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Review

Removal of organic micropollutants in waste stabilisation ponds: A review

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ABSTRACT

As climate change and water scarcity continue to be of concern, reuse of treated wastewater is an important water management strategy in many parts of the world, particularly in developing countries and remote communities. Many countries, especially in remote regional areas, use waste stabilisation ponds (WSPs) to treat domestic wastewater for a variety of end uses, including using the treated wastewater for irrigation of public spaces (e.g. parks and ovals) or for crop irrigation. Thus, it is vital that the resulting effluent meets the required quality for beneficial reuse. In this paper, both the performance of WSPs in the removal of organic micropollutants, and the mechanisms of removal, are reviewed. The performance of WSPs in the removal of organic micropollutants was found to be highly variable and influenced by many factors, such as the type and configuration of the ponds, the operational parameters of the treatment plant, the wastewater quality, environmental factors (e.g. sunlight, temperature, redox conditions and pH) and the characteristics of the pollutant. The removal of organic micropollutants from WSPs has been attributed to biodegradation, photodegradation and sorption processes, the majority of which occur in the initial treatment stages (e.g. in the anaerobic or facultative ponds). Out of the many hundreds of organic micropollutants identified in wastewater, only a limited number (40) have been studied in WSPs, with the majority of these pollutants being pharmaceuticals, personal care products and endocrine disrupting compounds. Thus, future research on the fate of organic micropollutants in WSPs should encompass a broader range of micropollutants and include emerging organic pollutants, such as illicit drugs and perfluorinated compounds. Further research is also needed on the formation and toxicity of transformation products from organic micropollutants in WSPs, since the transformation products of some organic micropollutants can be more toxic than the parent compound. Combining other wastewater treatment processes with WSPs for removal of recalcitrant organic micropollutants should also be considered.

Contents

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1. Introduction

Waste stabilisation ponds (WSPs) are large shallow ponds that utilise physical and biological processes to remove organic materials, pollutants and pathogens present in raw wastewater. They are one of the simplest methods of wastewater treatment and are widely employed all over the world, particularly in developing countries where sufficient land is normally available and the climate is more favourable (high temperature and sunlight) for their operation (Mara, 2006, 2004; Mara and Pearson, 1998; von Sperling, 2007). In Europe, WSPs are widely used for small rural communities of up to approximately 2000 inhabitants, although larger systems are found in France (over 3000 WSPs), as well as in Spain and Portugal (Keffala et al., 2013). Approximately one third of all wastewater treatment plants (WWTPs) in the USA are WSPs, usually servicing communities of up to 5000 people. In warm climates, such as North Africa, the Middle East, Asia and South America, WSPs are commonly used to treat wastewater from large (up to one million) populations (Keffala et al., 2013). In Australia, WSPs operate for a range of population sizes, from small remote communities of 1000–2500 inhabitants (Sheludchenko et al., 2016), to larger facilities servicing populations of up to 600,000 (Busine and Oemcke, 2003).

The scarcity of reliable water sources in many parts of the world, due to population increases, deterioration in the quality of surface waters, depletion of groundwater and climate change, has resulted in the use of recycled water as an alternative water source (Chen et al., 2012b). In many communities, treated wastewater is often the only irrigation option, particularly in developing countries with heavily utilized agricultural areas. However, to date, the majority of studies of wastewater reuse have focussed on large metropolitan treatment plants employing advanced activated sludge treatment and water recycling technologies, such as reverse osmosis membranes or advanced oxidation processes (Busetti et al., 2015; Rodriguez et al., 2009; Van Buynder et al., 2009). This type of treatment is considered to be “best practice” and produces high quality recycled water (Rodriguez et al., 2009). However, in rural WSPs, treatment for water reuse is often limited to disinfection by chlorine. Unlike reverse osmosis, chlorine disinfection does not provide additional chemical removal from wastewater, and may actually react with organic micropollutants present in wastewater to produce potentially harmful disinfection by-products (Krasner et al., 2008; Liew et al., 2012).

Organic micropollutants include a wide group of anthropogenic and natural compounds, such as pharmaceuticals and personal care products (PCPPs), steroid hormones and other endocrine disrupting compounds (EDCs), surfactants, industrial chemicals and pesticides (Luo et al., 2014). While current wastewater treatment processes can reduce the concentrations of many micropollutants, they are not specifically designed to remove them. Therefore, these contaminants can still be present in the resulting wastewater effluent, which is then either discharged to the environment or used in recycling schemes. Additionally, it is possible that some organic micropollutants are transformed into other compounds during wastewater treatment. The impact of these transformation products on the environment must also be considered, as these transformation products can be more toxic than the parent compound (Garcia-Rodríguez et al., 2014). The concentrations of organic micropollutants in wastewater effluents can range from a few nanograms per litre (ng/L) to several micrograms per litre (μg/L) (Luo et al., 2014).

Many organic micropollutants have been identified in wastewater and in wastewater-impacted environments (Barnes et al., 2008; Focazio et al., 2008; Schwarzenbach et al., 2006; Ying et al., 2009), with growing concern about the health and environmental impact of these chemicals in both environmental discharge and wastewater reuse applications. However, to date, there have been relatively few studies of micropollutant removal in WSPs. Most studies and reviews of micropollutants in wastewater have focussed on mechanised wastewater treatment processes, such as activated sludge treatment or membrane bioreactors (Bonvin et al., 2016; Fernandez-Fontaina et al., 2012; Homem and Santos, 2011; Jiang et al., 2013; Kim et al., 2007; Luo et al., 2014; Ternes, 1998; Verlicchi et al., 2012), reverse osmosis (Busetti et al., 2015; Rodriguez et al., 2009; Van Buynder et al., 2009) or advanced oxidation processes (Chen et al., 2012a; Huber et al., 2005; Lee and von Gunten, 2010; Reungoat et al., 2012; Ternes et al., 2003).

Choosing low cost and low energy technologies for wastewater treatment and reuse is of great importance, particularly in developing countries and remote rural communities. While some aspects of WSP performance, such as pathogen inactivation (Bolton et al., 2010; Curtis et al., 1992; Davies-Colley et al., 2000, 1999; Hosetti and Frost, 1998; Mara, 2013; Maynard et al., 1999) and nutrient removal (Brown and Shilton, 2014; Camargo Valero et al., 2010a, 2010b; Maynard et al., 1999; Mayo and Abbas, 2014; Powell et al., 2011a, 2011b; Senzia et al., 2002; van der Linde and Mara, 2010) have been well studied, WSP performance in the removal of organic micropollutants, has not been widely investigated. Garcia-Rodríguez et al. (2014) reviewed the ability of biologically based wastewater treatment systems (i.e. constructed wetlands, WSPs, high rate algal ponds (HRAPs) and *Daphnia* and fungal reactors) to remove organic micropollutants, however, much of the review focussed on constructed wetlands, with only a few studies on WSPs. Thus this paper reviews the current state of knowledge on the application of WSPs for the removal of organic micropollutants from wastewater, including the possible removal mechanisms and the impact of design and environmental factors on the removal efficiency. Potential knowledge gaps for further research in the future are also identified. This is the first comprehensive review of the performance of WSPs for the removal of organic micropollutants.

