic surfactants (AS-C12, AS-C14, AS-C16, and AS-C18) decreased. But after aerobic treatments, a decrease of the shorter chain surfactant was found for AS-C12, while concentrations of AS-C14 were not affected, and the concentrations of the longer chain surfactants (AS-C16 and AS-C18) increased [3]. These findings indicate that treatment technology and surfactant physicochemical properties determine the levels of these compounds in biosolids.

#### 2.1.3. Biocides and pesticides

Biocide chemicals such as triclocarban (TCC), triclosan (TCS), and benzotriazoles (BT and 5-TT) are the most prevalent biocide contaminants detected in biosolids. Lai et al. [100] reported that BT and 5-TT concentrations in biosolid-amended soils were around 150 µg/kg, while levels of TCC were approximately 440 µg/kg, which was much higher than TCS (2.2 µg/kg) in stabilized sludge [3]. It can be explained by higher use of TCC in producing personal care products [34]. Levels of biocides in biosolids vary by treatment technology. For example, Abril et al. [3] found that TCC and TCS, which are recalcitrant compounds, may be removed to a lesser extent under aerobic conditions. Similarly,

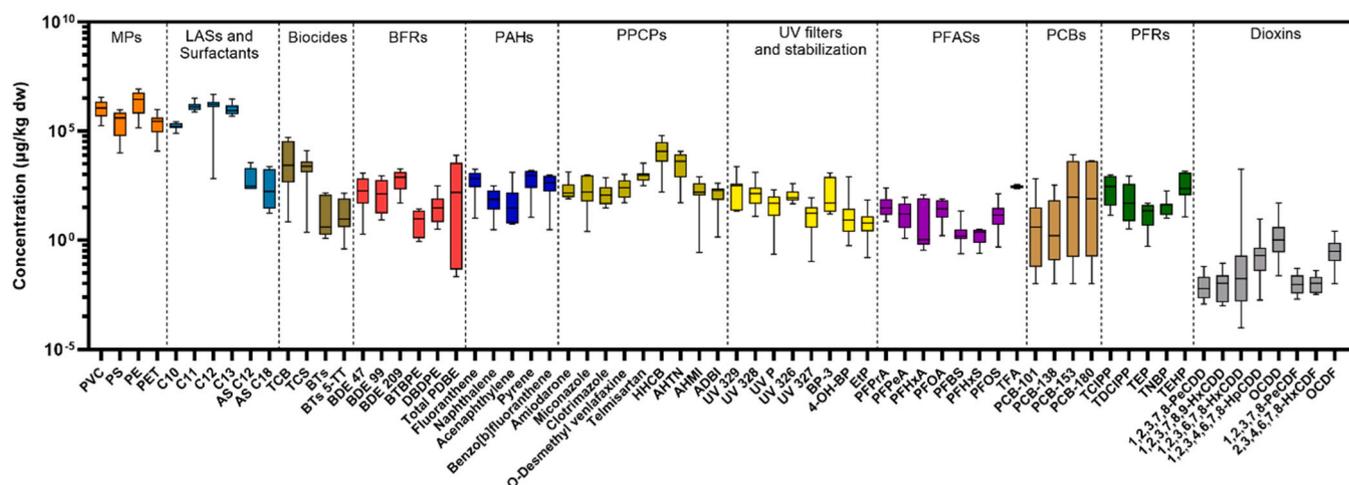


Fig. 3. Reported concentrations of MPs and the 10 selected OCs and their constituent compounds in biosolids. The number of studies involved in each group are similar to Fig. 1. Data are presented as box plots (box: 25–75% interquartile range; bar: median; whisker: Tukey).

the removal of TCC and TCS from wastewater (WW) depends on the adsorption, abiotic degradation, and microbial mineralization or transformation reactions in the WWTPs [161].

#### 2.1.4. Brominated flame retardants (BFRs)

A large difference was found in the concentration of BFRs chemicals. The discrepancy of the maximum and minimum concentration of total Polybrominated Diphenyl Ethers (PBDEs) was wide, with a 370 thousand-fold difference. The reason is due to the different sources where BFRs were discharged (e.g., domestic or industrial inputs). It might also be due to the appearance of MPs which causing the spike of PBDEs in biosolids. High concentrations of total PDBEs in biosolids were found in domestic wastewater, i.e., 4300 to 7800 µg/kg [44,49,71]. High occurrence of PDBEs, i.e., BDE 47, BDE 99, BDE 100 and BDE 209 constitute 90% of total PBDEs [48]. Occurrences of some PBDE replacements, such as BTBPE and DBDPE, were also detected at levels comparable to or lower than those of PBDEs [48].

#### 2.1.5. Polycyclic aromatic hydrocarbons

Polycyclic aromatic hydrocarbons (PAHs) are originated from both natural (e.g., fossil carbon, natural fires) and artificial (e.g., wood-burning, biofuel combustion) sources. Pertaining to artificial source, the meat processing industry is one considerable point source of PAHs contaminants [51]. Various studies have reported PAHs in biosolids at µg/kg to mg/kg levels. However, the occurrence levels of PAHs in biosolids are mainly driven by the characteristics of WW, treatment technologies, and geographical differences [227]. Chen et al. [32] report a total of 16 PAH concentrations in biosolids of WWTP ranging from 70 to 140 µg/kg dw, which was about 4 to 16 times higher than levels in the biosolid of drinking water treatment plants. This indicates that PAHs in biosolid is mostly produced from anthropogenic activities other than natural sources. However, even in WWTPs, levels of PAHs in biosolid are subject to a wide range of variation. Oleszczuk [150] confirmed this observation through the monitor of biosolids from five WWTPs which received mainly municipal WW. The results showed the variations of different PAHs in biosolids at five municipal sewage plants (3700 to 11,300 µg/kg). Among the lists of PAH compounds, the four- (pyrene) and three-ring (phenanthrene) compounds were dominant in all the biosolids. Phenanthrene presented in biosolid at 2100 µg/kg dw, accounting for > 38% of the total monitored PAHs, followed by pyrene (1500 µg/kg dw) [2].

#### 2.1.6. Pharmaceutical and personal care products

PPCPs are one of the most substantial chemical groups, with hundreds of compounds identified in biosolids at µg/kg to g/kg levels.

Castro et al. [29] reported 37 emerging PPCPs in sewage sludge from different WWTPs. The highest occurrence of PPCPs includes amidarone, miconazole, clotrimazole, O-Desmethyl venlafaxine, and telmisartan (e.g., 732 to 1303 µg/kg). Those PPCPs are high-production volume pharmaceuticals [79]. Synthetic polycyclic musks (PCMs) are fragrance compounds of interest and have been monitored in dewatered sludge samples [178,236,33]. Galaxolide (HHCB) and tonalide (AHTN) were the two predominant PCMs in biosolids with a concentration of 50 to 170,000 µg/kg and 80 to 704,000 µg/kg, respectively. The highest occurrence was found in biosolids from a cosmetic plant [236]. This suggests the association with the extensive use patterns and discharge sources of these two products.

#### 2.1.7. UV filters and stabilizers

UV filters and stabilizers have been used universally for skin and product protection against sun light. The occurrence of UV filters and stabilization chemicals has been reported, mostly at µg/kg to mg/kg levels [100,145,146]. The concentration of benzotriazole ultraviolet was found from 50 to 390 µg/kg in biosolid-amended soils [100]. The chemical 4-OH-BP is one of the most abundant benzophenones with high concentrations detected in influent wastewater [218]. Also, 4-OH-BP can be formed by alkylphenol degradation, which can transform BP-3 into 4-OH-BP [218]. UV-329, UV-328, and UV-P were monitored at high occurrence in biosolids from 10 to 100 µg/kg [100]. The relatively-high occurrence of UV filters in biosolids could be explained by its high sorption tendency (high log partition coefficient ( $K_{ow}$ ) value such as BP-3:  $\log K_{ow} = 3.8$ , and other surfactants and biocides ranged from 4.4 to 7.1) to biosolids after aerobic digestion treatment [3].

#### 2.1.8. Per- and polyfluoroalkyl substances

Per- and polyfluoroalkyl substances (PFASs) possess unique water and oil repellency characteristics and continue to be used in a broad variety of consumer and industrial products (e.g., various food packaging, cookware, performance clothing, class B aqueous film foaming foam). As can be seen in Fig. 3, the PFASs included in this OC group (perfluoropropanoic, -pentanoic, -hexanoic, and -octanoic acids (PFPrA, PFPeA, PFHxA, and PFOA, respectively), and perfluorobutane, -hexane, and octane sulfonic acids (PFBS, PFHxS, and PFOS, respectively) have a reported concentrations in the range of  $10^0$  to  $10^1$  µg/kg in biosolids.

According to Li et al. [108], the highest PFPrA concentrations were found in the biosolids from a facility with chemically-enhanced primary treatment that had a 70% domestic and 30% industrial feed stream. That study demonstrated that different ratios of domestic and industrial sources resulted in different concentrations of PFASs in biosolids. Also, Li et al. [108] reported PFPeA concentrations ranging from 1.7 to

46.7 µg/kg in the biosolids from other sources. Abril et al. [3] also found PFHxA levels up to 120 µg/kg in a stabilized sludge, and [108] found up to 110 µg/kg in the biosolids from a waste-activated sludge facility treating 93% domestic and 7% industrial sources. As hydrophobic compounds, the longer PFASs tend to absorb more into biosolids than the short-chain PFASs [116].

#### 2.1.9. Polychlorinated biphenyls

Polychlorinated biphenyls (PCBs) are used widely in manufacturing industries (e.g., electrical, heat transfer and hydraulic equipment). PCBs presented at low-level µg/kg in biosolids. Even though PCBs are banned, the legacy release of PCBs from old equipment, recycling operations, and old stocks can occur [89]. For example, PCB 28 (24.8 µg/kg), PCB 52 (10 to 12.8 µg/kg), PCB 101 (10 to 31.4 µg/kg), PCB 118 (10 to 17.5 µg/kg), PCB 138 (10 to 68.6 µg/kg), PCB 153 (10 to 92.6 µg/kg), and PCB 180 (10 to 80.2 µg/kg) are the dominant compounds which originated from urban water resource recovery facilities [89]. The occurrences of PCBs in WW and biosolids can be correlated to their main sources and transport, such as atmospheric deposition, surface runoff, and industrial discharges [23,152].

#### 2.1.10. Phosphorus flame retardants

Occurrences of PFRs in biosolids sharply increased at the same time BFRs were phased out, likely due to their use as substitutes [36]. Liu et al. [124] investigated the levels of different PFR compounds in a WWTP treatment train and found that some (tripropyl phosphate [TPP], and tricresyl phosphate [TMPP]) were detected in WW but not in biosolids. Total PFRs ranged from 120 µg/kg to 900 µg/kg. Tris (2-chloroisopropyl) phosphate (TCIPP) is the most prominent with > 370 µg/kg due to its dominant concentration in the influent and tris (2-ethylhexyl) phosphate (TEHP) > 114 µg/kg, which is less prominent in influent but has high adsorption ability to biosolids, while others ranged from 0.8 to 44.2 µg/kg [124].

#### 2.1.11. Dioxin

de Sena et al. [51] measured various dioxin levels (at pg/g levels) in the meat processing industry biosolids. They found that octachlorodibenzodioxin (OCDD) and octachlorodibenzofuran (OCDF) were of the highest levels, i.e., 23.1 ng/kg and 10.2 ng/kg, respectively. OCDD and OCDF are the highest occurrence compounds, contributing more than 62% and 27% of PCDD and PCDF, respectively. Other congeners of PCDDs and PCDFs had lower concentrations ranging from 0.4 to 5.3 ng/kg. Koyuncu [95] showed a total PCDD/F of 0.44 ng toxic equivalent (TE)/kg of biosolids in anaerobic digestion. Andrade et al. [10] reported higher concentrations of 2,3,7,8-TCDF and 2,3,7,8 TCDD of 32 and 16 ng/kg, respectively, in biosolids of exclusively urban origin. Differences in the occurrence of dioxin compounds in biosolids can be explained by their WW sources. Dioxin can be generated in greater amounts by industrial processes such as pulp and papers, chemical and pesticide manufacturing.

