Sand and Sand-GAC Filtration Technologies in Removing PPCPs: A Review

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Abstract

Concerns have been raised about the risks that pharmaceuticals and personal care products (PPCPs) in aquatic environments posed to humans and the environment. In recent years, sand filtration has been used to potentially remove these emerging contaminants from water. However, there has been no review of the effectiveness of this technology to date. This paper presents a brief introduction of sand filtration types, reviews the current progress in PPCP removal through sand filtration, and discusses the mechanisms behind this process and the combination of granular activated carbon (GAC) and sand as an enhanced sand-GAC filtration technology. Sand filtration achieves a reasonable but highly variable degree of PPCP removal. Biodegradation and adsorption are the two main mechanisms of PPCP removal, in particular the biodegradation since adsorption capacity of sand is relatively low. Other processes, such as bio-sorption and indirect adsorption, may also contribute to PPCP removal. To compensate for the inadequate PPCP removal through sand filtration, porous GAC has been combined with sand to develop sand-GAC filtration technologies. Serial, dual, and sandwich filters have been investigated, and significant removal enhancement has been observed, due to the strengthened adsorption capacity, suggesting the applicability of these variants. Future research focus, such as investigating the influence of different operational conditions on sand filter performance, obtaining a deeper understanding of the various removal mechanisms, and investigating of long-term performance of the filter used for PPCP removal, are suggested.

Keywords: PPCPs; Sand filtration; Mechanisms; GAC; Removal

1. Introduction

There have been concerns about pharmaceuticals and personal care products (PPCPs) for decades (Daughton and Ternes,1999). The term PPCPs comprises a large variety of emerging environmental contaminants, such as antibiotics, hormones, anti-inflammatory drugs, antiepileptic drugs for pharmaceuticals, and antimicrobial agents, synthetic musks, insect repellents, preservatives, and sunscreen ultraviolet (UV) filters for personal care products (Daughton and Ternes, 1999; Liu and Wong, 2013). Generally, effluents released from wastewater treatment plants (WWTPs) are considered as an important source of PPCP discharges into the environment (Chen et al., 2012). In recent years, the effectiveness of various wastewater treatment technologies (e.g., activated sludge treatment, biological nutrient removal processes, UV treatment, Fenton process, constructed wetlands, etc.) in removing PPCPs has been investigated, but the effectiveness of such technologies varies greatly, and some technologies are not cost-effective (Kim and Tanaka, 2009; Krishnan et al., 2021; Li et al., 2017, 2012; Sui et al., 2011; Zhang et al., 2019). Owing to the inadequate removal efficiency of WWTPs, PPCPs have been detected in various water sources (e.g., surface water, groundwater, drinking water, and seawater) around the world and it has become a new environmental problem globally (Caldas et al., 2019; Fu et al., 2019; Kallenborn et al., 2018; Li et al., 2016; Moldovan, 2006; Sengar and Vijayanandan, 2022). Despite the relatively low concentrations (ng/L-μg/L) of PPCPs in the aquatic environments, their persistence, toxicity and other related problems (e.g., antibiotic resistance) may pose potential risks to human health and other organisms (Li et al., 2016; Narayanan et

al., 2022; Sauvetre and Schroder, 2015; Sengar and Vijayanandan, 2022).

Sand filtration is one of the earliest water treatment technologies and remains as an important water purification process worldwide. Pure sand filtration for water treatment can be classified into either slow sand filtration (SSF) or rapid sand filtration (RSF), depending on the filtration rate (Bar-Zeev et al., 2012; Campos et al., 2002). In recent decades, a variant of SSF called biosand filtration (BSF) has emerged and has been used to treat household drinking water (Elliott et al., 2011; Pompei et al., 2017). These types of sand filtration have shown great potentials in removing suspended solids, pathogenic microorganisms, and traditional and emerging organic pollutants (Asami et al., 2016; D'Alessio et al., 2015; Escolà Casas and Bester, 2015; Haig et al., 2011; Nakada et al., 2007; Wang et al., 2021). Although other advanced water treatment technologies (e.g., biomembranes and advanced oxidation processes) are now widely used, sand filtration has received significant attention in recent decades owing to its simplicity, compatibility, low cost (for BSF), and relatively low chemical and electricity requirements and high water treatment volumes (for SSF and RSF) (Haig et al., 2014, 2011; Pompei et al., 2017; Wang et al., 2021). Till now, sand filtration has been successfully employed in the purification of a variety of water sources, including wastewater, surface water, ground water, rain water, etc. (Andreoli and Sabogal-Paz, 2020; Moreira Neto et al., 2012; Racar et al., 2019; Sobsey et al., 2008). In addition, to enhance the removal of organics (including PPCPs) in water treatment plants, granular activated carbon (GAC) has been combined with sand in various filtration technologies (e.g., Bauer et al., 1996; Gidstedt et al., 2022; McKie et al., 2016). Other materials, such

as vegetal materials (e.g., woodchips), clay, graphene, graphene oxide, zero-valent iron, anthracite, kinetic degradation fluxion media, and crushed limestone/brick, have also been employed together with sand for filtration, and some have exhibited good PPCP performance (Clyde et al., 2021; Fu et al., 2019; Kulkarni et al., 2019; Majdi et al., 2019; Rizzo et al., 2015; Shrestha et al., 2014; Vu and Wu, 2022; Zaman et al., 2017; Zhang et al., 2016; Zhao et al., 2019).

Studies on the removal of PPCPs through sand filtration are relatively few. Wang et al. (2021) reviewed the biodegradation potential of RSF for organic micropollutant removal from drinking water. To the best of the authors' knowledge, the PPCP removal performance of different sand filtration systems is yet to be comprehensively reviewed. Therefore, it is meaningful to look into and summarise the past and current trends in sand filtration processes for PPCP removal. This review describes the SSF, BSF and RSF technologies, their application in PPCP removal, the mechanisms involved in the PPCP removal processes, and the effectiveness of combing of GAC with sand filtration for PPCPs removal enhancement. In particular, the paper focuses on sand-GAC filtration as it is a widely used practice globally. Finally, suggestions for future research and development of these processes are also proposed and highlighted.