2. An overview of WSP treatment and performance

Conventional WSPs, often referred to as lagoons, usually consist of a combination of three different types of ponds: anaerobic ponds (APs), facultative ponds (FPs) and maturation ponds (MPs) (Mara, 2006, 2004; Mara and Pearson, 1998; Polprasert and Kittipongvises, 2011; Sah et al., 2012). The main features and functions of each pond type are summarised in Table 1. The ponds

Table 1
Main features and functions of anaerobic, facultative and maturation ponds. Adapted from Polprasert and Kittipongvises (2011).

Feature	Anaerobic ponds	Facultative ponds	Maturation ponds
Depth (m)	3–5	1.5–3	1–2
HRT (days)	10–50	5–20	5–10
OLR (kg BOD_5 ha^{-1} d^{-1})	High, >300	Medium, 100-300	Low, <100
Microorganisms responsible for BOD reduction	Anaerobic bacteria	Facultative bacteria and algae	Aerobic bacteria
Major functions	Pretreatment and BOD reduction	BOD reduction and nutrient removal	Polishing, SS and pathogen reduction and some nutrient removal
By-products formed	Biogas, methane, carbon dioxide	SS in the form of algal and bacterial cells, carbon dioxide	none

Key: HRT = hydraulic retention time; OLR = organic loading rate; BOD5 = 5 day biochemical oxygen demand; SS = suspended solids.

differ from each other in geometry, hydraulic flows, organic loading and important biochemical processes. The ponds also differ in their efficiency in carbon, nutrient and pathogen removal, with removal efficiency generally increasing with increasing number of ponds (von Sperling, 2007). Treatment performance in WSP is strongly influenced by: the type of wastewater, the amount of organic loading, the geometry and physical configuration of the pond system, and the hydraulic behaviour (Mara, 2004; Sah et al., 2012; Shilton, 2005). WSP treatment performance is also impacted by the many physical, chemical and biological processes (e.g. sedimentation, adsorption, biodegradation) operating within each pond, which are influenced by environmental factors, such as the presence of algae, light intensity, pH and temperature (Sah et al., 2012; Shilton, 2005; von Sperling, 2007). A complete and detailed overview of the physical, chemical and biological processes operating within each pond is presented in Shilton (2005).

Algae can enhance the removal of organic contaminants from wastewater (Matamoros and Rodríguez, 2016; Matamoros et al., 2016a, 2015; Muñoz and Guieysse, 2006; Zhou et al., 2014). Algae can be sensitive to toxic pollutants, however, some types of algae are more tolerant to pollutants than others (Muñoz and Guieysse, 2006; Palmer, 1969). When appropriate methods of algal selection and cultivation are used, it is possible to use algae to biodegrade a wide range of hazardous pollutants, such as polycyclic aromatic hydrocarbons, phenolic compounds and organic solvents (Muñoz and Guieysse, 2006), as well as PPCPs, EDCs (Matamoros et al., 2016a, 2015; Zhou et al., 2014) and pesticides (Matamoros and Rodríguez, 2016). In recent years, high rate algal ponds (HRAPs) have received increasing attention due to their effectiveness in treating wastewater and production of algal biomass for food or energy production (Craggs et al., 2014; Cromar and Fallowfield, 1997; Oswald, 1988; Park et al., 2011a, 2011b; Park and Craggs, 2011; Rawat et al., 2011). They have also been shown to be effective in removing certain organic micropollutants (e.g. pharmaceuticals) (Matamoros et al., 2016b, 2015).

The pH can affect the removal mechanisms of some organic micropollutants. For example, at neutral pH (7–8) of wastewater, acidic pharmaceuticals, such as gemfibrozil, diclofenac, ibuprofen and naproxen, occur as anions and thus will remain in the dissolved phase of the WSP, where the likely mechanisms of elimination will be biodegradation and/or photodegradation (Ying et al., 2009). The pH of WSPs varies diurnally and with depth based on rates of algal photosynthesis and respiration, with values varying during the day in the range of 7–9.4 or higher (Bolton et al., 2010; Kayombo et al., 2002; Paterson and Curtis, 2005). The pH of a pond system can be used as an indicator of overall pond performance. For example, low pH values (e.g. < 7) indicate incomplete consumption of CO_2 by photosynthesis (Kayombo et al., 2002). Reduced pH can also indicate overload of sewage, which can lead to reduced concentrations of dissolved oxygen and production of bad odours (von Sperling, 2007).

Temperature plays an important role in WSPs with respect to the activity of algae and bacteria and can impact the removal of micropollutants in WSPs, especially *via* biodegradation, with increased removals typically reported during warmer months (Li et al., 2013; Matamoros et al., 2015). For example, Li et al. (2013) observed higher removal efficiencies of PPCPs and EDCs in a US lagoon system in September (average temperatures of 20 °C) than in November (average temperatures of 4 °C), while Matamoros et al. (2015) observed that removal of both biodegradable compounds (oxybenzone, naproxen, benzothiazole, methyl paraben, benzotriazole, 5-methyl-benzotriazole, OH-benzothiazole and triphenyl phosphate) and moderately volatile compounds (e.g. galaxolide and tonalide) were better in the summer (average temperature 26 °C) compared to winter (average temperature 11 °C). Biodegradation is usually optimised at the physiological temperature of the microorganism, as this promotes optimal activity. For example, the optimum temperature for biodegradation rates for the antibiotic ceftiofur was found to be between 35 °C and 40 °C, close to the optimum temperature for gut microorganisms of 37 °C (Li et al., 2011).

Light plays a major role in the performance of WSPs by providing the energy source for photosynthesis (Heaven et al., 2005; Davies-Colley et al., 2005) and also provides a major pathway for micropollutant degradation through direct or indirect UV (200–400 nm) photodegradation (Boreen et al., 2003; Packer et al., 2003; Rivera-Utrilla et al., 2013; Ryan et al., 2011). The rate of photodegradation of a particular compound is influenced by the variation in the intensity of solar irradiance with both latitude and season (Andreozzi et al., 2003). For example, Matamoros et al. (2015) found that removal of photodegradable compounds (e.g. diclofenac and ketoprofen) was better in summer when the average daily solar irradiation was 282 W m^{-2}, compared to winter when the solar irradiation was much lower (74 W m^{-2}).

3. Removal of organic micropollutants in WSPs

There are only nine published studies on the removal of organic micropollutants in WSPs to date, and, of the hundreds of organic micropollutants that have been identified in wastewater, approximately forty have been studied in WSPs (Table 2), leaving a significant knowledge gap in this area given the prevalence of WSP technology worldwide. Most of the compounds studied in WSP to date are classed as PPCPs or EDCs. For the organic micropollutants which have been studied in WSPs, removal has been reported to be the same, or better, than conventional treatments, but removal performance is highly variable (Table 2), ranging from <30% for recalcitrant compounds, such as carbamazepine, gemfibrozil and propranolol, to > 95% for easily removed compounds (e.g. caffeine, ibuprofen, paracetamol and methyl dihydrojasmonate). In general, most compounds that are recalcitrant in conventional treatments (e.g. carbamazepine) are also poorly removed in WSPs, while

Table 2
Summary of organic micropollutants studied in WSPs to date.