### 2.2. Temporal distribution of OCs

The temporal trends of OCs in biosolids are driven by a range of factors, such as country-specific regulations on OC manufacturing or site-specific wastewater operating conditions (incoming WW amounts, conditions, and working conditions of technology treatment devices, etc.) of relevant WWTPs, local meteorological conditions (for labile compounds), and the extent of use of specific chemicals by the human population. From 1950 to 2016, the level of OCs in biosolids was found to rise over time due to an increase in OC usage with global economic expansion and population growth [148]. For example, microplastic concentrations in biosolids correlated with plastic production, which increased faster over 1950 – 2016 [148]. Some microplastics (PVC, PE, PET) were only quantified in biosolids from 1990 s onward [148]. The consumption rate of PFR from North America increased from 14,000

tons per year in 1986, to 38,000 tons per year in 2015 – an annual increase of 5.9% per year; while in China, PFR consumption was 70,000 tons per year in 2007 with an annual rate of increase of 15% [124]. Though China is not representative for Asia as a whole, it can be seen that the usage rate of OCs in each continent or countries will increase at different rates. Albero et al. [8] reported that levels of methylparaben tend to remain steady while propylparaben concentrations in biosolids slightly increased during a four-year sampling period.

Under the Stockholm Convention, PBDEs (one class of BFRs) were listed as persistent organic pollutants (POPs) and were phased out or restricted in the U.S., Canada, and Europe [201,205,79]. High production and consumption of PBDE caused an increasing concentration of biosolids from 1975. It remained stable from 2000 through 2009 due to the government phase-out from December 2004 in the U.S. Hale et al., [71]. However, DBDE levels (alternative flame retardants and a group of PBDEs) increased steadily from 1995 to 2008 due to rising use rates [71]. DBDE continues to be produced and used (for example, in television casings), increasing its occurrence in biosolids. In conjunction with phasing out of PBDE in 2004, the total concentration of 11 PBDEs was found to decrease from 7600 µg/kg in 2001 to 4100 µg/kg in 2006 [213]. This indicates that on-going efforts to phase out PBDEs is resulting in a reduction of their concentrations in biosolids over time.

In Australia, the national load of PFAS in biosolids was found to increase from 2016 to 2018 [62], due to the increase of population, usage of PFAS-laden products and the mass of biosolids generated (300,000 dry tonnes in 2010 to 327,000 dry tonnes in 2017). The increasing trend in Australia matches well with worldwide trends as countries upgrade WWTP infrastructure to capture more of the population and thereby increase biosolid generation. Since environmental and health concerns surrounding exposure and environmental contamination are increasing for some PFASs such as PFOS, PFHxS and PFOA, there have been recent shifts in regulatory action. In 2023, the PFAS National Environmental Management Plan (NEMP - version 3.0, 2023) has restricted the concentration of PFAS in biosolids to a maximum of 31 mg PFOS + PFHxS/kg, and 130 mg PFOA/kg [76]. The 2023 plan also establishes a baseline to prevent the spreading of PFAS through land application of biosolid. From 2016, NSW, Queensland, and South Australia also banned the use of PFAS in firefighting foam.

Annex A and annex B of the Stockholm Convention were amended to include PFHxS & PFOA, and PFOS, respectively in 2019 in conjunction with the phase-out of long-chain PFASs [82]. Despite those efforts, more short-chain PFASs have been produced after 2002, with research suggesting that only 30% and 60% of biosolids samples have PFBA and PFBS detections, respectively, during 2012 – 2017, which is likely due to lower sorption potential to the biosolids [104]. It is expected that the level of legacy PFAS in biosolids would reduce progressively, but few confirmation studies have been conducted. However, for some OCs, such as PCBs (listed as POPs), they have also been phased out of production and discharge in many countries in the 1970 s and 1980 s, but their continued detection in biosolids was still a concern [89]. This is mainly due to the persistence, bioaccumulation, and unintentional formation and release of PCBs from some sources (e.g., waste incinerators, cement kilns, metallurgical industry, and residential combustion) [202]. Similarly, BFRs are still found in biosolids even though they have been replaced by PFRs and usage essentially stopped in Australia in 2005 [61]. This indicates that persistent OCs (e.g., PFASs, PCBs, BFRs) will be likely to remain in the environment for several decades even after phasing out.

It is also interesting to know that reported concentrations of non-hydrophobic chemicals such as artificial sweeteners appear to be independent of WWTP operational parameters such as treatment capacity, population served, and hydraulic retention time [106]. This implies non-hydrophobic OCs may also continue to be found in wastewaters and biosolids over time.

Temporal variation in the occurrence of some combustion-related compounds (e.g., dioxins, PAHs) depends on emission sources such as

man-made combustion: waste, agricultural residue burning, and natural processes such as volcano eruption. Some dioxin compounds have been released during organochlorine manufacturing and paper bleaching with chlorine. A study showed that the concentrations for PCDD/Fs and dl-PCBs in December (after the hot season had started for more than one month) were in the same range as the concentrations in September (less heating activities), indicating that dioxin deposition from the atmosphere to the sewer system did not affect dioxin levels in biosolids [241].

Seasonal variation analysis has been conducted for only some contaminants in biosolids. For example, Chen et al. [35] found that concentrations of four parabens in sludge samples were similar between the 2009 spring, 2009 summer, and the spring of 2010 but increased during the fall and winter of 2009. Significant lower PPCPs concentrations in biosolids were observed in August compared to the February, May, and December periods [194]. High consumption rates of antibiotics, NSAIDs, and antilipidemic can be the reason for high occurrences in the cold seasons. Concentrations of total  $\Sigma$ PCB<sub>7</sub> in sludge were found the highest in July, and the lowest in March. PCB levels were higher in warmer months than colder months, possibly due to precipitation's dilution effect [89]. High levels of PFRs were recorded in autumn (September–November). In contrast, lower levels were found in spring (March to May), which may be caused by the dilution effect of infiltration of precipitation found in April and May within the study area [124].

### 2.3. Spatial distribution of OCs

Most studies focussed on the occurrence of contaminants in the Americas, Europe and Asia-Pacific (also including Oceania and Africa). It can be seen from Fig. 4 that total dioxin concentrations in biosolids were lowest in the Americas, where ng/kg levels were found, while high levels of dioxin were detected in biosolids, especially in Europe (less than  $\mu$ g/kg levels) and Asia. In China, dioxin levels in biosolids ranged from 14.3 to 33.2 ng/kg [241], while in Brazil, dioxin levels ranged from 0.4 to 23.1 ng/kg in biosolids from meat processing industries [51]. Varied dioxin concentrations in biosolids could be explained by their provenance through incineration processes such as waste incineration, trash burning, open burning and natural processes, such as forest fires and volcanoes [143].

For microplastic compounds, studies in Australia reported higher occurrences in biosolids than studies in the Americas and Europe which have similar levels (Müller et al., 2020; [148]; Tian et al., 2022; [240]). The difference among continents could originate from different

population-scale production and consumption, urbanization rate, consumer perception, and specific industrial WW sources for individual WWTP, which need to be examined in further studies. For example, in terms of mass load, Okoffo et al. [149] projected that Australia releases a lower annual mass of MPs in biosolid (3700 Mt/year), while China, Europe, USA and Canada emitted 14000 to 80000 Mt/year, 26000 to 150000 Mt/year, 21000 to 122000 Mt/year, and 1500 to 9000 Mt/year, respectively. The reason for this difference is that the Australia population (26 million people) is much lower than the others hence the total mass of MPs in biosolids are compromised. A lack of a fundamental understanding of the extent and impacts of plastics leakage and critical pathways for their entry into the environment may hinder comprehensive assessments of microplastics in biosolids [148].

The occurrence of BFRs in biosolids was higher in Asia and the Americas and lower in Europe. While PFRs, which accounted for about 20% of the flame retardant consumption in 2006 in Europe, are proposed as alternatives for BFRs since the ban on some BFRs in this continent [210]. In Asia, occurrences of PFRs were still lower than the other continents; however, BFRs in biosolids were higher, meaning that the consumption/use of BFR compounds is still prominent in some Asian countries.

Similar to dioxins, PAHs concentrations in biosolids were lowest in the Americas, i.e., from meat processing industries [51]. Whereas similar concentrations of PAHs levels in biosolids were found for other continents (Asia-Pacific, Europe, Africa), [131,150,173,2242,95].

PCBs were 1 to 2 logs higher in the Americas than Europe or Asia [131,69,89,94]. For pharmaceutical products, azole biocides, UV filters and stabilizers found in biosolids, the intensive uses reflect the routine usage by people, especially in the household, where these chemicals were in higher concentration in biosolids in the Americas and Europe than in the Asia-Pacific region.

For the remaining contaminants, the purposed use and production by households and industrial activities mainly impact the occurrence of chemical compounds in biosolids. For example, the highest occurrence of PFASs was found in Europe, followed by Asia-Pacific and America. Some PFAS compounds (e.g., PFOS and PFOA) have been voluntarily removed from products manufactured in the US [144]. The levels of total PFAS reported varied by country and was largely dependent on the number of compounds monitored. For example, 34 perfluoroalkyl acids (PFAAs) were monitor in Australia [116] while 22 PFASs including 13 perfluoroalkyl carboxylic acid (PFCAs) homologues, 5 perfluorosulfonic acid (PFSAs) homologues, and 4 perfluoroalkyl sulfonamides (FASAs)

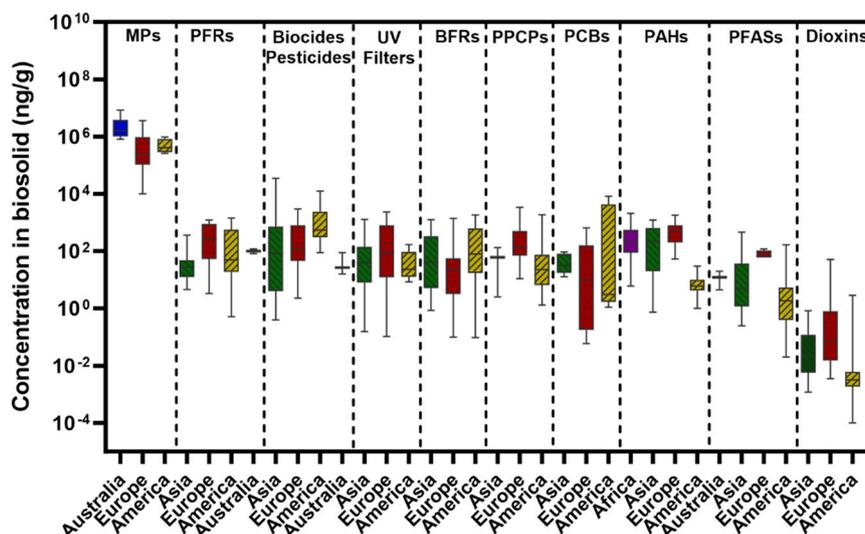


Fig. 4. The spatial distribution of MPs and the 10 OCs groups in biosolids (where data are available). Data of LAS group is not present as it is specific in Europe only. Data are presented as box plots (box: 25–75% interquartile range; bar: median; whisker: Tukey). The x-axis represents the compounds of contaminants groups. The acceptable representativeness of the data is ensured by including a wide range of countries in the continents as much as possible.

homologues were studied in Canada [104]. High PFAS levels were found in biosolids monitored from WW of industrial facilities, i.e., fluorochemical facilities in China [177], and electronic and semiconductor industries in China [108].