2. Overview of sand filtration

2.1 SSF

The first application of SSF as a mean of water treatment dates back to year 1804 when John Gibb designed and built a slow sand filter for his bleachery and sold the surplus treated water to the public (Huisman and Wood, 1974). For over two centuries,

it has remained an effective water treatment technology in both small and large community water supplies (Haig et al., 2011). Practically, SSF can either be applied as a tertiary stage in water treatment processes or can be used as an efficient single-stage treatment method for raw water within a certain water quality range, especially in low- and middle-income countries (LMICs), making it compatible and flexible (Bowles et al., 1983; Ellis and Wood, 1985; Matamoros et al., 2009; Pompei et al., 2017). SSF uses quartz sand to purify contaminated water. Traditional slow sand filters used in large treatment plants operate in continuous mode to meet large water treatment demands and the sand bed remains wet throughout operation. A schematic representation of a typical SSF filter is shown in Fig.1. In the filter, one gravel layer supports the sand media in the filter, and the treated water flows out through the filter underdrain. A thin, slimy, gelatinous biofilm, called *schmutzdecke*, grows at the top of the sand layer and plays an important role in the water purification. Before formal operation can commence, a maturation stage is usually required to allow the *schmutzdecke* to form.

2.2 BSF

Developed by David Manz at the University of Calvary in the 1990s, BSF, as a variant of SSF, has been successfully implemented as a small-scale, point-of-use (POU) technology for removing microbes from drinking water (Kennedy et al., 2013; Sobsey et al., 2008). Till now, BSF has been widely promoted by several organisations (e.g., Centre for Affordable Water and Sanitation Technology (CAWST)) and over 300,000 BSFs filters have been installed in more than 69 countries (Andreoli and Sabogal-Paz, 2020). Compared to the continuous operation mode of SSF, BSF is an intermittently-

operated slow sand filter. A schematic representation of a BSF filter is shown in Fig.1. Water enters the filter through a flow diffuser and *schmutzdecke* grows on the top of the sand media over time. Like SSF, a maturation stage is also needed for BSF. It is considered a cost-effective household water purification technology and is mainly used in LMICs.

2.3 RSF

Compared to the SSF, RSF is used as a tertiary polishing water treatment technology. The concept of RSF was brought out in USA at the end of the 19th century (Srivastava and Chattopadhyay, 2022). Unlike SSF and BSF, RSF is generally considered to lack biofilm layer on filter media (*schmutzdecke*) and is primarily used to remove large suspended solids through physical processes (e.g., size exclusion) (Srivastava and Chattopadhyay, 2022). However, biological processes may also occur in the RSF system, helping to eliminate contaminants (Wang et al., 2021). As this treatment method uses a high filtration velocity, significant amounts of debris can accumulate in a short period of time, leading to the need for frequent backwashing (Arndt and Wagner, 2003). Therefore, the operational costs of RSF are higher than those of SSF. In practice, RSF is always coupled with other technologies, such as coagulation, flocculation, or UV treatment (Asami et al., 2016; Berg et al., 1968; Heinonen-Tanski et al., 2003).

2.4 Comparison of sand filtration types

The design/operational parameters and properties of the sand media used for SSF, BSF and RSF differ. Table 1 lists some typical operational parameters. It should be

noted that the filtration design/operational parameters are closely aligned with the needs (e.g., feed water quality, financial budget, treatment requirements) and may vary considerably under real circumstances. Generally, the supernatant water in a SSF filter is 100~150 cm deep and the sand media depth is 0.6~1.2 m (Huisman and Wood, 1974; Young-Rojanschi and Madramootoo, 2014). In contrast, the standing head and sand depth in BSF are reduced to 5~20 cm and 0.4~0.55 m, respectively, owing to its small-scale configuration. For RSF, the World Health Organization (WHO) recommends a supernatant water height of 150~200 cm and a sand media depth of 0.5~1.0 m. In addition, the SSF filtration rate is usually within the range of 0.1~0.3 m/h (2.4~7.2 m/d) (Campos et al., 2002) and a retention time of 1~48 h is recommended by CAWST for BSF. However, unlike the slow filtration rates of SSF (cm/h) and BSF, RSF typically employs a much faster filtration rate of 100~475 m/d or 5~30 m^3/h (Arndt and Wagner, 2003; Bar-Zeev et al., 2012).

The effective size and uniformity coefficient are the two most important properties of sand media. The effective size (D_{10}) is the diameter at which 10 % of the sand's mass is comprised of particles with a diameter less than this value, whereas the uniformity coefficient is the ratio of D_{60}/D_{10}. Generally, fine sand is used in the SSF, and the effective size of the sand is 0.1~0.3 mm, with a uniformity coefficient of around 3 (Campos et al., 2002). For BSF, CAWST suggests the use of fine sand with an effective size of 0.15~0.20 mm and a uniformity coefficient of 1.5~2.5. In contrast, the effective size for RSF is usually greater than 0.55 mm with a uniformity coefficient of less than 1.5 (Casey and Casey, 1997). However, the fine sand used in SSF may cause quick

clogging (Mendoza-Espinosa and Stephenson, 1999). In recent years, coarse sand with an effective size of more than 0.3 mm has been used in some SSF studies. Table 2 lists the sand grain size/effective size and uniformity coefficient used in some previous studies.