Compound	Average removal efficiency (%)	References
Stimulants		
Caffeine	70–100	Camacho-Muñoz et al. (2012); Hijosa-Valsero et al. (2010b); Li et al. (2013); Matamoros et al. (2016b); Ying et al. (2009)
Pharmaceuticals		
Paracetamol	99	Matamoros et al. (2016b)
Ibuprofen	75–100	Camacho-Muñoz et al. (2012); Hijosa-Valsero et al. (2010b); Hoque et al. (2014); Li et al. (2013); Matamoros et al. (2016b); Ying et al. (2009)
Ketoprofen	87–100	Hijosa-Valsero et al. (2010b); Ying et al. (2009)
Naproxen	75–100	Camacho-Muñoz et al. (2012); Hijosa-Valsero et al. (2010b); Li et al. (2013); Matamoros et al. (2016b); Ying et al. (2009)
Diclofenac	65–90	Hijosa-Valsero et al. (2010b); Matamoros et al. (2016b); Ying et al. (2009)
Salicylic acid	95–100	Camacho-Muñoz et al. (2012); Hijosa-Valsero et al. (2010b)
Carbamazepine	6–29	Camacho-Muñoz et al. (2012); Matamoros et al. (2016b); Ying et al. (2009)
Gemfibrozil	15–20	Camacho-Muñoz et al. (2012); Ying et al. (2009)
Propranolol	10	Camacho-Muñoz et al. (2012)
Trimethoprim	44–100	Hoque et al. (2014); Li et al. (2013)
Sulfamethoxazole	82–100	Camacho-Muñoz et al. (2012); Hoque et al. (2014); Li et al. (2013)
Diphenhydramine	85	Li et al. (2013)
Found in Personal Care Products		
Methyl dihydrojasmonate	80–99	Hijosa-Valsero et al. (2010b); Matamoros et al. (2016b)
Galaxolide	75–96	Hoque et al. (2014); Matamoros et al. (2016b)
Tonalide	76–96	Hoque et al. (2014); Matamoros et al. (2016b)
Cashmeran	82	Matamoros et al. (2016b)
Furosemide	99	Matamoros et al. (2016b)
Oxybenzone	99	Matamoros et al. (2016b)
Methylparaben	81	Matamoros et al. (2016b)
Triclosan	77–100	Hoque et al. (2014); Li et al. (2013); Matamoros et al. (2016b); Ying et al. (2009)
Triclocarban	85	Li et al. (2013)
Endocrine disrupting compounds		
Octylphenol	71	Matamoros et al. (2016b)
Bisphenol A (BPA)	24	Qiang et al. (2013)
Androstendione	93–100	Coleman et al. (2010)
Androsterone	100	Coleman et al. (2010)
Eticholanolone	100	Coleman et al. (2010)
17β-Trenbolone	100	Coleman et al. (2010)
Estrone (E1)	31–100	Coleman et al. (2010); Pessoa et al. (2014); Qiang et al. (2013)
17β-Estradiol (E2)	42–62	Pessoa et al. (2014); Qiang et al. (2013)
Estriol (E3)	26–77	Coleman et al. (2010); Qiang et al. (2013)
17α-Ethinyl estradiol (EE2)	35–99	Pessoa et al. (2014); Qiang et al. (2013)
4-Nonylphenol (NP)	75	Qiang et al. (2013)
Artificial sweetener		
Sucralose	Not removed	Hoque et al. (2014)
Other		
Benzotriazole	50	Matamoros et al. (2016b)
5-Methyl benzotriazole	76	Matamoros et al. (2016b)
Benzothiazole	81	Matamoros et al. (2016b)
OH-Benzothiazole	86	Matamoros et al. (2016b)
Tributyl phosphate	59	Matamoros et al. (2016b)
Tris(2-chloroethyl) phosphate	21	Matamoros et al. (2016b)
Triphenyl phosphate	76	Matamoros et al. (2016b)

compounds that are well removed in conventional treatments (e.g. caffeine, paracetamol) tend to be well removed in WSPs. Very limited or no studies have been published on the removal of pesticides, perfluorinated compounds, illicit drugs or artificial sweeteners in WSPs.

Removal of organic micropollutants from wastewater is a complex process involving a variety of physical, chemical and biological processes which are influenced by many design aspects of the treatment system (e.g. hydraulic residence time (HRT) and the organic loading rate) and environmental conditions (sunlight, temperature, redox conditions, pH and presence of toxic compounds), as well as the composition of wastewater, operational conditions of the WWTP and the characteristics of the micropollutants (e.g. hydrophobicity, biodegradability and volatility) (Clara et al., 2005; Luo et al., 2014; Pessoa et al., 2014). For example, micropollutants with slow to intermediate degradation kinetics, such as fluoxetine or some antibiotics, will be less efficiently biodegraded at shorter HRTs or increased loading rates (Fernandez-Fontaina et al., 2012; Luo et al., 2014).

In WSPs, organic micropollutant removal often occurs in the early stages of treatment (e.g. either in the anaerobic or facultative ponds) and higher HRTs have resulted in better compound removals (Hijosa-Valsero et al., 2010b; Li et al., 2013). Removals tend to improve in more sophisticated systems, with an increasing number of ponds, or through combination of WSPs with another biologically based wastewater treatment process, such as a constructed wetland (Garcia-Rodríguez et al., 2014). Environmental conditions (or seasonality) can play an important role in the removal of organic micropollutants, since seasonality affects temperature, sunlight duration and intensity, and biomass production.

However, seasonal variations in organic micropollutant removal are mostly evident in climates with cold winters (ambient temperatures ≤ 4 °C) compared to more temperate climates with warmer winter temperatures ($\geq$11 °C) (Matamoros et al., 2016b).

3.1. Key organic micropollutant removal mechanisms in WSPs

The main reported removal mechanisms occurring in WSPs are photodegradation, biodegradation and sorption onto organic matter (Fig. 1.), processes which can be enhanced by the presence of algae or some aquatic plants (Garcia-Rodríguez et al., 2014). Organic micropollutants that are removed from the WSP wastewater effluent through sorption to the sludge can create potential issues for sludge disposal and be potentially remobilised back into the water column. Organic pollutants that are transformed *via* either photodegradation or biodegradation may produce transformation products that are more (or less) toxic than the parent compound. A general overview of the three possible removal mechanisms is presented below, with specific examples of chemical removal presented in subsequent sections.

3.1.1. Photodegradation

Photodegradation of organic micropollutants upon exposure to sunlight can occur either by direct or indirect mechanisms (Andreozzi et al., 2003; Garcia-Rodríguez et al., 2014; Rivera-Utrilla et al., 2013; Vione et al., 2014). Direct photodegradation involves direct absorption of solar light by the contaminant, followed by a chemical reaction (Andreozzi et al., 2003; Garcia-Rodríguez et al., 2014; Rivera-Utrilla et al., 2013; Vione et al., 2014). The structure of the organic micropollutant determines whether it will absorb radiation, thereby increasing its energy which can lead to bond breaking and degradation (Rivera-Utrilla et al., 2013). Indirect photodegradation reactions are promoted by reactive species (e.g. reactive oxygen species, such as hydroxyl radicals or singlet oxygen) produced by natural photosensitisers, such as nitrate and nitrite anions or chromophoric dissolved organic matter (i.e. the fraction of dissolved organic matter that can absorb sunlight), present in the water (Niu et al., 2016; Rivera-Utrilla et al., 2013; Vione et al., 2014). Although photodegradation of organic micropollutants is acknowledged as a key removal mechanism in WSPs (Garcia-Rodríguez et al., 2014), most studies on the photodegradation of organic micropollutants have been conducted under laboratory conditions with simulated sunlight or in natural waters (e.g. Andreozzi et al., 2003; Boreen et al., 2003; Lin and Reinhard, 2005; Packer et al., 2003; Rivera-Utrilla et al., 2013). Only limited studies on the photodegradation of micropollutants in WSPs have been published (Jasper et al., 2014; Ryan et al., 2011).