The occurrence data of MPs and the 10 OC groups indicate a substantial presence of a wide range of organic chemicals in biosolids at a wide margin of concentration (7 to 8 logs different). Although several OC groups have been regulated for a while, they are still present in significant quantities in biosolids (e.g., LASs, BFRs, biocides) which is of concern. It indicates further action is required to reduce the concentration of those OCs to an appreciated level. Europe shows a higher occurrence of those OCs in biosolids than other continents. It implies that the usage rate of Europeans is higher than others, probably attributable to their better social-economic profile. The occurrence of microplastic is exceptionally high than other OCs as there seem to be no regulations for microplastic in biosolids yet. It is also noticeable that the occurrence of microplastic in Australia is much higher than in other continents, potentially implying a higher microplastic consumption rate, which may reflect the differences in detection methods and estimates [54,232]. However, OCs in biosolids of Asia might not entirely reflect the actual situation as data set from Asian countries are limited, and predominantly come from middle to high-income countries like Japan, Korea, Taiwan (Republic of China), and China. Occurrence levels of OCs depend on several factors: current and historical usage, legislation, and other sets of geological, sociodemographic factors. Further data and discussion on the effect of those factors are provided in the next section.

The difference among continents could originate from different population-scale production and consumption, urbanization rate, consumer perception, and specific industrial WW sources for individual WWTP, which need to be examined in further studies. For example, antibiotic such as chloramphenicol appeared less in WWTPs of Europe and developed countries because it has been banned, while it was found substantial in China and developing countries [199]. The reason is accessibility to chemicals and PPCPs in developing countries is easy over the counter; while in developed countries, the prescription would be required for a list of chemical and PPCPs. In turn, chemical like gemfibrozil used for obesity is much higher in Europe and the Americas than Asia, reflecting the social issue pertaining to a high percentage of obesity in Europe and the Americas [199]. The trend of PPCPs is the mix up of several PPCPs hence does not totally reflect the issue accurately (Fig. 4). Our collected data agree with Tran et al. [199] that the Americas seems to have a high level of OCs, while Asia has a lower OCs in biosolid than other continent. Data of Europe and American might reflect the reality more accurately since all wastewater and chemical are collected and managed properly, so the illegal discharge is minimum. In developing countries, part of wastewater and chemicals are illegal discharged, hence the level of OCs in biosolids might be less than the real value.

## 2.4. Fate of organic contaminants in biosolids

Some intrinsic factors can cause the variation of OC concentrations in biosolids: (i) chemical characteristics of OCs, (ii) operation conditions of WWTPs, and (iii) sociodemographic factors.

### 2.4.1. Effects of OC characteristics on their occurrence

Many studies have used the solid–water distribution coefficient ( $K_d$ ) as one of the most popular parameters determining the affinity of OCs to biosolids. As shown in Fig. S2,  $\log(K_d)$  values of PPCPs, UV filters, biocides, PFASs, and PAHs were reported within the range of 0.3 to 4.3, 3.5 to 5.0, 1.1 to 4.4, 0.8 to 6.7 and 1.2 to 2.6, respectively. The BFR group has high  $\log K_{ow}$  and a higher  $\log K_d$  values than the others, corresponding to their greater sorption-hydrophobicity relationship [216]. Specifically, the  $\log(K_d)$  values for the BFRs are between 4.5 to 25 times greater than that of the other groups ( $P < 0.05$ ), and suggest a high potential for these compounds, especially the BFRs group, to accumulate in the biosolids. However, it was indicated that sample

homogeneity during quantification processes poses challenges for determining occurrence levels of contaminants in biosolids among studies; hence, it generally requires a large number of samples for analysis or an adequately representative sampling or sample-pooling procedure [52].

In selected OC groups where literature data are available, we observed significant correlations between  $\log K_d$  and molecular weights of chemicals (Fig. 5). The correlations show that UV filter group shows the highest correlation of adsorption tendency ( $\log K_d$ ) and molecular weights ( $R^2 = 0.95$ ), followed by BFRs ( $R^2 = 0.91$ ), PAHs ( $R^2 = 0.90$ ) and PFASs ( $R^2 = 0.88$ ). This indicates that the high tendency of chemicals adsorb to biosolids ( $\log K_d$ ) is well described by the corresponding to molecular weight of the OCs. While comparing the slope of OC groups, the PAHs and biocides present the strongest sorption tendency to biosolids (slope = 0.0108 to 0.011), followed by PFASs (slope = 0.009), UV filter group (slope = 0.002), and BFRs (slope = 0.001). The biocide and PPCP groups had the lowest propensity to adsorb to biosolids; however, the PPCP and biocide compounds in this review were probably more hydrophilic (e.g., chlorine, quaternary ammonium compounds) amongst a wide range of PPCPs and biocides used in practice. PAHs, PFASs and BFRs have been well-known as hydrophobic chemicals. For example, PFASs have been reported to be substantial in biosolids due to their proteinophilic nature [61]. PAHs are structured by a non-polar C (aromatic ring), while BFRs comprise non-polar C and bromide atoms, resulting in a significant hydrophobic characteristic.

### 2.4.2. Effects of WWTPs operational factors

Aside from specific WWTP factors e.g., wastewater parameters such as temperature, hydraulic retention time (HRT), solid retention time (SRT) and pH can affect the degradation, sorption mechanisms, and fate of the contaminants in biosolids (Table 1).

**2.4.2.1. pH.** pH is critical for determining the activities of microorganisms, as well as solubility, ionization state and hydrophobicity of OCs and biosolids, and hence related to the fate of OCs in biosolids [176]. The sewage pH can also influence how OCs accumulate in biosolids, by affecting the hydrophobicity, which can also be estimated using the octanol–water distribution coefficient ( $\log D$ ) [195]. For instance, by increasing pH, the  $\log D$  (effective hydrophobicity) of ionisable compounds such as ibuprofen, diclofenac, and ketoprofen can decrease, resulting in reduced sorption ability as well as reduced removal efficiencies of the compounds [195]. At pH between 6 and 8, OCs such as atenolol, azithromycin, erythromycin, clarithromycin, clindamycin, and tylosin are preferentially presented in cation forms enabling easier sorption on biosolids biomass which have negative charge [200]. In weakly acidic conditions (pH range from 5 to 6), the acidic pharmaceutical compounds (e.g., ibuprofen and ketoprofen) were observed to be efficiently degraded. In contrast, the degradation of other neutral PPCPs (e.g., propyphenazone and carbamazepine) was not affected by the pH of wastewater [200].

**2.4.2.2. Temperature.** In addition to pH, temperature directly affects microbial activities and the sorption equilibrium, impacting the fate of OCs in biosolid. Some studies reported increasing OCs degradations at increasing temperatures due to increased microbial activities [128]. For instance, in a lagoon system, degradation of PPCPs degradations at 20 °C was greater than degradation at 4.4 °C [115]. Other studies have also reported that biodegradation of other OCs was dependent on temperature in an activated sludge system and membrane bioreactor (MBR) [102,192]. On the other hand, sorption mechanisms also vary based on temperature. The removal of most hydrophobic compounds by the sorption mechanisms was significantly enhanced under temperatures between 10 to 35 °C [70,214]. However, at high temperatures, such as 45 °C, the removal of OCs decreased [70]. As most removal mechanisms of the hydrophobic OCs are driven by sorption onto the

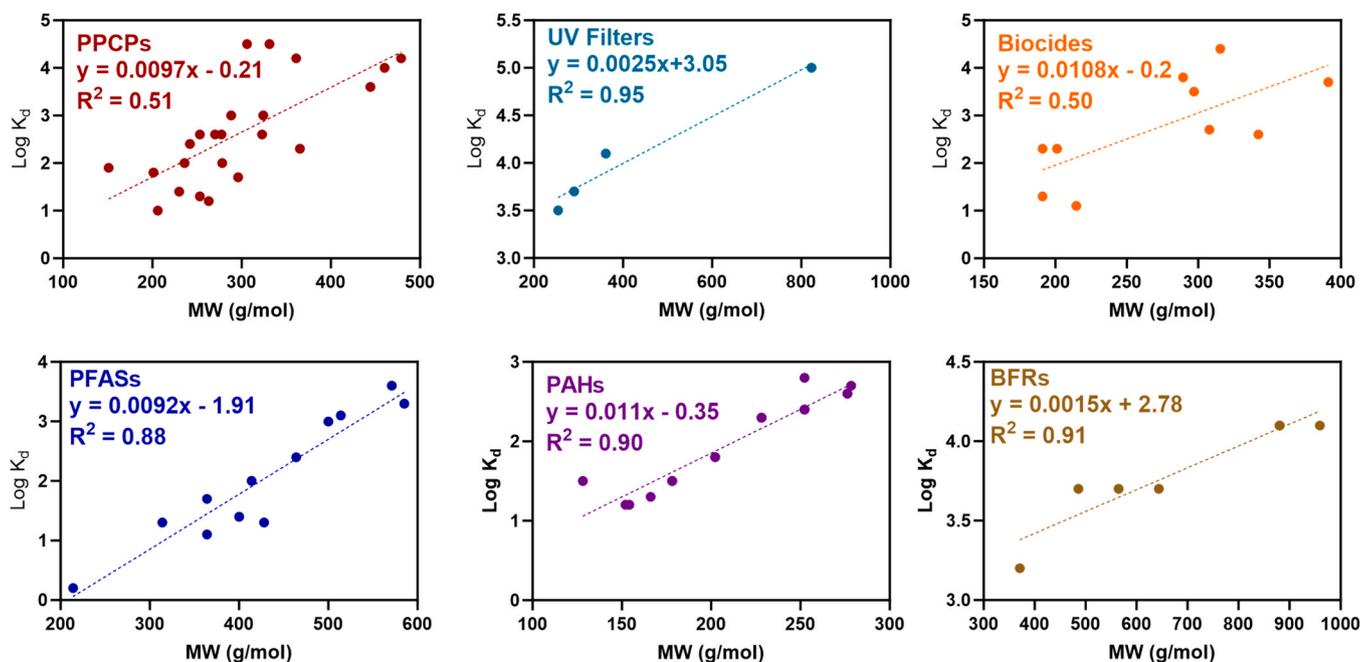


Fig. 5. Correlation of solid–water partitioning coefficient ( $\log K_d$ ) and molecules weight of OCs (where literature available). The x axis represents the molecules weight chemical groups. The y axis represents the  $\log K_d$  of the chemical groups. Arvaniti et al., [101,117,119,14,142,180,191,200,216,224,249,26,58,65,91,98].

biosolids, reduced equilibrium sorption can occur when thermophilic temperatures are reached in the reactor [70].

**2.4.2.3. HRT and SRT.** In conventional WWTPs, HRT's longer than one day can play an essential role in the biodegradation of OCs as the reaction time is critical for microbial activity to degrade them [56], while the sorption of the OCs onto the biosolids requires a shorter HRT [160]. In an MBR system, the degradation efficiencies of PPCPs (e.g., gemfibrozil and trimethoprim) in wastewater can be improved by increasing HRT from 3 to 6 h. Those OCs were well adsorbed onto the biomass surfaces at the HRT of 6 h [160]. For typical WWTPs i.e., activated sludge systems, removing some antibiotics such as clindamycin and ciprofloxacin has also been found to be dependent on the HRT [134]. Furthermore, Kim et al. [92] reported that in the activated sludge process, tetracycline degradation efficiencies significantly increased to 86% with the HRT of 24 h, respectively, corresponding to the sorption kinetics reaching equilibrium within 24 h [92].