Besides the operational mode, cleaning strategies differ as well. As RSF employs a high filtration velocity (m/h), backwashing is frequently required to avoid clogging (Wang et al., 2021). But backwashing usually consumes large quantities of clean water. In contrast, no backwashing is required for SSF and BSF, but scraping and replacement of the sand bed, which is beneficial for water-shortage areas (Huisman and Wood, 1974; Lantagne et al., 2006). Generally, sand filtration technologies share the advantages such as cost-effectiveness (relatively cheaper operation/maintenance costs), configuration simplicity, low chemical and electricity requirements, and practical compatibility (Li, 2019). In addition, a large proportion of pathogenic microorganisms, including bacteria, protozoan oocysts, cercariae and schistosomes, can be eliminated, although such processes primarily occur in SSF and BSF (Elliott et al., 2011; Escolà Casas and Bester, 2015; Haig et al., 2011; Srivastava and Chattopadhyay, 2022; Wang et al., 2021).

3. Removal of PPCPs through sand filtration

3.1 Overview of PPCP removal

The removal of PPCPs though sand filtration has not received as much research attention as that received by other treatment processes. Table 3 lists some published research on PPCP removal through sand filtration. These studies cover various types of contaminated water, including wastewater effluents, synthetic wastewater/ rainwater,

surface/reservoir water and tap water.

As shown in Table 3, the removal of PPCPs varies significantly, from negative (-186%) to full removal (100%). Negative removal is not uncommon in water treatment processes and can usually be ascribed to the desorption of molecules from the particles, water evaporation or quantitative errors caused by low concentrations (Li et al., 2017; Lin et al., 2016; Nakada et al., 2007). It is shown from Table 3 that some PPCPs are recalcitrant to sand filtration, such as carbamazepine, removal of which ranged from negative to below 25%, regardless of the filter type, influent concentration, inflow type, filtration rate and experimental scale. Carbamazepine is a compound with low biodegradability and its ineffective removal can be attributed to weak biodegradation and insufficient adsorption onto the surfaces of sand grains. In contrast, effective removal through sand filtration has been found for other PPCPs, such as ibuprofen. Apart from one case of low removal through RSF (30.1%, Nakada et al., 2007), studies have shown that ibuprofen was effectively removed (> 90%) through sand filtration, regardless of the filter types and experimental conditions (Nakada et al., 2007; Pompei et al., 2017, 2019; Zearley and Summers, 2012). It could be that ibuprofen's susceptibility to biodegradation facilitates its effective elimination during filtration. Other PPCPs with high removal include methylparaben, propranolol tylosin, etc.

Several researchers have investigated the elimination of estrogenic compounds, an important category of PPCPs, during filtration processes (Table 3; D'Alessio et al., 2015; Haig et al., 2015; Kennedy et al., 2013; Nakada et al., 2007). Haig et al. (2016) compared continuous SSF bioaugmented by three estrogen-metabolising isolates (E1,

E2, and E3) with nonaugmented SSF to remove those three estrogens (at ng/L concentrations in the influent). With bioaugmentation, the removal of E1, E2 and E3 were 79.46%, 34.58% and 11.66%, respectively. In contrast, the nonaugmented SSF achieved corresponding removal of 2.08%, -66.66% and -11.60%, respectively. These results highlight the dramatic variability in the removal of different compounds. However, poor removal of E1, E3 and EE2 (around 10~20%) were observed during BSF of influent with a concentration of 5 mg/L (Kennedy et al., 2013). Although larger errors can be expected at trace-level influent concentrations, the variations in removal behaviour among structurally similar or related compounds merit further investigation.

Table 3 indicates that the removal achieved for a given compound varies with the influent concentration, filtration rate, filter type and experimental scale. For instance, three contrasting studies that quantified the removal of triclosan yielded the following removal: 74.2% (25 μg/L, 5~20 cm/h or 1.2~4.8 m/d) through laboratory-scale SSF (Li et al., 2018); ⩾90% (190 ± 42 ng/L, 1.2, 2.4 m/h or 28.8, 57.6 m/d) through laboratory-scale RSF (Zearley and Summers, 2012); and 25.2~52.5% (158~360 ng/L, 110 m/d) through full-scale RSF (Nakada et al., 2007). Similarly, significant differences have also been observed for other PPCPs (e.g., acetaminophen, caffeine, diclofenac, gemfibrozil, naproxen, sulfamethoxazole and trimethoprim, Fig. 2). Therefore, it can be assumed that the PPCPs removal through sand filtration is influenced by the operational conditions, initial influent concentrations and filter capacity, and the removal effectiveness of specific PPCPs through sand filtration is inconsistent.

3.2 Comparison between SSF, RSF and BSF

Overall, RSF is less effective in removing PPCPs than SSF and BSF. Both SSF and RSF use the continuous filtration mode; however, as RSF employs a much faster filtration rate, theoretically, PPCPs experience a shorter contact time within the RSF system, leading to lower removal than that in SSF. Escolà Casas and Bester (2015) studied the degradation of seven PPCPs through SSF from effluent wastewater (at μg/L concentrations) which were recalcitrant in traditional activated sludge treatment. At a filtration rate of 0.012 m/h (0.288 m/d), 41%, 94%, 58%, 57%, 85%, 59% and 21% of diclofenac, propranolol, iopromide, iohexol, iomeprol, tebuconazole and propiconazole were eliminated, respectively. Except for tebuconazole and propiconazole, the removal of the other five PPCPs were influenced by the hydraulic retention time (HRT). In comparison, Hollender et al. (2009) studied the removal of 220 micro-pollutants in a WWTP equipped with post-ozonation followed by RSF (filtration rate: 14.4 m/h or 345.6 m/d). Before implementation of the ozonation, the RSF process achieved only limited removal of several PPCPs: diclofenac (20%); atenolol (15%); sotalol (15%); naproxen (30%); carbendazim (15%); and trimethoprim (15%). Relatively inefficient removal of PPCPs by RSF was also reported by Nakada et al. (2007). However, Table 3 shows an association between removal and influent concentration. When the initial concentration was of the order of ng/L, high removal values were achieved by RSF for some PPCPs such as caffeine (67~80%) and triclosan (> 90%) (Zearley and Summers, 2012). In contrast, lower removal of 25.3% and 74.2% were reported for treating 25 μg/L caffeine and triclosan through SSF, respectively (Li et al., 2018). Thus, the initial concentration is an influential factor of PPCP removal.