3.1.2. Biodegradation

During biodegradation, organic compounds are degraded by microorganisms (e.g. bacteria, fungi, algae) into simpler chemical structures, sometimes resulting in complete mineralisation (Garcia-Rodríguez et al., 2014). Biologically mediated degradation is a major process occurring in WSPs for removal of pollutants, occurring under aerobic and anaerobic conditions (Garcia-Rodríguez et al., 2014; Li et al., 2013). It has been reported that decomposition of micropollutants in biologically based wastewater treatment systems, such as WSPs, is faster in aerobic, rather than anaerobic, conditions (Garcia-Rodríguez et al., 2014; Hijosa-Valsero et al., 2010b; Jasper et al., 2014). However, even in anaerobic conditions in a particular treatment system, aerobic environments may be present around an algal bloom or near the surface. This coexistence of both aerobic and anaerobic conditions in biological treatment systems (e.g. WSPs) allows for the biodegradation of a range of different types of organic pollutants (Hijosa-Valsero et al., 2010b).

Complex reciprocal biological interactions occur between resident bacteria, algae and other microbial communities in WSP systems and these biological interactions and microbial communities may also play a part in the fate of organic micropollutants within the WSP environment (Coleman et al., 2010). For example, it has been reported that a microalgal-bacterial consortia can enhance

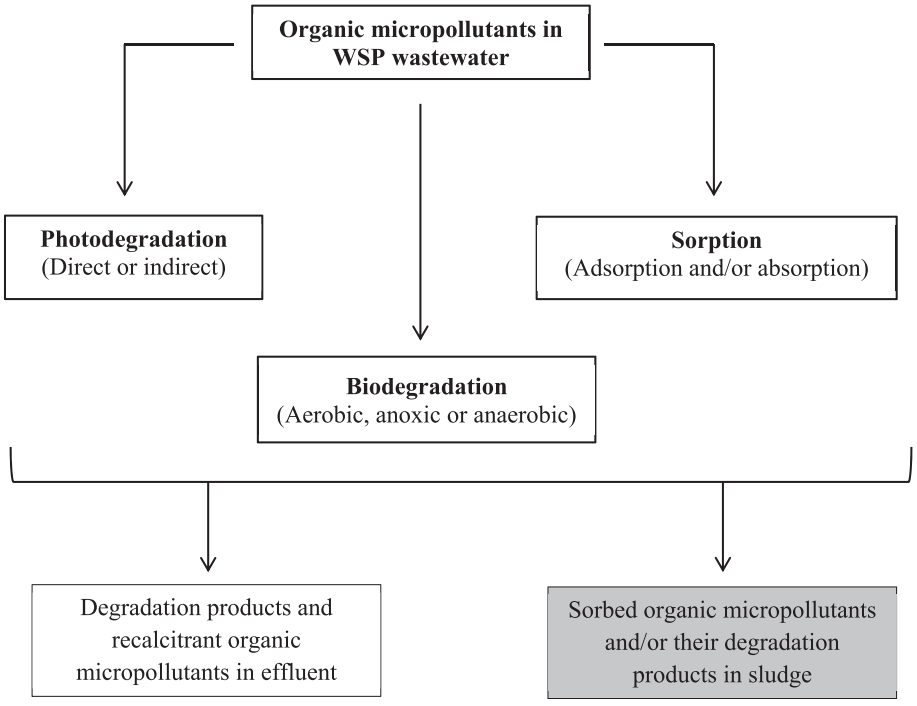

Fig. 1. Removal mechanisms for organic micropollutants in waste stabilisation ponds. Adapted from Garcia-Rodríguez et al. (2014).

the biodegradation of pollutants compared to individual microorganisms, where algal photosynthesis provides oxygen and organic exudates which can then be used by the pollutant-degrading bacteria (Matamoros et al., 2015; Muñoz and Guieysse, 2006). Many studies of micropollutant biodegradation using biological treatment processes have focussed on conventional activated sludge treatment (Verlicchi et al., 2012; Wang and Wang, 2016). However, it is still not clearly understood if the biodegradation of particular micropollutants (e.g. PPCPs) in the activated sludge treatment is attributed to specific microorganisms or microbial community (Wang and Wang, 2016) and no studies have been published on the contribution of microbial communities to micropollutant removal in WSPs, representing a significant knowledge gap in this area.

The advent of next-generation DNA sequencing (NGS) has revolutionized the approach utilized to survey the microbial communities. For instance, sequences from genes containing both conserved and highly polymorphic regions, such as the small ribosomal RNA gene (16S rDNA), are used for massive parallel identification and relative quantification of Bacteria and Archaea, in a method called metabarcoding (Caporaso et al., 2012, 2010). Microbial functional genes associated with certain microbial processes have been used in functional gene arrays to measure gene diversity, relative abundance and activity in natural environments (e.g. marine sediments) (Babbin et al., 2016; Rose et al., 2016). While these techniques have not yet been applied to profile the microbial communities contributing to biodegradation in WSPs, Rose et al. (2016) used functional microbial gene assays to measure the abundance and activity of genes associated with nitrogen removal in a WSP system (one facultative pond and four maturation ponds) in northern Australia. Fernandez-Fontaina et al. (2012) reported that good nitrifying activities increased biodegradation of certain compounds in a pilot −scale nitrifying activated sludge reactor. This co-metabolic biodegradation increase was attributed to the activity of the ammonium monooxygenase enzyme (Fernandez-Fontaina et al., 2012). Thus it is possible that similar interactions between nitrogen removal and micropollutant biodegradation could exist in WSPs.

The biodegradability of micropollutants depends on their bioavailability, with the complexity of the compound structure (e.g. monocyclic or polycyclic) and its functional groups (e.g. halogen groups) playing an important role in determining pollutant biodegradability (Luo et al., 2014). Easily degraded substances include linear compounds with short side chains, unsaturated aliphatic compounds and compounds containing electron donating functional groups. Compounds which are more difficult to degrade include compounds with long and highly branched side chains, saturated or polycyclic compounds and compounds containing sulphate, halogen or electron withdrawing substituents (Luo et al., 2014). For example, it has been suggested that the two chlorine substituents in the structure of diclofenac make it recalcitrant to biodegradation (Hijosa-Valsero et al., 2010b; Kimura et al., 2005). This could account for its reported moderate removal (65%) from a WSP (Hijosa-Valsero et al. (2010b). Since diclofenac is easily photodegraded (Andreozzi et al., 2003; Packer et al., 2003), it is likely that the partial removal of diclofenac from WSPs is due to photodegradation. However, in some cases, there is no clear relationship between the chemical structure and removal. For example, ibuprofen and ketoprofen are considered structurally similar, however they showed different removal efficiencies, including across different wastewater treatment processes, with ibuprofen generally being eliminated more efficiently (Camacho-Muñoz et al., 2012; Luo et al., 2014; Matamoros et al., 2016b).