SRT is another operating condition that influences the fate of OCs in biosolid. Some previous studies conclude that SRT mainly affects OCs with high  $\log K_d$  and low biodegradation rate (mostly hydrophobic compounds) [187,43,81]. Although the OCs were mainly eliminated by biodegradation, some PPCPs and biocides such as carbamazepine, verapamil, and triclocarban tended to be increasingly adsorbed and accumulated onto the biosolids with the increasing SRT (Yang et al., 2016). With SRT of 3 d, the OCs, namely *n*-nonylphenol (4-*n*-NP), triclosan (TCS) and bisphenol A (BPA), were observed to have higher values of the sorption coefficients than those compounds with the SRTs of 10d and 20 d. Probably, at 3 d SRT, the activated sludge is greatly effective and may increase extracellular polymeric substance (EPS) production and consequently enhance sludge hydrophobicity and sorption affinity of the OCs [187]. Meanwhile, the increasing biotransformation rate for all the OCs were also observed at 3 d SRT. This indicates that microorganisms appear to be more active at younger sludge ages. At 20d SRT, the proportion of heterotrophic biomass per dry sludge matter was double than that of 3d SRT [187].

Accordingly, the methods used to treat biosolids (e.g., biological, thermal, advanced oxidation process) can also influence OC levels [13, 33]. Technologies like thermal and advanced oxidation process are the

most effective for reducing OCs, however, are usually more costly to build and operate. In turn, biological treatment is more cost-effective but time-consuming. The concentrations of OCs also depend on treatment capacity, where concentrations tended to be higher in low- and medium-capacity WWTPs (1 to 20 million gallons per day) [240]. Further discussion regarding the influence of treatment technologies on the occurrence and fate of OCs are provided in detail in Section 3.

#### 2.4.3. Effects of socio-demographic factors (seasonal use/consumption, occasional events, daily activities)

Several socio-demographic factors directly affect OCs in influent wastewater, resulting in their presence in biosolids from the WWTP. Seasonality, daily activities, culture, and occasional events, among others, can contribute to this variability of OCs in municipal wastewater and the resulting biosolids.

**2.4.3.1. Seasonal conditions.** Some OCs have seasonal uses resulting in their influent loading rate being varied throughout the year. For example, in the UK, antihistamines utilized to treat allergies peaked from May to August when pollen production is greatest [157]. The India's municipal wastewater treatment plants observed the highest antibiotic concentrations, including tetracycline, erythromycin, and ciprofloxacin [181]. The levels of antibiotics in influent wastewater exhibited highest concentrations during both the summer (57.8 ng/L) and winter (54.7 ng/L) seasons [181]. It was anticipated that antibiotics are typically used to treat respiratory infections, which are more common during the winter months [181]. Seasonality affected ciprofloxacin and norfloxacin concentrations in receiving wastewater for a WWTP in Switzerland over one year (Coutu et al., 2013). In the winter and spring seasons, there was an increase in the PPCPs concentrations, which could be attributed to the seasonal need for medical treatment (Coutu et al., 2013). For instance, in these seasons, ciprofloxacin was employed in the management of respiratory infections of the throat, nasal passages, and ears (Coutu et al., 2013). A comparison study of a full-scale MBR system between summer and winter showed an increase in biodegradation of 17 $\beta$ -estradiol, estrone, bisphenol A and triclosan during summer (Trinh et al., 2016). The underlying reason is possibly attributable to the elevated temperature that has conditioned biodegradation activity and

**Table 1**  
Effect of WWTP operational parameters on fate of OCs in biosolid.

Influential operating conditions	Targeted contaminants	Treatment process in WWTPs	Degradation efficiency (%)	Refs.
pH: 5-9	PPCPs – Carbamazepine, sulfamethoxazole, ibuprofen, diclofenac, ketoprofen	MBR	0.2 - 97	[1]
Temp: 10-45 °C	PPCPs - Ibuprofen, acetaminophen, naproxen, ketoprofen, diclofenac, primidone, carbamazepine, salicylic acid, metronidazole, gemfibrozil, triclosan Pesticides – fenoprop, pentachlorophenol Surfactants – 4-tert-butylphenol, 4-tert-octylphenol, 4-n-nonylphenol	MBR	20 - 99	[2]
Temp: 35-55 °C	PPCPs - Estrone, 17β-estradiol, estriol, sulfate conjugate of estrone LAS and surfactants - Nonylphenols, nonylphenol polyethoxylates, and nonylphenol ethoxycarboxylates	Anaerobic digestion	17 - 96	[3]
Temp: 4.4-20 °C	PPCPs - Caffeine, carbamazepine, diphenhydramine, erythromycin, fluoxetine, gemfibrozil, ibuprofen, naproxen, triclocarban, triclosan, trimethoprim, sulfamethazine, and sulfamethoxazole, 17α-estradiol-3-sulfate, 17β-estradiol-3-sulfate, estrone-3-sulfate, estriol, 17α-estradiol, 17β-estradiol, estrone, and ethynylestradiol	Aerated lagoons	60 - 99	[4]
HRT: 3-6 d	PPCPs – Diclofenac, furosemide, ibuprofen, naproxen, ramipril, atenolol, caffeine, carbamazepine, ketoprofen, paracetamol, estrone, 17β-estradiol, 17α-estradiol	Fixed bed reactor	52 - 62	[5]
HRT: 2-4 d	PPCPs – Diclofenac, furosemide, ibuprofen, naproxen, ramipril, atenolol, caffeine, carbamazepine, ketoprofen, paracetamol, estrone, 17β-estradiol, 17α-estradiol	Trickling filter	30 - 46	[5]
HRT: 3-6 d	PPCPs – Diclofenac, furosemide, ibuprofen, naproxen, ramipril, atenolol,	Fluidized bed biological reactor	40 - 56	[5]

**Table 1 (continued)**

Influential operating conditions	Targeted contaminants	Treatment process in WWTPs	Degradation efficiency (%)	Refs.
	caffeine, carbamazepine, ketoprofen, paracetamol, estrone, 17β-estradiol, 17α-estradiol			
HRT: 7 - 24 h SRT: 3 - 10 d	PPCPs - Tetracycline	Activated sludge processes	78 - 86	[6]
HRT: 3 - 6 h	PPCPs - Trimethoprim and gemfibrozil	MBR	10 - 90	[7]
HRT: 12 - 24 h	PPCPs - Bisphenol A, 2,6-di-tert-butylphenol, di-butylphthalate, di-(ethylhexyl)-phthalate, carbamazepine, diclofenac, N,N-diethylmeta-toluamide	Two-stage AS	55 - 81	[8]
HRT: 1 - 4 d	PPCPs - Ketoprofen, ibuprofen, galaxolide, tonalide, triclosan, diclofenac, terbutrin, and oxybenzone; naproxen, cashmeran, methyl dihydrojasmonate and caffeine, diazinone, tributyl fosfate, carbamazepine, and tris(2-chloroethyl) phosphate)	Biological filtration processes	10 - 99	[9]
HRT: 2 - 5 d	PPCPs - Ketoprofen, naproxen, ibuprofen, diclofenac, salicylic acid, carbamazepine, caffeine, galaxolide, tonalide and methyl dihydrojasmonate	Constructed wetlands	22 - 99	[10]
pH: 4.4 - 7.2	PPCPs - Estradiol, estrone, -ethynilestradiol, bisphenol A, benzophenone, clofibrac acid, gemfibrozil, ibuprofen, fenoprofen, ketoprofen, naproxen, diclofenac, indomethacin, propyphenazone, and carbamazepine,	Activated sludge	Directly effect on solid-water partitioning coefficient (K <sub>d</sub> )	[11]
SRT: 3 - 20 d	PPCPs - 4-n-nonylphenol, triclosan, and bisphenol A	Activated sludge	90 - 99	[12]
Hydraulic Loading Rate (HLR): 13 - 160 L/m <sup>2</sup> /d	PPCPs - Caffeine, salicylic acid, methyl dihydrojasmonate, CA-ibuprofen, hydrocinnamic acid, oxybenzone, ibuprofen, OH-ibuprofen, naproxen, diclofenac, galaxolide, and tonalide, carbamazepine	Constructed wetland	20 - 99	[13]

Reference: [1] Tadkaew et al. [195], [2] Hai et al. [70], [3] Paterakis et al. [155], [4] Li et al. [115], [5] Ejhed et al. [56], [6] Kim et al. [92], [7] Prasertkulsak and Chiemchaisri [160], [8] Boonnorat et al. [24], [9] Matamoros

et al. [136], [10] Hijosa-Valsero et al. [78], [11] Urase and Kikuta [203], [12] Stasinakis et al. [187], [13] Matamoros et al. [135].

reduced sorption of chemicals to biomass.

**2.4.3.2. Occasional events.** There is limited data on the impact of occasional events on the loading of OCs in wastewater. Increased drug use can occur during music festivals and periods of increased tourism or public holidays/vacations, resulting in increased measured concentrations in wastewater [19,130]. Pandemics such as influenza and COVID-19 are another scenario resulting in a relatively increased load. For example, during a period of increased consumption of PPCPs for controlling and preventing COVID-19, antiviral drugs such as ribavirin, and antibiotics such as moxifloxacin hydrochloride were found to rise sharply in the WWTPs in China [37]. Because there is limited information for managing WWTPs to maintain expected performance in such an event, a greater proportion of PPCPs could accumulate onto the biosolids, disrupting biosolids useability [157].

**2.4.3.3. Diurnal patterns.** There is a diurnal variation of some OCs concentrations in wastewater (Coutu et al., 2013). In addition, differences in weekday/weekend flow can be specific to the social demographic pattern of the catchments. For example, a residential (commuter) or tourist area may have increased flow during weekends [157]. Intra-day dynamics of PPCPs use (e.g. triclosan, caffeine) were reportedly the cause of fluctuations of PPCPs concentrations in influent and hence in the biosolids [112]. Some PPCPs were removed by sorption onto the biosolids [112].

Overall, OCs characteristics play an important role in their adsorption capacity to biosolids. Two important ones are OCs hydrophobicity and molecular weight, given the OCs with higher hydrophobicity tend to have greater affinity to biosolids. Organic matter is a rich component in biosolids in which also hydrophobic, making them more likely to adsorb hydrophobic OCs. Additionally, larger molecules or higher molecular weight OCs tend to be more readily absorbed by biosolids due to their increased mass and size, which allows for greater interactions with biosolids particles.

Operational conditions of WWTPs such as pH, temperature, and HRT and SRT are also critical to the sorption of OCs to biosolids. pH of wastewater and biosolids impacts ionization state of both OCs and other organic matter in biosolids, then compromising the sorption of OCs. pH alters the charges on surface of biosolids hence change the interaction with OCs as a result. Temperature is also an influential factor for sorption of OCs onto biosolids. It influences the sorption rate as higher temperature often results in faster kinetics, which means that the equilibrium between OCs in the wastewater and those adsorbed to biosolids may be reached quicker. Hence, OCs could be greatly removed during the season with high temperature (summer). Together with pH and temperature, HRT is essentially important as an increasing HRT between biosolids, and wastewater allows more contact time for the contaminants to interact with biosolids. However, there may be diminishing returns with excessively long contact times because adsorption sites on biosolids become saturated.

Socio-demographic factors (seasonal use/consumption, occasional events, daily activities) could have effects on the influent OCs concentrations of wastewater. The initial concentration of OCs in wastewater could influence sorption of OCs onto biosolids. However, the higher concentrations of the OCs in the wastewater may require longer contact times to achieve effective sorption.

### 3. Treatment technologies for OCs in biosolids

The treatment technologies for OCs in biosolids comprise four main groups to be discussed herein: biological, thermal, advanced oxidation process, and other physical/chemical technologies. In this section, the

treatment technologies focus on emerging chemicals such as PFASs, and PPCPs, with a less attention to biocides, BFRs, UV filters and stabilization, and PFRs. Although MPs and LAS is a highly prevalent OCs in biosolids, this study do not focus on potential treatment technologies for these compounds. Similarly, PCBs, PAHs, and dioxins have been widely studied hence they are not included in this section.