As both SSF and BSF can be used for small-scale applications, it is meaningful to compare their performance. Considering PPCP removal, generally, the intermittent mode favours better contact between the compounds and media and can lead to greater removal. Pompei et al. (2017, 2019) conducted two studies on the removal of the same six PPCPs (acetaminophen, diclofenac, naproxen, ibuprofen, benzophenone-3 and methylparaben) through continuous SSF (Pompei et al., 2019) and intermittent BSF (Pompei et al., 2017), with identical spiked influent concentrations of 2 μg/L. Both operational modes yielded good removal. Despite some differences in the filter configurations, effective sand size (0.25 mm versus 0.21 mm) and influent, their results provide some interesting comparisons. For naproxen and ibuprofen, the removal were similar under both operational modes (Table 3). However, intermittent BSF achieved higher removal of benzophenone-3, diclofenac and methylparaben. In contrast, continuous SSF achieved higher removal of acetaminophen. This may be attributed to the aerobic degradation of acetaminophen promoted by oxygen replenishment derived from continuous water inflow (Yu et al., 2006). A comparison of two filtration modes for bisphenol A removal was conducted by Sabogal-Paz et al. (2020), who employed a pilot-scale sand filter with the same media and configuration in both modes. Poor elimination was observed with continuous flow mode, with negative removal of -14 ± 16%, compared to 3 ± 8% removal with intermittent mode. The authors attributed this low-to-negative removal to desorption from the sand surface and the release of compounds from dead cells.

3.3 Treatment of wastewater versus surface/reservoir water

Most studies on sand filtration have been associated with wastewater treatment or surface/reservoir water treatment for drinking water purposes. At a filtration rate of 0.06 m/h (1.44 m/d) and initial concentrations of 2 μg/L or 5 μg/L, SSF removed less than 15% of sulfamethoxazole and clarithromycin, 15~50% of amoxicillin and oxytetracycline, and 50~80% of trimethoprim from surface water (Xu et al., 2021). A study by van Gijn et al. (2021) revealed that SSF removed approximately 20% of benzotriazole, 60% of caffeine and trimethoprim, 40% of clarithromycin, <20% of carbamazepine and diclofenac, 60~80% of metoprolol and propranolol, and 20~60% of naproxen and sulfamethoxazole from WWTP secondary effluent (with an initial concentration of 2 μg/L and filtration rate of 1 L/h or 24 L/d). Also, Escolà Casas et al. (2022) compared the efficacy of PPCP removal from synthetic wastewater and WWTP secondary effluent through SSF (filtration rate of 288 mm/d or 0.288 m/d). With high initial concentrations of 100 μg/L, SSF removed 9%, 33 ± 12% and 20% of carbamazepine, diclofenac and sulfamethoxazole from synthetic wastewater, respectively, compared to 0%, 20% and 17% from WWTP secondary effluent (initial concentration of 20 μg/L), respectively.

Considering RSF, Nakada et al. (2007) conducted a two-year study on the elimination of 24 PPCPs in a WWTP. The removal of ng-μg/L level PPCPs through RSF (filtration rate: 110 m/d) was considered inefficient, including carbamazepine (-52.1~22.4%), diethyltoluamide (-19.8~18.9%), ketoprofen (-186~20.5%), crotamiton (-5.2~16.3%), naproxen (-11.0~58.8%), triclosan (25.2~52.5%) and sulfamethoxazole (26.9%). Comparatively, Zearley and Summers (2012) systematically investigated the

removal of 34 trace PPCPs (at ng/L concentrations) from drinking water (tap water with 3 mg/L dissolved organic matter) through RSF (filtration rate of 1.2, 2.4 m/h or 28.8, 57.6 m/d). Higher removal were observed for some PPCPs, such as naproxen (72~86%) and triclosan (≥90%). However, other PPCPs, such as carbamazepine (0.5~1.6%) and sulfamethoxazole (2.4~4.1%), were minimally removed. These investigations provide preliminary insights into PPCP removal under varying influent quality. However, in the absence of comparative studies, it is difficult to draw conclusions on the variability of PPCP removal from wastewater and surface/reservoir water through sand filtration.

3.4 Laboratory-scale versus pilot/full-scale experiments

Understanding the implications of the experimental scale can help in evaluating the practicality of implementing sand filtration at larger-scales. Generally, the pilot/full-scale tests achieved relatively lower PPCP removal than laboratory-scale studies. For instance, laboratory-scale RSF (filtration rate of 1.2, 2.4 m/h or 28.8, 57.6 m/d) (Zearley and Summers, 2012) eliminated 72~86% of naproxen and 83~92% of trimethoprim (both at ng/L level) from tap water mixed with dissolved organic matter, whereas full-scale RSF of real wastewater achieved lower removal for the same compounds (30% or -11.0~58.8% for naproxen, 15% or 66.2% for trimethoprim; Table 3) (Hollender et al., 2009; Nakada et al., 2007). The removal of sulfamethoxazole (<4%, filtration rate of 0.15 m/h or 3.6 m/d) (Rooklidge et al., 2005) and carbamazepine (0%, filtration rate of 0.05 m/h or 1.2 m/d) (D'Alessio et al. 2015) observed in pilot-scale SSF studies were also lower than those observed by laboratory-scale SSF studies (Escolà Casas et al., 2022; van Gijn et al., 2021; Xu et al., 2021). Similarly, unsatisfactory removal of other PPCPs,

such as lincomycin, erythromycin, gemfibrozil, and phenazone, have also been observed in other pilot/full-scale filtration studies (Table 3).