3.1.3. Sorption

Sorption can occur through both adsorption and absorption, where adsorption is the physical adherence of molecules or ions of the micropollutant onto the surface of a sorbent, while absorption involves incorporation of the pollutant into the sorbent (e.g. algal uptake) (Garcia-Rodríguez et al., 2014). Adsorption of a micropollutant to solids mostly depends on its hydrophobicity. The octanol-water partition coefficient (K_{OW}) of a compound can be used to predict its sorption potential. As a general rule, compounds with $\log K_{OW} < 2.5$ will have low sorption potential, $\log K_{OW}$ values between 2.5 and 4.0 indicate moderate sorption potential, and compounds with $\log K_{OW} > 4.0$ will have strong sorption potential (Luo et al., 2014; Rogers, 1996). Thus, hydrophobic compounds, such as galaxolide and tonalide ($\log K_{OW} > 5$), have been reported to be sorbed to the biomass (i.e. algae) in HRAPs (Matamoros et al., 2015). On the other hand, compounds that are highly or moderately hydrophilic, are unlikely to significantly sorb to the organic matter in pond sediments and will remain in the dissolved phase of the pond where the likely mechanisms of elimination will be biodegradation and/or photodegradation (Hijosa-Valsero et al., 2010b; Matamoros et al., 2015; Ying et al., 2009).

3.2. Pharmaceuticals and personal care products (PPCPs) in WSPs

The major pathway for PPCPs to enter the aquatic environment is through wastewater, after human usage of pharmaceuticals and their excretion in faeces and urine, and after human application of personal care products and their loss to the wastewater system during showering, as well as from disposal of unused medications into the toilet (Luo et al., 2014; Ying et al., 2009). Most of the research on the removal of PPCPs during wastewater treatment has been conducted in conventional activated sludge treatment plants (Fernandez-Fontaina et al., 2012; Kim et al., 2007; Luo et al., 2014; Ternes, 1998; Wang and Wang, 2016). Membrane bioreactors (Clara et al., 2005; Verlicchi et al., 2012) and advanced treatment technologies, such as ozonation (Chen et al., 2012a; Huber et al., 2005; Lee and von Gunten, 2010; Reungoat et al., 2012; Ternes et al., 2003), have also been studied. While there has been relatively little research on PPCP removal in WSPs, this class represents the class of compounds that have been most studied in WSPs, compared to other classes (Camacho-Muñoz et al., 2012; Hijosa-Valsero et al., 2010b; Hoque et al., 2014; Li et al., 2013; Matamoros et al., 2016b; Ying et al., 2009). Constructed wetlands have also become a popular option for PPCP removal in recent years (e.g. Hijosa-Valsero et al., 2010a; Li et al., 2014; Matamoros et al., 2009; Verlicchi and Zambello, 2014; Zhang et al., 2014; Zhu and Chen, 2014).

Comparison of different WWTP configurations in Australia (activated sludge with six lagoons; two oxidation ditches; three bioreactors with UV; two parallel anaerobic lagoons followed by eight aerobic lagoons, with only one lagoon being aerated) showed that, while the lagoon system had the lowest removal efficiencies for the compounds studied (caffeine, carbamazepine, triclosan, gemfibrozil, diclofenac, ibuprofen, ketoprofen and naproxen), it still exhibited good removal efficiencies (>77%) for most of the compounds, except for carbamazepine and gemfibrozil (Ying et al., 2009). Carbamazepine is known to be poorly removed by many types of WWTP (Camacho-Muñoz et al., 2012; Luo et al., 2014; Ternes, 1998; Verlicchi et al., 2012; Wang and Wang, 2016), however, gemfibrozil is typically better removed in more complex WWTP processes. For example, in this study, the removal efficiency for gemfibrozil was only 15% in the lagoon treatment system but 90% in the oxidation ditch WWTP. Poor removals for carbamazepine and gemfibrozil, compared to other PPCPs, have been reported in several other studies of WSP treatment. For example, Camacho-Muñoz et al. (2012) reported poor removals for carbamazepine (10%) and gemfibrozil (20%), as well as propranolol (10%), in a WSP

system, compared to a conventional activated sludge treatment, and a constructed wetland. The authors proposed that the WSP's poor performance could be due to low production of sewage sludge or poor aeration (Camacho-Muñoz et al., 2012). Li et al. (2013) also observed poorest removals for carbamazepine, gemfibrozil and trimethoprim, while other PPCPs (i.e. caffeine, naproxen, ibuprofen, sulfamethoxazole, triclosan, triclocarban and diphenhydramine) were effectively removed (88–100%), in a WWTP consisting of two aerated lagoons in series, followed by a sand filter. In this study, 70%–100% of PPCP removal occurred in the first aerated lagoon, with removals better in the warm season than the cold season. Hoque et al. (2014) also reported higher elimination rates in the summer (15–27 °C) compared to winter (0–5 °C) for ibuprofen, triclosan, galaxolide, tonalide, sulfamethoxazole, and trimethoprim in a WSP system consisting of an aerated lagoon and a settling pond in series, with a HRT of six weeks.

While some studies have found most (>80%) removal of PPCPs occurs in the early stages of the WSP system (Hijosa-Valsero et al., 2010b; Li et al., 2013), other systems that incorporate treatment through constructed wetlands can be even more efficient for overall removal of some PPCPs. For example, Hijosa-Valsero et al. (2010b) found that diclofenac, and methyl dihydrojasmonate were about 15–20% better removed in a combined facultative pond-constructed wetland (FP-CW) system than in a WSP system consisting of two anaerobic ponds, one facultative pond and one maturation pond, although removals of naproxen and ibuprofen were about 10% lower in the hybrid FP-CW system. However, even in the FP-CW system, most of the PPCP removal occurred in the facultative pond, rather than the constructed wetland.

Many pharmaceutically active compounds can be photodegraded, as they usually contain aromatic rings, heteroatoms and other functional groups that allow direct absorption of solar radiation or undergo reactions with reactive species (e.g. nitrate, nitrite or hydroxyl radicals) that promote their photodegradation in natural waters (Boreen et al., 2003; Packer et al., 2003; Rivera-Utrilla et al., 2013). For example, singlet oxygen and dissolved organic matter were shown to play a role in enhancing the photodegradation of the antibiotic norfloxacin under simulated sunlight conditions (Niu et al., 2016). Most studies on the photodegradation of pharmaceuticals have been conducted under laboratory conditions with simulated sunlight, however some studies in WSPs have been conducted. Ryan et al. (2011) observed that the half lives of two common antibiotics (sulfamethoxazole and trimethoprim) were reduced by between two (sulfamethoxazole) and ten (trimethoprim) times in summer sunlit WSPs, compared to sunlit non-wastewater matrices. Effluent organic matter (EfOM) and nitrate were identified as the main photosensitisers in the wastewater matrix, while the natural organic matter (NOM) present in the nearby surface water was not an efficient photosensitiser (Ryan et al., 2011). Jasper et al. (2014) also reported that photodegradation accounted for most of the removal of propranolol and sulfamethoxazole in a pilot-scale open wetland during the summer. However, photodegradation only accounted for approximately 10% of the removal of atenolol and metoprolol and less than 40% of the removal for trimethoprim (Jasper et al., 2014).