#### 3.1. Biological treatment

Biological treatment is widely used to stabilize biosolids from wastewater treatment plants. The most common biological sludge stabilization processes include aerobic and anaerobic digestion. The purpose of sludge stabilization processes is to reduce the bulk volume of biosolids, eliminate pathogens, and minimize odour. After digestion, the biosolids often further undergo composting to reduce the load of OCs to a regulated level.

##### 3.1.1. Aerobic digestion

Aerobic digestion is used when degradation of OCs in biosolids occurs in the presence of molecular oxygen. This process is often preferred for smaller WWTPs to reduce capital costs [68]. Moreover, effectiveness in degradation of OCs, increased degradation efficiencies, and social acceptability are raised as advantages of aerobic digestion [208].

SRT and temperature are two of the most influential factors on the OC degradation efficiency of aerobic digestion. Aerobic digesters are often operated at retention times beyond 15 d [85]. A retention time of less than 6 d limits degradation of organic matter [121]. Half-life measurements of TCB and TCS showed that 4 to 46 d of aerobic digestion is essential to reach degradation efficiencies of more than 90%. The upper end of this timeframe is likely not practical in conventional treatment systems [147]. Regarding temperature, a study was conducted to understand the effect of variation in temperature during both warm and cold seasons. It was found that degradation efficiencies of OCs (e.g., PPCPs, PCMs, and PBDEs) were improved at warmer temperatures due to increased microbial metabolic activity at higher temperatures. As a result, the efficiency of aerobic digestion for OC treatment can be improved by operating the digester under thermophilic conditions (55 to 70 °C) rather than mesophilic (i.e., ambient temperatures); however, the operational costs are also increased which need to be pondered for economic benefit. Autothermal thermophilic aerobic digestion is an advanced configuration of aerobic digestion that can provide a thermophilic condition without the requirement for an external heat source.

The post-anaerobic aerobic digestion process is an exemplary configuration, which can further reduce the sludge volume and micropollutants such as PPCPs. Six PPCP compounds (e.g., cotinine, DEET (N, N-diethyl-m-toluamide), hydroxycotinine, codeine, acesulfame, and ibuprofen) were removed effectively beyond 80% using this process [118]. The transformation of organic chemicals largely depends on what bacterial strain is involved in the biotransformation process, resulting in different transformation pathways. For example, the transformation of DEET depends on the redox condition, while ibuprofen transformation comprises hydroxylation, carboxylation, methylation, and ring-opening reactions.

Several pretreatment methods can improve digestion efficacy, such as chemo-mechanical [165], electrolysis [185,235], sonication [248], or enzymatic or microbial pretreatment [66,209]. After a 20-day aerobic digestion, Diethylhexyl phthalate (DEHP) degradation was 72%. However, when employing ultrasonic and Fenton methods, DEHP degradations increased to 89% and 85%, respectively [158]. Combining aerobic digestion with conventional activated sludge process can reduce waste sludge volume and shorten retention time to 5 to 15 d [85]. With regard to enzymatic pretreatment, laccase is known as a highly effective enzyme for biodegradation of various OCs [209].

Some chemicals, like triclosan, are preferable to a specific degradation condition, that is, aerobic process. In an activated sludge reactor, 75% of the triclosan was removed in an aerobic condition, whereas

degradations were very low in anoxic and anaerobic conditions [38]. Triclosan was also well degraded in an ammonia oxidation condition. For example, 36% to 59% of triclosan was oxidized by ammonia-oxidizing bacteria in 24 h [103]. Rapid degradation of NP2EO and PAs (such as phthalate esters and DEHP) also has been achieved under aerobic conditions [151,182,40]. Nevertheless, longer sludge retention time, higher operating cost, and no energy recovery are aerobic digestion's drawbacks compared to anaerobic digestion [1233].

### 3.1.2. Anaerobic digestion

Anaerobic digestion comprises four consecutive steps (e.g., hydrolysis, acidogenesis, acetogenesis, and methanogenesis) performed by different microorganisms. The advantages of this process include lower energy costs and a potential revenue stream from the production of biogas. Operation and maintenance can, however, be more cost-intensive than aerobic digestion.

The physicochemical properties of the target OCs, the temperature of the process, SRT, organic loading rate (OLR), and sludge inoculum are among the most important factors of anaerobic digestion to control the degradation of OCs. The mechanism of action is mostly based on variation in sorption, microbial population, and metabolism, which subsequently affect biodegradation capacities. A low OLR and high SRT provide a better cost-benefit value [75]. As a result, anaerobic digestion is normally operated at 15 to 30 d HRT [133].

There is a notable issue in the analysis of anaerobic biodegradation data that both dry matter and moisture of sludge decline during the process. As a result, higher or similar downward trends should be considered as an efficient degradation in anaerobic digestion [197]. Based on this definition, a wide range of degradation rates was reported for OCs in the anaerobic digestion. While biodegradation capacity of some compounds (such as HHCb or AHTN, TBBPA, PAEs, NPEOs, and phthalates) is limited in this process, some others (such as PHPs) have efficient degradation rates [132].

Bisphenol A, Nonylphenol and Nonylphenol monoethoxylate were moderately removed (40% to 80%) [172]. Samaras et al. [172] also studied PPCPs degradation efficiency in three lab-scale anaerobic digestion systems (e.g., a single-stage mesophilic, a single-stage thermophilic and a two-stage thermophilic/mesophilic) and found that degradation efficacy was not affected by the type of system. All reactors had a high degradation rate of diclofenac, ibuprofen, naproxen and ketoprofen (>80%). Huang et al. [80] have reported indications of PFOA biotransformation under anaerobic conditions by *Acidimicrobium* sp. strain A6, and even reported evidence of production of F<sup>-</sup> and shorter chain carboxylates (i.e., PFBA, PFPeA, PFHxA and PFHpA), however these claims remain controversial and have yet to be repeated by others (Liu et al., 2023). Similarly, Li et al. [107] have reported the biotransformation of 8:2 fluorotelomer alcohol to PFPeA and PFBA during anaerobic sludge digestion, indicating the concentration of PFBA and PFPeA could increase after anaerobic digestion, however the degradation or transformation of PFAS from biological processes such as those present in a WWTP or a sludge digestion process is an active and ongoing area of research.

Low efficiency, bioaccumulation and toxic metabolites are the main disadvantages of anaerobic digestion. In some scenarios, the persistency and potency of OCs by-products might be more than their parental OCs. In this case, it is vital to identify the influential factors, such as the physico-chemical properties of OCs, operational conditions, and the involved microbial compositions [64,212]. Moreover, pre- or post-treatment processes (such as advanced oxidation or bio-augmentation of sludge) are also practical to increase the degradation of OCs from biosolids.

Similar to aerobic digestion, there are also a few options for sludge pre-treatment to improve process efficiency of anaerobic digestion, including chemical [109,198], thermal [122,164,231], biological [5,6] and mechanical processes [21,114], or a combination of these techniques [164]. Most of the pre-treatment methods result in a substantial

increase of OC degradation through an increase in solubility and bioavailability of OCs in the biosolids. For instance, co-digestion with Fe<sub>3</sub>O<sub>4</sub> improves the degradation efficiency of tetracycline by 40%, given that Fe<sub>3</sub>O<sub>4</sub> can adsorb tetracycline and make it more available for microbes. Significant improvements were reported for the degradation of PPCPs by applying high temperature pre-treatment and complementary advanced technologies (such as Fenton oxidation, microfiltration and ultrafiltration) [212]. Reports showed that ultrasonication and thermal hydrolysis also increased the biodegradation efficiencies of di(2-ethylhexyl)phthalate and DEHP by 13% and 19% respectively [175, 212,40].

In a comparative study between anaerobic and aerobic digestion of OCs, Zhao et al. [247] reported that anaerobic digestion removed a wide range of chemicals (e.g., bezafibrate and fluoroquinolones, acetaminophen, atenolol, bezafibrate, carbamazepine, 17 $\alpha$ -ethinylestradiol), which was a greater number than aerobic digestion.

### 3.1.3. Composting

Composting is an aerobic stabilisation process in which sludge is mixed with different types of bulking agents (wood chips, green waste, and sawdust) to enhance biological degradation. This process is usually the last step before the land application of biosolids. Conventional composting can be divided into three main types: aerated static pile, windrow, and in-vessel.

Composting was superior to other sludge stabilization methods for higher degradation of total alkylphenol ethoxylates (APEOs). While anaerobic digestion resulted in 887 mg/kg (dry weight) of APEO, composting could achieve a sludge with far lower concentrations as low as 5 to 14 mg/kg [99]. There are some other contradicting trends in composting process of di(2-ethylhexyl) phthalate [175].

Composting also appeared to be an effective alternative to degrade surfactants. Keller et al. [90] found that estrogenic nonylphenol polyethoxylates (e.g., nonylphenol, nonylphenol mono-ethoxylate, nonylphenol di-ethoxylates) were significantly reduced by composting after two months. A higher degradation rate (over 75%) was attributed to the rising temperature (40–50 °C) because it benefited microbial diversity and activity. This highlights the significant role of temperature in composting. Thermophilic conditions could elevate the degradation of a wide range of OCs (BFRs, PFRs, PPCPs, etc.) [197]. Xia et al. [229] also investigated the impact of composting temperature on nonylphenol degradation. Nonylphenol reduction was greater at 65 °C (76%) than at 45 °C (41%) during the first 6 d of incubation during composting. However, after 15 d, the differences between degradations narrowed and were not significantly different between temperatures. Results indicated that high temperatures could enhance emerging contaminants degradation in biosolids over short periods as lower temperatures obtained similar results over more extended periods. These findings suggest that thermophilic microorganisms were more effective for nonionic surfactant degradation than mesophilic microorganisms during composting treatment [25].

OCs in biosolids may leach out during the composting process, resulting in potential ecotoxicological risks, and is still poorly investigated. Liu et al. [123] evaluated the impact of biochar and woodchips on the leachability of four PPCPs (e.g., carbamazepine, gemfibrozil, naproxen and ibuprofen) in biosolid. Results indicated that composting significantly decreased total and leachable concentrations of selective compounds in unamended and amended biosolids. Microbial and hydrophobic interactions were suggested as the synergistic degradation mechanism of selected compounds [140,175]. The positive effects of the amendment were observed for carbamazepine, gemfibrozil and naproxen, which was likely due to the functional groups of the amendment, improved air penetration and additional carbon sources for more active microbial degradation [45]. To accelerate the degradation of organic matter in biosolids during the composting process, reactive oxygen species (e.g., calcium peroxide, soluble Fe (II)) were mixed and shortened the maturity time by 25% [16,193]. Biosolids were mixed with

garbage enzymes to enhance enzyme activity and fungal and bacterial communities [83,84]. Hormones may pose some constraints in composting due to the transformation into another hormone or conversion from conjugate forms. A good example of this is some steroids, such as methyltestosterone, that are typically detected in post-composting residues [46,238].

### 3.1.4. Myco-remediation

Myco-remediation applies enzymatic machineries of fungi to efficiently degrade complex compounds. Unique capabilities of fungi have been extensively studied in metabolism of pharmaceuticals which was deemed responsible of their elevated reduction (specifically the fungus *T. versicolour* in the degradation of ranitidine, fenofibrate, atorvastatin, diclofenac, and hydrochlorothiazide) [168,169,4,53].

Myco-remediation using white-root fungi has been adapted for the treatment of OCs from its first application for lignin cellulose degradation [190]. The white-root fungi have been evaluated for the treatment of OCs such as PPCPs [126,170,87]. Fungi can be sourced from both land and marine environments. There is a study that introduces adaptive capabilities of marine fungi in the degradation of OCs. Specifically for OCs treatment, both fruiting body-forming and purely filamentous fungi can be applied. Some species that have been reported include *Aspergillus*, *Trichoderma*, *Penicillium*, and *Fusarium* [190]. There are two common enzyme types associated with fungi: (i) laccase of class phenol oxidases and peroxidase of class heme-containing peroxidases, and (ii) intracellular cytochrome P450 monooxygenase [87,189].