Occasionally, PPCPs showed higher removal at larger-scale compared to that at the laboratory-scale. More than 99% of trimethoprim was removed in a pilot-scale SSF study (filtration rate of 0.15 m/h or 3.6 m/d; initial concentration of 0.2 mg/L) (Rooklidge et al., 2005) compared to removal of 50~85% by laboratory-scale SSF studies (filtration rate of 1.44 m/d or 24 L/d; initial concentration of 2 μg/L) (van Gijn et al., 2021; Xu et al., 2021). The removal of sulfamethoxazole was also higher with full-scale RSF compared to that with laboratory-scale RSF (Table 3). Furthermore, some PPCPs exhibited removal in a wide range, such as E1, E2 and caffeine, showing removal difference under various filtration conditions (Table 3). In general, differences in the influent quality, initial PPCP concentration and operational conditions could cause significant removal discrepancies. For more valid comparisons and evaluations, further investigations on PPCP removal are required at various experimental scales.

Overall, sand filtration systems reviewed herein achieved a reasonable but highly variable degree of PPCP removal. In a comprehensive study, Paredes et al. (2016) examined the removal of 18 PPCPs through sand biofiltration, and classified the compounds into three categories based on their removal behaviours: I) compounds exhibiting biotransformation and adsorption, e.g., celestolide; II) compounds exhibiting biotransformation only, e.g., sulfamethoxazole; and, III) compounds recalcitrant to both biotransformation and adsorption, e.g., carbamazepine. Paredes et al. (2016) considered biotransformation (biodegradation) and sand adsorption as the main PPCP removal

mechanisms during this treatment process. As these two mechanisms have also been studied by other researchers, a review of them is warranted.

4. Mechanisms of PPCP removal through sand filtration

The sand bed remains wet throughout the filtration process in both the continuous and intermittent modes. Therefore, the removal processes of PPCPs mainly occur in the aqueous phase or on the sand media surface. As sand filtration involves both physico-chemical and biological processes, various mechanisms are involved in this process, including absorption, diffusion, screening and sedimentation as mechanical mechanisms (Haig et al., 2011), and predation, scavenging, adsorption and bio-oxidation as microbiologically mediated purification mechanisms (Haig et al., 2014; Wang et al., 2021).

As PPCPs are soluble in water and usually present in trace concentrations, they rarely aggregate as pellets. The two main mechanisms of PPCP removal through sand filtration are biodegradation and adsorption (Escolà Casas and Bester, 2015; Wang et al., 2021). Other mechanisms, such as bio-sorption, may also play roles in PPCP removal (Li et al., 2018; Rolph et al., 2018).

4.1 Biodegradation

The main mechanism responsible for PPCP removal through SSF is considered to be biodegradation (Escolà Casas et al., 2022; Li et al., 2018). Biodegradation occurs in the *schmutzdecke* and upper sand layer (Campos et al., 2002). However, the effect of biodegradation in RSF is considered to be weak, but it can be enhanced (Srivastava and Chattopadhyay, 2022; Wang et al., 2021). Generally, PPCPs that are recalcitrant to

biodegradation are less likely to be effectively removed through sand filtration. For instance, carbamazepine is a low-biodegradable compound and its removal through sand filtration is often unsatisfactory, whereas ibuprofen, which is easily biodegraded, tends to be effectively removed (Table 3). An additional consideration is that, although some PPCPs are susceptible to aerobic and/or anaerobic biodegradation, aerobic conditions are normally more favourable for biodegradation (Conkle et al., 2012).

During the SSF/BSF maturation period (usually 10 d) and with continuous water inflow, microbes enter the filters, attach to, and grow on the sand surface of the upper sand layer using deposited organic matter in the influents as food, thereby forming the *schmutzdecke* (Ahammed and Davra, 2011; Elliott et al., 2008). The biodegradation of PPCPs occurs mostly within the *schmutzdecke* layer and gradually decreases with the increase in sand depth. However, as PPCPs are not energy sources (e.g., glucose) for general microorganisms, they can only be bio-degraded by microbes with specific degrading genes, or may share the same degradation pathways as other nutrients in the filter (Li et al., 2017, 2014). The oxygen level of the water decreases with the increase in the depth of the filter bed and may lead to anaerobic conditions (Reungoat et al., 2011; Young-Rojanschi and Madramootoo, 2014). Different types of microbial communities can develop at various depths, including anaerobic microbes. As PPCPs can be bio-degraded either aerobically or anaerobically (Suarez et al., 2010), different compounds may be removed at different sand depths. Generally, the *schmutzdecke* (within a 10 cm depth) is responsible for most microbial activity, but below a certain sand depth, biochemical reactions still take place (Huisman and Wood, 1974; Nakhla

409 and Farooq, 2003). Through biodegradation, PPCPs may undergo mineralisation, or
410 transformation into more hydrophobic/hydrophilic compounds (Halling-Sorensen et al.,
411 1998; Kümmerer, 2003; Zhang et al., 2014). Under mineralisation, degradable PPCPs
412 are gradually oxidised into simpler organic matters or inorganic compounds (e.g., water,
413 carbon dioxide, sulphates, and nitrates), either providing the energy required for the
414 metabolism and growth of the microbes or are discharged in the effluent (Huisman and
415 Wood, 1974).