Given the high water solubility of most PPCPs, for example ibuprofen, diclofenac, methyl dihydrojasmonate, naproxen, ketoprofen, caffeine, carbamazepine and salicylic acid all have $\log K_{OW}$ between 0.16 and 4.0, sorption to sludge is unlikely to be a major removal mechanism for these chemicals. However, the importance of multiple removal mechanisms (e.g. biodegradation, sorption and photodegradation) in PPCP removal in WSPs was demonstrated by Matamoros et al. (2016b), who compared the performance of a WSP, a CW, an extended aeration system and a rotating biological contactor for the removal of several pharmaceuticals, sunscreen compounds, fragrances and antiseptics. The WSP system (two facultative ponds in parallel, followed by a maturation pond) had the highest overall removal efficiency, attributed to the coexistence of multiple removal mechanisms, as well as the high HRTs (20–30 days), in the WSP system. In this study, Matamoros et al. (2016b) found a strong relationship (p value <0.05) between biodegradable compounds, such as caffeine, paracetamol, ibuprofen, methyldihydrojasmonate, naproxen, benzotriazole, methyl benzotriazole, tributyl phosphate and triphenyl phosphate, and NH_4-N removal for all treatment technologies (activated sludge, CW, WSP and rotating biological contactor). In contrast compounds that have been reported to be removed by sorption onto organic matter (e.g. galaxolide and tonalide) or photodegradation (triclosan and diclofenac) did not show any correlation with NH_4-N removal. Recalcitrant compounds such as carbamazepine or tris(2-chloroethyl) phosphate also did not show any correlation with NH_4-N removal (Matamoros et al., 2016b). No seasonal variation was observed in the removal of the organic pollutants, possibly due to the smaller difference in temperature between the warm and cold seasons of the Mediterranean climate, compared to other studies.

None of the studies discussed above addressed the formation of transformation products in WSP systems. Yet transformation products are important to identify because some transformation products may be more toxic than the parent compound. For example, the photodegradation products of naproxen are more toxic than the parent compound (Packer et al., 2003; Rivera-Utrilla et al., 2013). Photodegradation of carbamazepine produces a number of products, including acridine which is mutagenic and carcinogenic (Rivera-Utrilla et al., 2013). Most studies of transformation products in wastewater have been conducted under controlled laboratory conditions and many have used advanced oxidation process (e.g. Lee and von Gunten, 2016; Deeb et al., 2017) while others have been conducted in pure water or natural waters (e.g. Packer et al., 2003; Rivera-Utrilla et al., 2013). One of the challenges in identifying transformation products in wastewater has been the availability of analytical methods sensitive enough to allow detection of low concentrations of compounds in a complex matrix. Recent advances in analytical instrumentation such as the use of high resolution mass spectrometry (HRMS) has allowed the analysis of a wide range of known and unknown compounds in complex samples including parent compounds, metabolites and transformation products (Deeb et al., 2017 and references therein). However, there is still a significant knowledge gap with respect to occurrence data of transformation products in wastewater systems, in particular in WSPs.

3.3. Endocrine disrupting compounds in WSPs

EDCs can disrupt developmental and reproductive functions in wildlife and humans (Coleman et al., 2010; Pessoa et al., 2014), and include compounds such as natural and synthetic hormones, alkyl phenols (Andersen et al., 2003; Kinani et al., 2010; Lagana et al., 2004) and polycyclic aromatic hydrocarbons (PAHs) (Busetti et al., 2006). The removal of EDCs from wastewater can be studied by measuring the concentrations of individual EDCs or by measuring the estrogenic activity of the wastewater using sensitive and specific *in vitro* bioassays, which detect compounds that share a specific mode of action (Dagnino et al., 2010; Gomez et al., 2007). The removal of EDCs in conventional WWTPs, such as activated sludge plants, has been well studied (Andersen et al., 2003; Clara et al., 2005; Kim et al., 2007; Svenson et al., 2003; Tan et al., 2008, 2007; Van Buynder et al., 2009), however, there has only been limited research on removal of EDCs in WSPs.

Removal of steroidal hormones in WSPs is typically high. For

example, Coleman et al. (2010) found androgenic steroid removals in two Australian WSPs that ranged between 93% (androstendione) to 100% for androsterone, eticholanolone and 17β-trenbolone. While removal of estrogenic hormones estrone (E1) and estriol (E3) was lower, 26% and 82%, respectively. Multiple studies highlight the importance of long HRTs for EDC removal (Gomez et al., 2007; Servos et al., 2005). Gomez et al. (2007) found that the removal of estrogenic activity (ERα bioassay) in two French WSPs with long retention times (78 days and 43 days) was more than twice that of a WWTP consisting of a trickling filter and post tertiary ponds with a total HRT of 20 days, even though both treatment systems had high COD removals. Servos et al. (2005) has also shown that lagoons with long residence times had consistently high removals (80%–98%) of hormones, while Pessoa et al. (2014) observed much lower removals (44%–80%) of estrogenic hormones in Brazilian WSPs with shorter HRTs (21, 18.4 and 26.4 days).

Although longer HRTs can enhance the removal of estrogens, unfavourable redox conditions (i.e. poor aeration), and low biological activity, can hinder biodegradation (Qiang et al., 2013). Both activated sludge and constructed wetlands were found to better remove six EDCs including E1, 17β-estradiol (E2), E3, 17α-ethinyl estradiol (EE2), bisphenol A (BPA) and 4-nonylphenol (NP) than a WSP system comprised of a primary sedimentation pond, followed by a facultative pond, especially in winter (mean temperature 7 °C). The poor performance of the WSP system was attributed to the decreased activity of algae and heterotrophic bacteria at the colder temperature (Qiang et al., 2013). Octylphenol has also been found to have higher removal in pilot-scale HRAPs (Matamoros et al., 2015), than conventional WSPs (Matamoros et al., 2016b), again possibly related to higher activity of algae.

Another consideration for EDC removal is consideration of partitioning between the solid and liquid phase. Dagnino et al. (2010) found that suspended solids contributed up to 80% of the total ERα and AhR activities of WSP treated effluent, a much higher proportion than the 20% contribution of the suspended solids to the total estrogenic activity in effluents from a trickling filter or activated sludge plant with biofilter. Despite this, total removal efficiencies (dissolved and suspended solid) for ERα and AhR activities were higher than 90% for the WSP plant; close to 90% for the activated sludge plant with biofilter plant and less than 65% for the trickling filter.

Based on previous studies on other wastewater treatment processes (De Mes et al., 2005; Coleman et al., 2010), the likely mechanisms for the removal of EDCs is WSPs may include adsorption to particulate matter and sedimentation; both direct and indirect photolysis and aerobic and anaerobic biodegradation. Sorption to both inorganic and organic suspended solids as well as the pond biomass may represent important pathways removal of EDCs in WSPs, however the direct contribution of pond organisms, such as phytoplankton to EDC removal is not well understood. Therefore, additional research should be conducted to evaluate the importance of biomass adsorption for removal of EDCs in WSPs.