Several studies have shown fungi to be effective in the reduction of OCs. *Penicillium ostreatus* could completely degrade oxytetracycline up to 97% in 14 d. Despite their use for OC treatment, fungal degradation is rarely applied to sludge and biosolids.

The role of ligninolytic and non-ligninolytic enzymes has also been known for degradation of OCs. Oxidation of OCs using *Dentipellis* sp. KUC8613 highlighted the initial role of cytochrome P450 followed by a list of ligninolytic enzymes catalyze enzymes (such as glutathione transferase, dehydrogenases, and epoxide hydrolases) [7]. Biosolids indeed contain important beneficial enzymes excreted by microbes; hence using external enzyme sources is an add-on to accelerate the degradation of OCs. Another issue with using fungi is that the treatment time seems to be rather long (14 to 70 d) compared with other conventional biotreatment processes.

Most of the technologies in the biological treatment group (anaerobic, aerobic and composting) have been well developed so far and are now being used widely for the treatment of nutrients, odour, and pathogens, not just specifically on OCs [17,154]. Except for myco-remediation, this technique aims to boost the remediation of OCs and recently emerged as a promising one; however still at bench scale study.

## 3.2. Thermal treatment

Thermal technologies like incineration have attracted increasing interest in recent years for biosolids management due to the effective destruction of recalcitrant compounds such as PFASs. Most of the sewage sludge treatment plants in countries like Germany, Switzerland, Netherland are incinerated either with sole biosolid or in combination with other solid wastes [174]. Direct incineration has become more prevalent in China since it has a low energy deficit, investment and operation cost [74].

To date, thermal treatment is one of the most prevailing technologies for the treatment of OCs in biosolids due to the high temperatures produced, which favour the decomposition of organic molecules. Thermal technologies appear in various forms such as, but not limited to, pyrolysis, gasification, supercritical water oxidation, hydrothermal alkaline treatment, and thermal desorption oxidation [226,243,244,97]. Thermal treatment can produce secondary emissions (e.g., carbon monoxide, sulphur dioxide) and should be managed to comply with

emission regulations [174]. The main limitation of these technologies is the high-moisture content of biosolids that requires a lot of energy to dry. Although thermal treatment has been able to be coupled as pre-treatment before some other degradation processes in the degradation of OCs, the results were not always similar. While thermal hydrolysis pre-treatment of sludge before anaerobic digestion had an inhibitory effect on the degradation of nonylphenol ethoxylates [138], improvements were reported in the thermal pretreatment for aerobic digestion of hormones and methane generation [28,156].

### 3.2.1. Hydrothermal alkaline treatment

Hydrothermal alkaline treatment (HALT) is a subcritical water process that leverages an alkaline condition to break down OCs and is especially targeted to PFAS destruction. This technology is in early stages of development but has been used for the treatment of halogenated pollutants, such as trichloroethylene, polyvinyl chloride, PFAS [111,60,73]. The kinetics of HALT degradation are considerably faster than other destructive technologies (e.g., electrochemical oxidation).

Although HALT is effective for degradation of OCs, this technology is preferentially applied for the liquid media such as wastewater, hence HALT is not a favourable option for direct treatment of solid samples like biosolids.

### 3.2.2. Incineration, pyrolysis and gasification

Incineration, pyrolysis and gasification of biosolids can theoretically break down OCs entirely and form the relevant by-product vapours. The major differences between these three processes are operating temperature, by-products, and the presence of oxygen. Incineration and gasification are similar in principle (high-temperature operation with oxygen); however, the energy produced from incineration is high-temperature heat, while gasification produces combustible gas. In turn, pyrolysis operation does not require oxygen but inert gas to make it happen.

Incineration, including biosolid, is one of the most popular waste-to-energy technology for waste management. Incineration of biosolids has appeared at some pilot to full-scale level. For example, a biosolids incinerator was part of WWTP in Dunedin (New Zealand) [55]. In addition, over 200 of WWTPs in the USA have been using incineration for biosolids treatment [207]. Yet, there is no report about the OCs degradation for those cases. Alternatively, we can qualitatively estimate the effectiveness of incineration by comparing the working temperature. Most organic compounds have thermal destruction from 590 °C to 650 °C, while the working temperature of incineration plant is up to 850 °C. Theoretically, this technology can destroy most of OCs, except for PFASs with the mineralization temperature taking place at > 1000 °C. It can be concluded that incineration cannot remove all, but effectively remove most of OCs.

Gasification processes will break down the OCs in biosolids, including PFAS, and produce biochar for agricultural applications [162]. The pre-dried biosolids is gasified with air at 850 to 880 °C with a 30-min residence time. The gas product is driven through a cyclone and scrubber to separate the particles from dust, aerosol and synthesis gas. To what extent the OCs are degraded would depend on the operating conditions of the thermal process (e.g., applied temperature and residence time). A semi-pilot pyrolysis system was able to achieve greater than 90% PFAS degradation and yielded 36 to 45% biochar using temperatures 500 to 600 °C [97]. Slow pyrolysis at a heating rate of 0.1 to 1 °C/s produces maximum biochar compared to fast pyrolysis (100 °C/s), which generates a product with a higher bio-oil yield [39].

The question is raised which technology is better and in which scenario. Winchell et al. [225] compared these three technologies for the treatment of biosolids, the result is that pyrolysis and gasification offer more advantages over incineration in terms of carbon-rich solid products, less flue gas, and lower combustion air requirements. Although incineration is a mature technology that has been applied worldwide for its simpler design and higher capacity, the pyrolysis and gasification are

progressively showing their effectiveness in OCs destruction and lower emissions for future application.

### 3.2.3. Supercritical water oxidation

The SCWO technology is particularly well suited for destruction of OCs, particularly recalcitrant compounds like PFASs. SCWO is based on heating and pressurizing water above 374 °C and 22.1 MPa, respectively, and has been used for many years for destruction of recalcitrant organics. SCWO has high treatment rates (6 to 30 tons per day) and short residence times (6 to 8 s) [96], and an energy usage rate of  $1.1 \times 10^5$  kWh/kg PFAS, which is approximately on par with other destruction technologies (e.g., plasma, electrochemical oxidation, sonolysis). A few drawbacks of SCWO include the potential emission of volatile organofluoride by-products [110] and capital costs of up to \$2.5 to \$5.5 million. Several pilot plants and full scale treatment plants have been installed for the treatment of sewage sludge; however, some of the full scale treatment plants face technical difficulties such as corrosion. Therefore, feasibility of this technology for biosolids treatment in large scale remains uncertain.

### 3.2.4. Cement kiln incinerator

Cement kiln incineration has been used most commonly to treat soil and spent sorbent media. This technology is operated at temperatures of 1400 °C to 2000 °C, providing for the destruction of recalcitrant OCs. The clinker (an intermediary product) production capacities can be up to 3200 t/d, with coprocessing of biosolids at 100 t/d [129]. The typical gas and solid residence times are 10 s and 30 min, respectively, shorter than other thermal treatment technologies. However, a cement kiln requires an excess of air for better mixing and can be economically beneficial as a low-energy technique. Although cement kiln incineration technology is well developed, the application for co-processing with biosolids has some challenges in the gas-phase emission of PFASs, and although not an OC, heavy metals [129,183,215].

Good practice in controlling the number of harmful elements such as alkali-chlorine-sulfur in the feed, can reduce, or even eliminate the detrimental impacts of these constituents on the operational performance of the cement kiln [153]. Cement kiln incineration can result in the production of dioxins, however operation of a kiln at higher temperatures can help mitigate against their formation.

As mentioned above, although used for treatment of soils and spent sorbent media at scale, cement kiln incineration is less widely used for biosolids treatment. One example of biosolids treatment however is an Australian gasification treatment that has only been in operation since early 2022 at Loganholme, Queensland, in which can destroy more than 94% PFAS [162]. Although several trials and pilot systems have been commissioned in Germany, a reliable continuous operation of a full scale system is still not in practice [174]. As with many of the other technologies previously discussed, cement kiln treatment of biosolids is maturing from a technology for treatment of different matrices (e.g., soil) into application for biosolids [174].

## 3.3. Advanced oxidation processes

The principle of advanced oxidation processes for biosolids treatment is either to use its highly reactive oxygen species to decompose OCs directly in biosolids or as a pre-treatment step to indirectly decrease the OCs contents in biosolids. The advanced oxidation processes comprise of three main technologies: ozone, Fenton, and UV.

### 3.3.1. Ozone

Ozone has been studied widely as a pretreatment method to degrade OCs such as PPCPs, and antibiotics [113,163,20,228,27,50]. Most ozone applications appear to be applied as a pre-treatment step for anaerobic digestion, while the effectiveness of this pre-treatment technique is still being debated. With this technology some of the cells in the sludge are exposed to a sufficient ozone dose to be lysed, destroying the cell and

releasing intracellular substances. From a biology standpoint this lysing is incomplete, meaning most of microbial mass survives, however this breakdown in the cell structure can increase OC concentrations at first, but further oxidation of OCs upon exposure to ozone completes the degradation process [163]. For instance, it has been used to completely degrade levofloxacin in a biosolids [246].

Ozone can also be incorporated with other technologies for OC treatment. The hybrid system of ozone and electrooxidation was reported to remove 86% of bisphenol A, 68% of nonylphenol, and 67% of triclosan and leave no secondary pollutants following treatment [50].

The end-point target of ozone application in biosolids is however to increase the hydrolysis step by breaking down the non-biodegradable fractions. Using ozone for biosolid treatment have been shown feasible via techno-economic assessment. The amount of biosolid for disposal was reduced 50% to 75% which save 235 to 655\$ per day [41]. However, the application of ozone in full scale goes to the reduction of biosolid for disposal other than OCs treatment.

### 3.3.2. Fenton chemistry

The Fenton reaction is the Fe(III/II)-catalyzed oxidation of organic substrates by hydrogen peroxide, and is a powerful technique to support OC decontamination indirectly. Like ozone, pre-treatment of biosolids with Fenton chemistry improves the availability of OCs, reduces matrix effects of biosolids, and increases overall treatment performance. Although the performance of the Fenton process depends on the characteristics of the matrix, operating conditions and the occurrence of contaminants, the degradation efficiencies are typically high. For example, Fenton chemistry was reported to remove up to 70% of estrone, 90% of 17 $\beta$ -estradiol, 84% of 17 $\alpha$ -ethinylestradiol and 98% of estriol, and additionally increased soluble total organic carbon by 24 times compared with non-Fenton conditions [120]. In a hybrid treatment system with a membrane bioreactor, solar Fenton oxidation removed sulfamethoxazole and erythromycin to levels below detection limits, and 84% of clarithromycin at the original concentration 100  $\mu$ g/L after 120 min [88].

Application of Fenton chemistry improved DEHP degradation efficiency by 10% when using 0.01 mL H<sub>2</sub>O<sub>2</sub>/g suspended solids and a [H<sub>2</sub>O<sub>2</sub>]<sub>0</sub>: [Fe<sup>2+</sup>]<sub>0</sub> ratio of 150 [158]. Fenton reactions have also been used to decompose biosolid matrix for analytical quantification of microplastic (Maw et al., 2022). Following one hour treatment with 30% H<sub>2</sub>O<sub>2</sub> and 0.05 M Fe<sup>2+</sup>, microplastics such as high-density polyethylene (HDPE), low-density polyethylene (LDPE), polypropylene (PP), and polystyrene (PS) were retained in their original forms indicating the effectiveness of Fenton chemistry for the pre-treatment of biosolids for microplastics quantification.