416 The key factor influencing biodegradation is the HRT, which determines the
417 duration of contact between PPCPs and microbes in the filter (Escolà Casas and Bester,
418 2015; Reungoat et al., 2011). Ideally, the longer the HRT, the higher the PPCP removal
419 through biodegradation. However, only a very few studies (e.g., Sabogal-Paz et al.
420 (2020)) provide this information. Therefore, it is recommended that future research
421 focuses on investigating the effect of HRT on PPCP removal. In practice, the filtration
422 rate of SSF should not be inordinately low to prolong the filtration time because this
423 reduces the volume of water treated per unit of time. In addition, to ensure robust
424 biodegradation, neither the SSF operation temperature nor the dissolved oxygen (DO)
425 can be too low. Proper temperatures ensure good enzyme activities inside microbial
426 cells. At low temperatures, microbial metabolism decreases, slowing down the removal
427 of PPCPs and other pollutants, consequently deteriorating the water quality (Huisman
428 and Wood, 1974). Pompei et al. (2017) observed that the presence of standing
429 supernatant water in the filters for >24h reduces the DO in the effluent and should be
430 avoided, regardless of the operation mode. Usually, DO concentration in the effluent

should not be too low to avoid anaerobic conditions (Huisman and Wood, 1974).

Although some microbes can degrade specific PPCPs, the microbial community may also be affected by the PPCPs (Halling-Sorensen et al., 1998; Ribeiro et al., 2018; Tamura et al., 2017). After continuous dosing of four PPCPs (25 μg/L) in a SSF treatment system, Li et al. (2019) observed that the abundance of *Proteobacteria* phylum decreased from approximately 76% to 40%, while some other subdominant phyla (e.g., *Bacteroidetes*, *Planctomycetes*) increased, probably due to the spiked compounds. In another study in which 2 μg/L of mixed PPCPs were treated with household-scale BSF, more bacterial species were detected during the period without the PPCPs injection than with the PPCP injection (Pompei et al., 2017), though the addition of PPCPs did not affect the filter performance. Besides, changes in algae and cyanobacteria communities by PPCPs during sand filtration were also observed (Pompei et al., 2022).

4.2 Adsorption

Since sand is not a porous material, it cannot provide a sufficient surface area for effective adsorption like other media, such as activated carbon. Clean sand has few functional groups for chemical adsorption. Generally, the adsorption of PPCPs onto sand surface is considered negligible or hard to occur, and as a result, it is excluded as the dominant removal mechanism in eliminating PPCPs through sand filtration, compared to biodegradation (Escolà Casas and Bester, 2015; Reungoat et al., 2011).

Theoretically, hydrophobic PPCPs are more likely to be adsorbed onto sand surface than hydrophilic PPCPs. However, the removal of PPCPs does not always

correlate with the octanol-water distribution coefficient (log K_{OW}) in sand filters (Ternes et al., 2002; Zearley and Summers, 2012). For example, Nakada et al. (2007) investigated the elimination of 24 PPCPs through RSF in a WWTP by carrying out four sampling campaigns (July 2003, November 2003, June 2004 and October 2005). They observed that compounds with log K_{OW}<3 (e.g., diethyltoluamide, crotamiton, sulfapyridine, sulfamethoxazole, and E3) had removal above 50% in all four campaigns, and higher removal of > 80% were detected for strongly hydrophobic compounds with log K_{OW}>3 (e.g., ibuprofen, mefenamic acid, octylphenol, and bisphenol A) in some campaigns. Using the data from their study, we fitted the removal of PPCPs with their corresponding log K_{OW} values. Figure 3 shows the fitting graphs using the entire data set (four sampling campaigns) and average removal. The fitting graphs for each sampling campaign are shown in Fig. S1. The removal of the target compounds was not linearly correlated to the log K_{OW} (fitting R^2 of 0.0370 and 0.0021 for the entire data set and average removal, respectively). Several hydrophilic compounds (e.g., trimethoprim) exhibited higher removal than hydrophobic compounds (e.g., nonylphenol). In addition, the removal of some compounds were highly variable and not consistent among the campaigns. The same contradiction was also observed by Kennedy et al. (2013) who discovered that although EE2 had the highest log K_{OW} in a BSF process, E3 exhibited the highest adsorption affinity. Thus, it is assumed that adsorption process may be affected by other factors (e.g., biodegradation, hydraulic conditions, seasonal difference) and the likelihood of the adsorption of specific PPCPs during sand filtration is not dependent solely on their hydrophilicity/hydrophobicity only.

475 Unlike particle pollutants or pathogens that can be removed by mechanical forces
476 or biological effects (e.g., screening, predation), PPCPs are usually dissolved in water
477 at trace concentrations. Generally, two main mechanisms at the molecular level may
478 contribute to adsorption: van der Waals forces and electrostatic attraction (Huisman and
479 Wood, 1974; Wang et al., 2021).

480 Van der Waals forces, including dispersion forces, induction forces and dipole-
481 dipole attraction, can operate between the PPCP molecules and the surface of the sand,
482 which consists of silicon dioxide and other substances. Van der Waals forces can also
483 operate between PPCP molecules, leading to multi-layer adsorption on the sand particle
484 surface. Generally, van der Waals forces are considered weak forces. However, as the
485 distance between the centres of masses is very short (nm level), once the contact is
486 established, the attraction is considerably enhanced and cannot be ignored as these
487 forces increase with the reciprocal of the sixth power of the distance.

488 Electrostatic attraction operates between electrified bodies with electrical charges
489 and is inversely proportional to the square of the distance. Generally, mineral quartz
490 sand surface has a negative charge during SSF. Positively charged PPCPs can be
491 attracted to the sand surface through electrostatic attraction. This process usually leads
492 to the oversaturation of this attraction, making the sand particles and the attracted
493 chemicals become positively charged. Consequently, negatively charged PPCPs are
494 then attracted onto the already adsorbed chemicals. Once this process begins, the charge
495 reversal and accumulation of the two types of charged molecules continues throughout
496 the filtration process.