3.4. Pesticides in WSPs

Agricultural run-off of pesticides from crops has been reported as the main source of pesticides in the aquatic environment (Matamoros et al., 2015). Thus pesticide concentrations in WSPs are more likely to result from agricultural run-off or spray drift from aerial application from nearby farms, rather than domestic wastewater, where only small contributions from personal insect repellents, or disposal of unused pesticide solutions would be expected. Consistent with these proposed sources, concentrations of some pesticides in agricultural run-off have been reported to be as high as several hundred μg/L (Davis et al., 2013; Schulz, 2004), but Matamoros et al. (2015) reported concentrations of diazinon, atrazine and 2,4-dichlorophenoxyacetic acid (2,4-D) to be less than 0.5 μg/L in urban wastewater. Because of this, most studies of pesticide removal in low cost treatment processes have considered treatment of agricultural run-off, rather than domestic wastewater. There are a significant number of studies on pesticide removal using constructed wetlands (Matamoros et al., 2007; Moore et al., 2002; Runes et al., 2003; Schulz, 2004; Schulz and Peall, 2001), as well as a combination of non-vegetated open ponds and vegetated constructed wetlands (Rose et al., 2008, 2006; Schulz et al., 2003), with findings that aquatic vegetation enhances pesticide removal compared to non-vegetated open water systems, but there is very little information on the removal of pesticides in conventional WSPs. For example, Matamoros et al. (2016b) was not able to assess seasonal pesticide removal in a study of a Spanish WSP because the pesticides were only detected in the primary effluent wastewater at very low concentrations in the warm season and were not detected at all during the cold season.

A few laboratory or pilot-scale limited studies have focussed on the use of algal systems for pesticide removal (Matamoros and Rodríguez, 2016; Matamoros et al., 2015). In the laboratory study, the detection of transformation products for alachlor, lindane and endosulfan suggested that biodegradation was their major route of removal (Matamoros and Rodríguez, 2016). However, photodegradation or algal uptake could not be ruled out for other pesticides, with no transformation products detected. In a study of pilot-scale HRAPs, improved removal for diazinon, atrazine and 2,4-D was observed for the longer HRT of 8 days compared to 4 days (Matamoros et al., 2015).

While studies of the removal of pesticides, and the formation of transformation products, in WSP are very limited, this is not a high priority area for future research since few pesticides enter WSP through domestic wastewater and the concentrations of pesticides in WSP are very low.

3.5. Perfluorinated compounds in WSPs

Perfluorinated compounds (PFCs) are a class of emerging persistent organic pollutants consisting of a fully fluorinated hydrophobic alkyl chain tethered to a terminal hydrophilic moiety (Arvaniti and Stasinakis, 2015; Lindstrom et al., 2011; Xiao, 2017). PFCs are persistent in the environment, bioaccumulative and potentially toxic to humans and wildlife (Arvaniti and Stasinakis, 2015; Lindstrom et al., 2011; Sepulvado et al., 2011; Xiao, 2017). For many years, PFCs have been used in a range of industrial and household applications, such as nonstick coatings, stain-repellent fabrics, paper packaging products, surfactants and firefighting foams (Arvaniti and Stasinakis, 2015; Sepulvado et al., 2011). Due to their widespread use, PFCs have been widely detected in the influent, effluent and sludge/biosolids at urban WWTPs worldwide (Arvaniti and Stasinakis, 2015; Sepulvado et al., 2011). PFCs are resistant to degradation and are difficult to remove during wastewater treatment (Arvaniti and Stasinakis, 2015). Two of the most widely studied and most frequently detected PFCs are perfluorooctanoic acid (PFOA) and perfluorooctane sulphonate (PFOS) (Arvaniti and Stasinakis, 2015; Lindstrom et al., 2011). PFOS and PFOA concentrations of up to several hundred ng/L and several hundred ng/g have been reported in wastewater samples and sludge, respectively (Appleman et al., 2014; Arvaniti and Stasinakis, 2015; Dickenson and Higgins, 2016).

While some studies on the removal of PFCs from wastewater using conventional and advanced treatment processes have been published (Appleman et al., 2014; Arvaniti and Stasinakis, 2015 and references therein; Dickenson and Higgins, 2016), no studies of the removal of PFCs in WSPs have been published. The most effective

treatments for PFCs to date have been sorption to the sludge (Arvaniti and Stasinakis, 2015), granular activated carbon and anion exchange (Appleman et al., 2014; Dickenson and Higgins, 2016), reverse osmosis (Appleman et al., 2014; Dickenson and Higgins, 2016), and a number of photolytic methods (Arvaniti and Stasinakis, 2015 and references therein, Hori et al., 2004). While photodegradation of PFCs in WSPs may be possible, to date most photolysis experiments have been conducted under laboratory conditions with pure water and high concentrations of PFCs. It is unlikely that complete degradation of PFCs would be achieved under full-scale conditions in WWTPs (Arvaniti and Stasinakis, 2015). Therefore, while partial degradation of PFCs in sunlit WSPs may occur as a result of photolysis, it is most likely that the bulk of the PFCs will persist in the wastewater and accumulate in the bottom sludge layer. Due to the environmental and health risks of these persistent pollutants, further studies of their partitioning between the wastewater and sludge phases and of strategies for their improved degradation in WSPs are essential. Furthermore, several studies have shown that PFOS, PFOA and a few other perfluoroalkyl compounds can be formed during biological wastewater treatment (Xiao, 2017 and references therein). Non-ionic compounds, such as *N*-ethyl perfluorooctane sulphonomidoethanol (EtFOSE) and 8:2 fluorotelomer alcohol (FTOH) have been proposed as potential precursors to their formation (Xiao, 2017). However, the reported yields of PFOA and PFOS from these two precursors are low (1–2%), suggesting the presence of other unidentified compounds which are readily biodegradable during wastewater treatment (Xiao, 2017). Therefore, identification of these unknown precursor compounds is vital to estimating the secondary formation of PFOS and PFOA during wastewater treatment and controlling the discharge of PFOA and PFOS in treated wastewater into the environment.

3.6. Other organic micropollutants in WSPs

Other organic micropollutants that have been reported to be present in wastewater include artificial sweeteners, such as sucralose, acesulfame, cyclamate and saccharin (Busetti et al., 2015; Hoque et al., 2014; Lange et al., 2012; Van Buynder et al., 2009), illicit drugs (e.g. Jones-Lepp et al., 2004; Kasprzyk-Hordern et al., 2009; Yadav et al., 2017; Zuccato et al., 2008), flame retardants, corrosion inhibitors, plasticizers and surfactants (Busetti et al., 2015; Luo et al., 2014; Matamoros et al., 2016a, 2016b, 2015; Van Buynder et al., 2009).

Acesulfame and sucralose are persistent and have been suggested as anthropogenic markers for wastewater impacted environments (Busetti et al., 2015; Lange et al., 2012). They are not removed in conventional WWTPs (Busetti et al., 2015; Lange et al., 2012). In contrast, removals of more than 90% have been reported for cyclamate and saccharin during secondary or tertiary wastewater treatment in several European WWTPs (Lange et al., 2012). To our knowledge, the only study of artificial sweeteners in WSPs to date is that by Hoque et al. (2014), who reported that sucralose was not removed during WSP treatment.