Other pre-treatment methods, such as CaO<sub>2</sub>, alkaline thermal treatment, free ammonia, and free nitric acid, showed promising results on sludge dewaterability, hydrogen and methane production, and short-chain fatty acid degradation. However, there are no clear conclusions on how effective they are on OC remediation [217,219,221,222].

### 3.3.3. Ultraviolet

Unlike O<sub>3</sub> and Fenton chemistry processes, UV was not reported as a common technology for biosolids treatment. UV was reported for degradation of endocrine-disrupting compounds in waste-activated sludge [237]. It was recommended that UV should be used in conjunction with H<sub>2</sub>O<sub>2</sub> to maximize the degradation efficiency of OCs (45 to 197 times higher than UV only). At the optimal conditions, for example, pH = 3, UV wavelength = 253.7 nm, UV intensity = 0.069 mW/cm<sup>2</sup>, and H<sub>2</sub>O<sub>2</sub> dosage = 0.5 mol/L, the degradation efficiencies of those chemicals was reported to range from 67 to 97%.

### 3.3.4. Microwave-assisted persulfate oxidation

For microwave-assisted persulfate oxidation, the activation temperature and dose of persulfate are important factors. One study found that at 20 °C, only 5% of PFAS were removed in 72 h, while at temperatures

of 50 °C and 70 °C, the degradation efficiency increased to 28% and 42%, respectively [72]. This study also found that persulfate dosages above 0.2 g/g wet biosolids did not result in further improvement to PFAS treatment due to self-scavenging of persulfate at high doses [72]. Although this technology may have some merits, it requires treatment times on the order of several hours, resulting in relatively high energy consumption.

### 3.4. Other chemical and physical treatment technologies

Apart from the major treatment technologies mentioned above, it is worth noting that ultrasonic and microwave-assisted persulfate oxidation technologies are also attracting interest in biosolids treatment.

#### 3.4.1. Ultrasonic

Ultrasonic treatment has been researched for the treatment of PFASs in biosolids; however, results are not as promising as the degradation of other OCs. Ultrasonic treatment has been reported to actually increase measured PFAS concentrations in biosolids [245]. It is thought that ultrasonic treatment may disrupt the sludge floc and release sorbed PFASs when sonic treatment extend longer than 15 min [245]. Similarly, oxidative agents like permanganate were also reported to enhance the release of PFASs from biosolids without improving degradation of PFASs themselves [245]. For that reason, ultrasonic treatment may be a method for pretreating biosolids upstream of other treatment technologies by disrupting the physical, chemical and biological properties of the sludge [18,30,31]. OCs such as estrogens, and BFRs are degraded in biosolids following pre-treatment with ultrasonic energy. For example, pre-treatment using ultrasonic energy was found to reduce 20% of estrogen mass in the anaerobic digester [31]. The combination of ultrasonic pre-treatment and biodegradation was also found to work well for removing phthalic acid esters [30].

#### 3.4.2. Alkaline stabilization

Alkaline stabilization of biosolids requires alkaline materials (e.g., hydrated lime, quicklime, fly ash, lime and/or cement kiln dust) to adjust pH for microorganism development [42,230]. Due to the wide availability and relatively low cost of these alkaline materials, alkaline stabilization is often considered a cost-effective process for biosolids stabilization and can be retrofitted to an existing facility as a secondary or backup stabilization method.

There are few studies on the fate of OCs in biosolids driven by alkaline stabilization. PPCPs can be solubilized from biosolids through alkaline treatment to enhance the degradation efficiency of PPCPs. For example, 90.5% of acetaminophen was removed at pH 12.5 in 4 h by alkaline-treated biosolid [166]. In another study, Vaithyanathan et al. [209] compared four pre-treatment options (e.g., ultrasonic, alkaline, freeze-drying and enzymatic), finding alkaline pre-treatment was the most reliable method with the highest degradation of contaminant mass from the biosolid matrix (72 OCs with maximum degradation efficiencies of 63%).

Further investigation on the solubility of emerging OCs in biosolids after alkaline treatment is necessary since treated biosolids might contain other contaminants, increasing hazard potential. Overall, alkaline stabilization does support the treatment of OCs indirectly as AOPs via biosolid pretreatment step.

#### 3.4.3. Landfilling

Landfilling is commonly used where in-situ clean-up methods do not work quickly enough or are cost-ineffective. It is often the fastest way to deal with high levels of contamination that pose an immediate risk to human health or the environment. It can be cost-effective for small volumes of contaminated media and can be incorporated with composting and incineration as pre-treatment steps prior to landfilling. However, landfilling contaminated media like biosolids should include attention to considerations like regulatory restrictions and the potential

need to treat surface runoff and leachate. In Australia, 5% of biosolids produced goes into landfills, at the rate of approximately 350,000 tonnes annually [12]. However, the biggest challenges for landfilling biosolids are the limited availability of landfill space and the need for leachate treatment over time, even with a well-engineered landfill. Substantial PFASs and BFRs have been detected in landfill leachate so far. For example, PFHxA was detected at 12 to 5700 ng/L in landfill leachates [61]. More than 172 PPCPs were reported in landfill leachates from 1993 to 2018 [234]. The reported PPCP concentrations varied depending on the PPCP type, with the highest PPCP leachate concentration belonging to diethyltoluamide at 0.05 mg/L.

#### 3.4.4. Summary

The treatment technologies and relevant information from the reviewed publications are shown in Table 2 below. According to our data collection, biosolids treatment technologies are at different stages of research, development, and commercialization. Many of the biological technologies studied have been applied at full scale for treatment of OCs in biosolids, exceptions include mycoremediation, HALT, and SCWO.

## 4. Multicriteria assessment of the treatment technologies

To support an in-depth analysis of treatment technologies, a multicriteria analysis for different treatment technologies was developed to help assess the suitability of technology and to aid in the selection process (Table 3) based on selected criteria developed elsewhere [125, 15,9]. The detail of the criteria and rationale for selecting the criteria are given in Appendix 6. The assessment was developed based on the availability of relevant MPs and OCs and team knowledge, particularly for PFASs, as they can be recalcitrant to destructive treatment.

For treatment performance criteria, biological technologies are not an essential high-performance option for the complete degradation of OCs compared to other thermal- and chemical-based technologies. Microbial activity in aerobic, anaerobic digestion, and composting requires time to establish the culture, hence taking longer to completely degrade OCs in biosolids. Mycoremediation also carries a high risk of losing enzyme activity. The advantage of biological technologies is their potential for selective degradation of specific OCs; however, some recalcitrant OCs cannot be degraded completely by biological technologies. Thermal treatment technologies demonstrate outstanding performance with the fastest degradation rate and complete mineralization of OCs. Thermal technologies are less selective for OCs than biological technologies. Their drawback, however, is that using thermal technologies for degradation of OCs is a secondary application rather than the primary purpose, such as in cement kilns. SWCO is also not an option for most OCs. SWCO has only recently emerged for PFAS destruction because PFASs are stable and resistant to most conventional biological processes. SWCO is still in progress to demonstrate viability of biosolids treatment at full-scale treatment. AOPs and other chemical technologies can also provide rapid degradation of OCs. They exhibit high versatility, being applicable for specific OCs and conditions using certain chemicals. However, the complexity of chemical and thermal technologies is more advanced than that of biological technologies. The limitation of all three technology groups is the potential generation of by-products. Biological technologies are associated with secondary waste such as greenhouse gases and excess biosolids, which can also be problematic. The use of chemicals in chemical technologies can also result in the formation of undesirable toxic by-products.

The socio-environmental criteria show that biological processes can be affected by seasonal variations in temperature [35]. It is expected that during winter, performance of biological process is less. Biological process and landfilling also require a reasonably large amount of land to deal with large amount of biosolid generated, but overall, they are well performed in social acceptability, management of co-contaminants and resource recovery (e.g., biogas). For an example of water security,

**Table 2**  
Summary of information for remediation technologies for OCs treatment in biosolid.

Treatment technology	Treatment Principle	General Operating Conditions	Reported Degradation Performance	References
Aerobic digestion	Anaerobic biodegradation	pH: 6.0 – 7.5 Temperature: 55 – 70 °C	80% PPCPs	[1-3]
Anaerobic digestion	Aerobic biodegradation	Thermophilic range: 50 – 60 °C Mesophilic range: 30 – 38 °C	40 – 80% PPCPs	[4,5]
Composting	Biodegradation	Aerated static pile or in-vessel: 55 °C in 3 d Windrow: 55 °C in 15d.	60 – 85% PPCPs	[6,7]
Mycoremediation by fungi	Fungal decomposition	Retention time: 14d	100% PPCPs (Oxytetracycline)	[8-10]
Hydrothermal alkaline treatment	Elevated temperature, pressure, and pH	Temperature: 350 – 450 °C	99.9% long chain PFASs	[11,12]
Incineration, Pyrolysis and gasification	Incineration: high temperature in the presence of molecular oxygen Pyrolysis: high temperature with absence of oxygen Gasification: limited amount of oxygen	Incineration: 980 – 1200 °C Pyrolysis: 400 – 800 °C. Gasification: 700 – 1000 °C.	90% PFASs	[13,14]
Super Critical Water Oxidation	Temperatures and pressures above the critical point of water	Temperature: >374 °C Pressure: >22.1 MPa	99.99% long chain PFASs	[15]
Cement kiln	Elevated temperatures in the presence of molecular oxygen	Temperature 1400 °C – 2000°C 10 s gas residence time 30 min solid residence time	100% PPCPs	[16,17]
Ozone	Degradation of OCs using oxidation by O <sub>3</sub> In combination with electrooxidation	5.2 g O <sub>3</sub> /h Flowrate: 0.28 m <sup>3</sup> /h	67 – 86% PPCPs (Nonylphenol, Bisphenol A, Triclosan)	[18]
Fenton	Degradation of OCs using oxidation by iron-catalyzed decomposition of H <sub>2</sub> O <sub>2</sub>	H <sub>2</sub> O <sub>2</sub> : 0.1 - 1% Fe <sup>2+</sup> catalyst: 10 - 100 mg/L pH: 2 - 4	84% PPCPs (Clarithromycin)	[19,20]
UV	Degradation of OCs using ultraviolet energy	pH = 3, UV wavelength = 253.7 nm UV intensity = 0.069 mW/cm <sup>2</sup>	(Not applicable for biosolids)	[21]
Microwave-assisted persulfate oxidation	Degradation of OCs using microwave radiation	Persulfate concentration: 1 - 10 mM Fe <sup>2+</sup> or Mn <sup>2+</sup> : 0.1 - 1 mM	28 - 42% PFASs	[22]
Ultrasonic (pretreatment method)	Degradation of OCs using high frequency sound waves	20 kHz to 1 MHz Power density: 0.1 to 10 W/cm <sup>2</sup>	20% PPCPs	[23,24]
Alkaline stabilization	Raising pH	pH: above 12 in 72 h Temperature: 52 °C High moisture content	90% PPCPs	[25,26]
Landfill	Physical isolation	Low permeability site Impermeable liner system	(Not applicable for treatment purpose)	[27,28]

References: [1] Li et al. [106], [2] Li et al. [118], [3] Pham et al. [158], [4] Samaras et al. [172], [5] Huang et al. [80], [6] Bao et al. [16], [7] Sun et al. [193], [8] Sadañski et al. [170], [9] Kaewlaoyong et al. [87], [10] Luo et al. [126], [11] Pinkard [159], [12] Soker et al. [184], [13] Kundu et al. [97], [14] Chen et al. [39], [15] McDonough et al. [137] [16] Kadam et al. [86], [17] Pang et al. [153], [18] de Leon-Condes et al. [50], [19] Flotron et al. [59], [20] Karaolia et al. [88], [21] Salihoglu et al. [171], [22] Hamid and Li [72], [23] Benabdallah El-Hadj et al. [18], [24] Chawla et al. [31], [25] Rathankumar et al. [166], [26] Vaithyanathan et al. [209], [27] Gallen et al. [61], [28] Silva et al. [179]

anaerobic digestion shows the highest net-negative value, saving 158 m<sup>3</sup> net water usage to treat 1000 m<sup>3</sup> of secondary effluent [196]. Also, anaerobic digestion produces the least secondary air emission or by-products, while incineration/cement kiln produces the most. Anaerobic digestion is also the best technology in response to energy criteria (–192 kWh), while pyrolysis consumes the highest energy at 364 kWh [196]. Thermal treatment also scores well in the socio-environment criteria, except the resource utilization as it is an energy consumption process. Chemical technologies such as AOP do not fit well in the socio-environmental criteria as they are advanced process which consume energy, resources, and require a high level of monitoring and operation. AOP and thermal are typically only necessary to meet the stringent discharge standard of certain OCs.