497 The removal of PPCPs solely through biodegradation is often unsatisfactory (Li et
498 al., 2018; Pompei et al., 2017). Although adsorption by sand media may play a role, its
499 effectiveness is often insignificant, and desorption may also occur because physical
500 adsorption is reversible (Rizzo et al., 2015). As sand is a non-porous material with a
501 small surface area, adsorption may be significantly enhanced by employing other
502 porous materials (e.g., activated carbon) as filtration media.

503 **4.3 Other mechanisms**

504 Besides direct adsorption onto the sand surface, PPCPs can also be adsorbed onto
505 the biomass/biofilm accumulated during filtration (Kennedy et al., 2013; McKie et al.,
506 2016). In general, biomass/biofilm is a mixture of water, microbes and their metabolic
507 products, making it slimy and gelatinous (Weber-Shirk and Dick, 1997). The bio-
508 sorption of PPCPs onto microbial surfaces and other relevant substances might also
509 occur, even if some PPCPs might not be easily biodegraded (e.g., diethyltoluamide).
510 However, this process is considered insignificant, especially for hydrophilic
511 compounds (Paredes et al., 2016). Zearley and Summers (2012) also observed that the
512 bio-sorption of PPCPs onto biomass was insignificant, and that the maximum biomass
513 adsorption capacity was reached within two hours of operation. In addition, light-
514 sensitive PPCPs (e.g., triclosan) may experience photodegradation when exposed to
515 direct light in the water or absorbed onto the upper surface of sand bed (Li et al., 2018).

516 PPCPs may also be indirectly removed along with the removal of particle
517 pollutants (Hollender et al., 2009). By screening and sedimentation, particle solids can
518 be retained in the filter (mainly at the upper layer). When PPCPs are adsorbed onto the

surface of a material with a stronger adsorption capacity than sand, these pollutants do not flow out in the effluent. However, few studies have focused on this aspect, and it is difficult to draw reasonable conclusions.

5. Sand-GAC filtration technologies

Given the fact that removal of PPCPs during sand filtration varies considerably, some studies aiming to enhance the adsorption process have been conducted, to combine it with GAC, which has a large surface area (Babaei et al., 2019; Gabarrón et al., 2016). GAC, whose surface area can exceed 1,000 m^2/g (Rossner et al., 2009), is a porous medium that is widely used as an adsorbent in drinking water and tertiary wastewater treatment processes worldwide (Dwivedi et al., 2018; Rizzo et al., 2015; Yu et al., 2022). Compared to sand, GAC provides a much larger surface area for physical adsorption if no specific functional groups exist (Li et al., 2018), as well as for chemical adsorption when functional groups (e.g., carboxyls, lactones, aldehydes, ketones) exist (Jung et al., 2001). Nevertheless, as GAC is more expensive than sand, single GAC adsorption in water treatment units may be not affordable in LMICs. Therefore, combining GAC with sand filtration can provide an optional solution. However, when sand filtration is combined with GAC unit/part, changes of operational parameters are sometimes necessary (e.g., change of filtration rate to better suit GAC filtration). In the sections that follow, three types of sand-GAC filtration technologies are reviewed: serial, dual and sandwich filters.

5.1 Serially connected filters

A typical serially connected sand-GAC filtration system consists of a front sand

filter unit with a GAC tank behind it (Fig. 4). Solid pollutants are filtered in the sand tank and further adsorption occurs in the GAC unit. A pilot-scale study was conducted using serially connected sand-GAC filtration (media parameters not shown) to treat 12 PPCPs from tertiary-treated wastewater (Gidstedt et al., 2022). PPCP removal decreased with the increase in the filtration rate (empty bed contact time, EBCT: 5~30 min; removal data in sand filter not shown). Rizzo et al. (2015) observed that less than 10% of four tested PPCPs (namely caffeine, carbamazepine, ibuprofen and diclofenac, 1 mg/L) were removed from WWTP biological effluent through sand filtration (effective size of 0.6 mm) during a 62-day operation (EBCT: 75 min); in contrast, the GAC reactor (surface area of 875 m^2/g) achieved a maximum removal of 62 % at the start of the process, which subsequently decreased to 24% constantly after 14 hours. Paredes et al. (2016) used coarse sand (particle size of 1~2 mm) and GAC (surface area not specified) to remove 18 PPCPs (EBCT: 17 min~3.2 d) and observed an improvement in effluent quality. Compared to sand, the removal of organic matter, ammonium and nitrate improved with GAC. Furthermore, carbamazepine, diazepam and diclofenac were only removed through adsorption by GAC. No influence of filtration rate or type of secondary effluent was observed on GAC performance. With respect to drinking water treatment, Gabarrón et al. (2016) investigated the removal of 49 PPCPs in a drinking water treatment plant (water velocity of up to 3 m^3/s), finding that GAC filtration was one of the most efficient technologies, whereas the efficacy of sand filtration (parameters not specified) varied considerably (0~100%).

5.2 Dual-layer filters

563 The treatment of PPCPs using dual-layer sand-GAC (sand above GAC) filters is
564 not common. Babaei et al. (2019) reported that 86.7% of linear alkylbenzene sulfonate
565 was removed through dual-layer sand-GAC filtration (EBCT: 50~100 min). In contrast,
566 dual-media filtration of GAC-sand (GAC above sand) mode is more used (Fig. 4). A
567 pilot-scale drinking water treatment plant using ozonated lake water was evaluated by
568 McKie et al. (2016). The dual-layer filters (EBCT: 16 min) comprised 50~150 cm GAC
569 (surface area not specified) over 15~50 cm of sand (effective size not specified). Two
570 of the nine PPCPs evaluated in this study had a removal of more than 50% (average
571 removal of 39 %) without coagulation. By adding polyaluminum hydroxychloride in
572 the concentration of 0.2 to 0.8 mg Al^{3+}/L, the average removal of the target PPCPs
573 increased from 45 % to 70 %. Altmann et al. (2016) compared a dual-layer filter (GAC-
574 sand, downflow) with a monolayer GAC filter (upflow) for removing various PPCPs
575 (e.g., gabapentin) from wastewater (6 m/h). The dual-layer filter comprised a 1.4 m
576 GAC layer (surface area not specified) and a 0.6 m quartz sand layer (0.7~1.1 mm).
577 Both filters exhibited similar removal for most PPCPs. Well-adsorbing compounds,
578 such as carbamazepine and benzotriazole, were reduced by almost 40% at 25,000-bed
579 volumes. Ma et al. (2018) investigated PPCP removal (e.g., atenolol) using GAC-sand
580 filters and anthracite-sand filters (4.88–9.76 m/h, media parameters not specified). A
581 higher mean removal of 49.1~94.4% was achieved using GAC-sand filters compared
582 to a removal of 0~66.1% using anthracite-sand filters due to a combination of
583 adsorption and biodegradation mechanisms. Although these studies indicate that better
584 removal can be achieved through GAC-sand filtration than sand filtration alone, putting