Illicit drugs (e.g. cocaine, amphatamines, cannabinoids, opiods and their metabolites) are considered to be emerging environmental pollutants and knowledge of their concentrations in raw wastewater can provide valuable information on the usage of illicit drugs in local communities, while assessing their concentrations in treated wastewater provides information on the removal efficiency of the wastewater treatment process (Kasprzyk-Hordern et al., 2009; Thai et al., 2016; van Nuijs et al., 2011; Yadav et al., 2017). While no studies on the removal of illicit drugs in WSPs have been published, a few studies on the removal of illicit drugs in other wastewater treatment systems have been published. Removal of illicit drugs, such as cocaine, benzoylecgonine, nor benzoylecgonine, cocaethylene, amphetamine, methamphetamine, 3,4-methylenedioxy-methamphetamine (MDMA), methadone, 2-ethylidine-1,5-dimethyl-3,3-diphenylpyrrolidine (EDDP), codeine, morphine, tetrahydrocannabinol (THC) and 11-nor-9-carboxy-Δ9-tetrahydrocannabinol (THC-COOH) have been studied in activated sludge and trickling filter wastewater treatment (Yadav et al., 2017 and references therein). Activated sludge treatment was found to be more efficient in removing illicit drugs than trickling filters most likely because activated sludge processes provide conditions (e.g. more active microbial biomass content) for more efficient degradation of micropollutants compared to trickling filters. The higher oxygen input due to mechanical aeration in the activated sludge treatment would also enhance volatilisation (Kasprzyk-Hordern et al., 2009; Baker and Kasprzyk-Hordern, 2013; Yadav et al., 2017 and references therein). For example Baker and Kasprzyk-Hordern (2013) found that activated sludge removed 91% of cocaine, 99% of amphetamine and 39% of methamphetamine, while the removal efficiency of trickling filters for cocaine, amphetamine and methamphetamine was 37%, 89% and 25%, respectively. More recently, Mackul'ak et al. (2015) investigated the use of aquatic plants in tertiary treatment of wastewater effluent for the removal of several illicit and prescription drugs, using laboratory-scale reactors. Removals ranged from 25% to 93% for codeine, 60%–85% for methamphetamine, 28%–82% for benzoylecgonine, 63%–87% for methadone, 48%–70% for tramadol and 8%–58% for oxazepam, depending on the type of plant used (Mackul'ak et al., 2015).

Reported percentage removal efficiencies of benzotriazole and benzothiazole corrosion inhibitors by conventional WW treatment processes are variable (Reemtsma et al., 2002; Voutsa et al., 2006), and similar variable removal has been observed in WSPs (Matamoros et al., 2016b). Removal of some benzotriazoles (benzotriazole and 5-methylbenzotriazole) was better in a pilot-scale HRAP, but removals of benzothiazole and hydroxybenzothiazole were poorer (Matamoros et al., 2015). Benzotriazoles and benzothiazoles are highly water soluble (i.e. $\log K_{OW} < 2.5$) (Busetti et al., 2015), and so sorption to sludge is unlikely. A recent study of benzotriazole and benzothiazole degradation in constructed wetlands and by artificial sunlight found that most degradation was caused by microbial action in the wetlands, with artificial sunlight only effective in the presence of titanium oxide (Felis et al., 2016). Thus, it is feasible that these compounds would also be biodegraded in WSPs.

Comparisons of the removal of the plasticizers and fire retardants, tris(2-chloroethyl)phosphate, tributyl phosphate and triphenyl phosphate, in a range of WWTPs (WSP, HRAP, activated sludge and constructed wetland) showed very variable removal between compounds, with no overall pattern (Matamoros et al., 2015, 2016b), suggesting that these compounds must be studied individually to understand removal mechanisms.

With the potential environmental and health risks of discharge or reuse of treated wastewater containing illicit drugs, future studies of their behaviour in WSP treatment and methods for their improved removal are essential. Similar studies of artificial sweeteners, corrosion inhibitors, plasticizers and flame retardants are important but of lower priority.

3.7. Summary of future research directions for removal of organic micropollutants in WSPs

Only a limited number (40) out of hundreds of organic micropollutants identified in wastewaters have been studied in WSPs. Published research on the elimination of organic micropollutants in conventional WSPs has been mainly focussed on PPCPs and EDCs, with limited studies on the removal of other classes of chemicals.

No studies have been published on the fate of illicit drugs or perfluorinated compounds in WSPs and limited data was found on the removal of pesticides in conventional WSPs. Thus, there is considerable scope for the study of removal of these classes of organic micropollutants in WSPs, as well as better identification of health risk and degradation mechanisms through the identification of transformation products and in the case of PFCs the identification of PFOS and PFOA (and other perfluoroalkyl compounds) precursors.

Organic micropollutants that are removed from the WSP wastewater effluent through sorption to the sludge can create potential issues for sludge disposal and be potentially remobilised back into the water column. Therefore, studies of sorption mechanisms for removal of organic pollutants should also include an assessment of their desorption/dissolution potential. Organic pollutants that are transformed *via* either photodegradation or biodegradation may produce transformation products that are more (or less) toxic than the parent compound. Therefore identification of transformation products and assessment of their toxicity should be undertaken in order to understand the environmental and health impact of the treated wastewater for discharge or reuse.

It is still not clearly understood if biodegradation of micropollutants in WSPs is attributed to specific microorganisms or microbial communities, therefore research should be undertaken to profile the microbial communities in WSPs to better understand the biodegradation pathways. Eventhough photodegradation has been cited as one of the key mechanisms for micropollutant removal, only limited studies have been published on photodegradation in WSPs.

Another future research area is to examine a combination of different biological wastewater treatment processes, e.g. a hybrid of an unvegetated WSP and a CW or a combination of conventional treatment with a WSP, for removal of the more recalcitrant organic pollutants.

4. Conclusions

WSPs have proven to be an effective and low-cost technology for wastewater treatment, particularly in developing countries or small regional communities. However, the performance of WSPs in terms of organic micropollutant removal is highly variable and depends on many different factors (e.g. pond configuration, composition of wastewater, environmental conditions and the type of chemical). Seasonal variations in removal performance appeared to be mostly evident in climates with cold winters (ambient temperatures ≤ 4 °C) compared to more temperate climates with warmer winter temperatures ($\geq$11 °C).

Only a limited number (40) out of hundreds of organic micropollutants identified in wastewaters have been studied in WSPs. Therefore, there is a wide scope for future research on WSP removal of organic pollutants, particularly emerging compounds, such as illicit drugs and perfluorinated compounds. There is a need for further studies of the formation and fate of transformation products of organic micropollutants in WSPs to identify whether the transformation products persist in the wastewater effluent, are further degraded to other transformation products (or eventually to carbon dioxide) or are sorbed onto the sludge. This knowledge would then allow full environmental risk assessment of the organic micropollutants and their transformation products.

Better removal of recalcitrant organic pollutants may also be possible by combining different biological wastewater treatment processes, e.g. a hybrid of an unvegetated WSP and a constructed wetland or a combination of conventional treatment with a WSP. Ultimately, the degree of chemical removal required for WSP effluent depends on the desired end use, such as discharge to natural waters or beneficial reuse (e.g. irrigation of crops or parks). In some cases, additional treatment of the WSP effluent may be required to produce fit-for-purpose treated wastewater, meeting the relevant discharge or reuse guidelines.

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