For the techno-economic criteria, biological process and landfilling benefit from their maturity as available commercial technologies, with process automation, low capital costs and high resilience to geological conditions. While landfill is a cost-effective technology, other biological technologies require less land use and are potentially more widely accepted. The thermal treatments and AOPs are far from a satisfactory level for these criteria. It is well known that AOP and some of the thermal technologies (i.e., incineration, pyrolysis, gasification) are commercially available but their complexity and capital costs are drawbacks. Advanced oxidation processes play a minor role in remediation of OCs from biosolids and are usually an add-on with other treatment technologies, hence offering little value. After a multicriteria

assessment, An et al. [9] concluded that composting and thermal technologies had the lowest score as sustainable technologies for biosolids treatment. Practically, a combination of these technologies in form of a treatment train may be used to balance between the overall efficiency of OCs degradation, and economic viability.

Based on all the above information, scoring outcome of multicriteria analysis, and knowledge of the team, we map out the relevance of the development and practicability of the technologies in Fig. 6 relative to their applicability for treatment of biosolids.

## 5. Challenges and outlook

Based on the knowledge and data collected from this review, the following issues should be considered for better understanding of technologies applicable for treatment of OCs in biosolids:

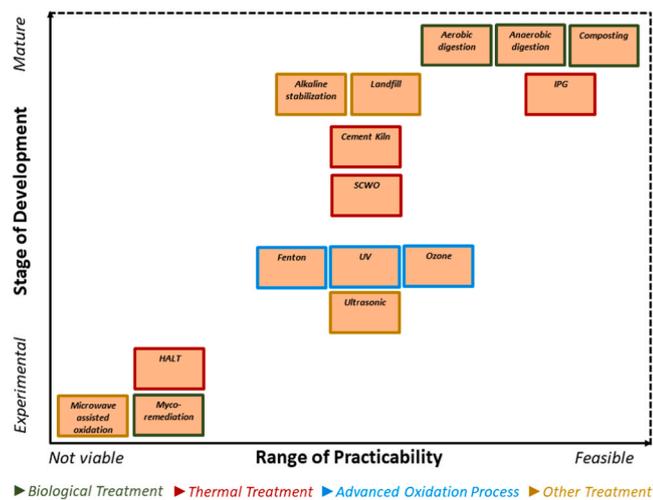
- Only a few studies have investigated the occurrence of LASs/surfactants and PFRs in biosolids, with a general lack of spatiotemporal trends of the chemicals.
- Studies on the fate of several chemicals (e.g., PFASs, PFRs, PCBs, dioxins) in biosolids are still necessary. Governments are keenly interested in determining the impact of wastewater treatment processes on PFASs, which can be exported to the environment through biosolids.

**Table 3**  
Multicriteria analysis of biosolids treatment technologies.

Criteria		Acrobic digestion	Anaerobic digestion	Composting	Myco-remediation	HALT	SCWO	Cement kiln	Ozone	Fenton	UV	Microwave-assisted	Ultrasonic	Alkaline stabilization	Incineration, Pyrolysis, Gasification	Landfilling
Treatment performance criteria	Low sensitivity to variation of the biosolids matrix	Green	Green	Yellow	Green	Green	Green	Green	Green	Green	Green	Green	Green	Green	Green	Green
	Minimal secondary waste generated (air, dust, solid, liquid)	Green	Green	Green	Green	Green	Red	Green	Green	Green	Green	Red	Red	Green	Red	Yellow
	Potential for adverse fugitive emissions	Green	Yellow	Green	Green	Green	Green	Green	Green	Green	Green	Green	Green	Green	Green	Green
	High destruction efficiency of OCs	Yellow	Green	Green	Green	Green	Red	Green	Green	Green	Green	Green	Red	Green	Green	Red
	Low level of complexity	Green	Green	Green	Green	Yellow	Red	Green	Red	Red	Red	Green	Green	Green	Green	Green
Socio-environmental criteria	Resilience to seasonal variation	Yellow	Green	Green	Green	Green	Green	Green	Green	Green	Green	Green	Green	Green	Green	*
	Low level of monitoring	Green	Green	Green	Green	Green	Green	Green	Red	Red	Red	Green	Green	Green	Green	Green
	High social/regulatory acceptability	Green	Green	Green	Green	Green	Green	Green	Green	Green	Green	Green	Green	Green	Green	Green
	Adequate management of co-contaminants	Green	Green	Yellow	Green	Green	Green	Green	Green	Green	Green	Green	Green	Green	Green	Green
	Efficient use of land/space	Yellow	Yellow	Green	Red	Green	Green	Green	Green	Green	Green	Green	Green	Green	Green	Red
	Efficient use of resources	Yellow	Green	Green	Green	Green	Green	Red	Red	Red	Red	Green	Green	Red	Green	Green
Techno-economic criteria	Degree of commercialization for treatment of OCs	Green	Green	Green	Green	Green	Red	Green	Green	Green	Green	Green	Green	Green	Green	Green
	Automated process with minimum labour required	Green	Green	Green	Green	Red	Green	Green	Green	Green	Green	Green	Green	Green	Green	Green
	High capital costs	Green	Green	Green	Green	Green	Green	Green	Green	Green	Green	Green	Green	Green	Red	Green
	Low sensitivity to geographical setting	Green	Green	Yellow	Green	Green	Green	Green	Green	Green	Green	Green	Green	Green	Green	Green
	Low level of maintenance/operation	Yellow	Yellow	Green	Green	Red	Green	Red	Yellow	Yellow	Yellow	Yellow	Yellow	Green	Red	Green

\* The indicators are Good (green), Fair (yellow), Bad (red).

\*\* All technologies meant to either target OCs directly or after extraction of OCs from biosolids.



**Fig. 6.** Practicability and development of technologies for the treatment of OCs in biosolid. The treatment technologies are either sole or in combination with other technologies. All the boxes are same size. The figure is subject to be revision for treatment of PFASs where biological treatment, and alkaline stabilization are essentially ineffective. Conversely, SCWO and HALT would be scored as performing better than AOPs for treatment of PFASs. \*IPG is Incineration, Pyrolysis, Gasification.

- The quantification of OCs in biosolids is difficult and subject to wide variation due to matrix effects, depending on sampling and testing methods. Most studies rely on grab sampling, which only reflects the concentration at a specific time point in biosolids treatment. Composite samples, representing better average concentrations, should be investigated to improve the quality of produced data. Understanding the effect of seasons, climate, and social-economic patterns also remains a knowledge gap that should be addressed in future studies. Difficulties with adequate quantification methods should also be addressed to reduce data variation. For example, several methods have been used for microplastics, such as microscopy, Raman spectrometry, FTIR, and pyrolysis GC-MS. At this stage, there is no standard method for measurement of MPs. There is also a challenge with cross-contamination due to plastic consumables in the lab, which can compromise QA/QC of the results.
- Thermal treatment is a viable option for biosolids as it completely degrades OCs, however the energy costs are high. Currently, the global perspective is to balance the application of thermal treatment and resource recovery. Some countries have been thermally treating 90 to 100% of their biosolids (e.g., Netherlands, Switzerland, Belgium), while others are moving towards balancing with resource recovery (e.g., UK, Australia, Spain, France).
- Although the list of regulated OCs in biosolids is continually evolving, current regulations are early and do not completely cover all of the potentially harmful compounds being identified in biosolids (e.g., MPs, pharmaceuticals, PFASs). Reducing the use and

consumption of OCs would be a better option than retrofitting WWTPs as the latter will be costly and ultimately less sustainable, stated in the Australian – New Zealand Biosolids Partnership fact-sheet [11]. It is also agreed that controlling the source of all plastics entering the environment is a complex but important global and regional goal. However, more effort on the research and development of suitable replacement products and materials is critically needed. In addition, it is challenging to assess and justly allocate financial responsibility for the treatment of OCs, either the consumers, manufacturers, or WWTPs themselves.

## 6. Conclusions

This review provides comprehensive progress on the occurrence and treatment of OCs in biosolids. The focus on OC occurrence in biosolids was intended to address limitations of previous studies, which were focused on treatment of OCs in the aqueous phase. This review also approached an extensive data set consisting of MPs and 10 classes of OCs. The key conclusions of this study are:

- Data have shown that the MPs and LASs/surfactants account for the highest fractions of OCs (up to mg/g) in biosolids, whereas dioxin, PCBs and PFRs were the lowest. The main reasons lie in their usage and production in the population. Chemicals that originate from natural sources or have been banned or regulated appeared in biosolids to a lesser extent than others.
- Europe seems to have higher biosolids concentrations of dioxin, PFASs, PAHs, PPCPs, UV filters, and PFRs than Asia and the Americas. The Americas had the highest levels of BFRs and biocides than others. However, Australia was found to have the highest levels of microplastics in biosolids compared to other countries, which data compiled from limited available literatures, hence, required more monitoring data for in-depth analysis.
- The Log  $K_d$  of most OCs is less than 4, except BFRs, which indicates BFRs will exhibit substantial partitioning within biosolids. However, BFRs were banned in 1979; hence the concentration of BFRs in biosolids is low as they are a group of legacy chemicals.
- Although several technologies have been developed for biosolids treatment, biological treatments (e.g., anaerobic treatment, composting) and physical treatments (landfilling) are currently the options most frequently used. Anaerobic digestion has historically worked well as a treatment methodology for a wide variety of organic pollutants in biosolids and continues to be a popular and mature technology for OC treatment. It has high water and energy recovery and produces fewer secondary pollutants. Thermal treatment will remain an energy-intensive, higher cost treatment option that also must meet stringent air discharge requirements in order to gain legislative and public acceptance.

## CRedit authorship contribution statement

**Phong H.N. Vo:** Writing – review & editing, Writing – original draft, Visualization, Supervision, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Gia Ky Le:** Writing – original draft, Methodology, Data curation. **Lai Nguyen Huy:** Writing – original draft, Investigation, Formal analysis, Data curation, Conceptualization. **Lei Zheng:** Writing – original draft. **Chawalit Chaiwong:** Writing – original draft. **Nam Nhat Nguyen:** Writing – original draft. **Hong T.M. Nguyen:** Writing – review & editing, Supervision, Investigation. **Peter J. Ralph:** Writing – review & editing, Investigation, Formal analysis. **Unnikrishnan Kuzhiumparambil:** Writing – review & editing. **Soroosh Danaee:** Writing – original draft. **Sonja Toft:** Writing – original draft, Supervision, Resources. **Craig Madsen:** Resources, Formal analysis. **Mikael Kim:** Writing – review & editing. **Jim Fenstermacher:** Writing – review & editing, Supervision, Resources. **Ho Truong Nam Hai:** Writing – original draft. **Haoran Duan:** Writing – review & editing,

Validation, Methodology. **Ben Tscharke:** Writing – review & editing, Validation, Supervision, Methodology, Formal analysis, Conceptualization.

## 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.

## Data Availability

Data will be made available on request.

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## Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at [doi:10.1016/j.jhazmat.2024.133471](https://doi.org/10.1016/j.jhazmat.2024.133471).

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