585 GAC above the sand may cause quick clogging of GAC micropores owing to the
586 screening of particles and the growth of biofilms on the GAC top layer, which reduce
587 the inherent advantages associated with the high adsorption performance of GAC (Li,
588 2019).

589 **5.3 GAC-sand sandwich filter**

590 A typical sandwich-layer filter consists of an upper sand layer, a middle GAC layer
591 and a lower sand layer (Fig. 5). GAC sandwich filters were was first designed and tested
592 by M. Bauer (Thames Water Utilities Ltd., United Kingdom) to eliminate pesticides
593 that could not be removed through SSFs, while avoiding constructing extra GAC
594 contactors (Bauer et al., 1996). Each layer has a specific function in this design: the
595 upper sand layer acts as the primary screener and provides a suitable medium for
596 *schmutzdecke* growth. The contaminants that are not degraded in the upper sand layer
597 are adsorbed onto the middle GAC layer. The last lower sand layer acts as a supporting
598 layer to minimise the potential of biological entities and GAC fines from entering the
599 effluents. In this study, none of the 20 target pesticides were detected in the effluent
600 using the GAC sandwich SSF, whereas various pesticides were detected in the effluent
601 of the SSF control.

602 The removal of four PPCPs (diethyltoluamide, acetaminophen, caffeine and
603 triclosan) from synthetic wastewater using GAC sandwich SSF at various GAC
604 proportion and filtration rates were explored by Li et al. (2018). An average removal of
605 98.2% was achieved at a filtration rate of 10 cm/h using a 10 cm sand/20 cm
606 GAC/20 cm sand filter, making a significant difference in the removal performance

607 compared to that using conventional SSF ($p < 0.05$). In addition, no significant
608 difference for PPCP removal ($p > 0.05$) at 10 cm/h and 20 cm/h filtration rates further
609 demonstrated the flexibility of this technology. The application of GAC sandwich SSF
610 following constructed wetland system has been successfully implemented to remove
611 PPCPs from natural water (Li et al., 2019). Good performance of GAC sandwich SSF
612 (0.06 m/h) in removing antibiotics was also observed by Xu et al. (2021). An average
613 removal of 97 ± 2% was achieved for amoxicillin, clarithromycin, oxytetracycline,
614 sulfamethoxazole, and trimethoprim, compared to a removal of just 20 ± 19% with
615 conventional SSF.

616 The studies discussed above demonstrate that sand-GAC filtration technologies
617 significantly improve the PPCP removal, and GAC-sand sandwich filters provide better
618 performance. However, the investigations on GAC-sand sandwich filters have only
619 been conducted at the laboratory-scale, whereas most studies on serially connected and
620 dual-layer sand-GAC filters have been carried out at larger-scales. The filter
621 configurations, media properties, inflow water quality and operational conditions could
622 lead to considerable PPCP removal differences. Although the available research on
623 sand-GAC filtration is limited, the enhanced PPCP removal performance indicates that
624 these technologies have great potentials to effectively remove PPCPs.

625 In practice, choosing a suitable sand or sand-GAC filtration technology for PPCP
626 removal depends on various factors such as the capital costs, materials, inflow water
627 quality and maintenance requirements (Li, 2019). Although GAC can enhance the
628 PPCP removal through SSF, the service life of GAC also needs to be considered, as it

varies considerably (from several weeks to years) with the filtration mode and rate, influent type and pH, GAC type, surface area and size (Bayer et al., 2005; Kennedy et al., 2015; Zearley and Summers, 2012). Generally, the GAC breakthrough of PPCPs comes earlier in treating wastewater than in treating drinking water. This is because of the adsorption competition associated with higher concentrations of low molecular weight acids and neutral organics in wastewater (Zietzschmann et al., 2016). At the end of GAC service life, reactivation for regeneration of GAC is needed, among which thermal and chemical ways are the two commonly used processes (Haig et al., 2014; Lantagne et al., 2006). However, compared to the scraping and washing strategies used for sand cleaning, GAC reactivation processes are expensive (Li, 2019). Therefore, the operation and maintenance costs should also be considered when selecting an appropriate filtration technology.

6. Future considerations on the application of sand filtration for PPCP removal

Sand and sand-GAC filtration technologies have potential for practical application in PPCP removal. To enhance the understanding of PPCP removal, the authors recommend the need for further investigations as follows.

Exploration of PPCP removal under different operational conditions for a wider range of PPCPs. Currently, more data are needed to enable valid comparisons between various filter types, influent quality and experimental scales. Besides, owing to the development of detection technology and quantification methods, more than a hundred of PPCPs can now be detected simultaneously, and various new PPCPs have been investigated (e.g., glucocorticoids, mineralocorticoids) (Archer et al., 2017; Weizel et al., 2018; Yang et al.,

651 2018). Therefore, to allow relevant comparisons, future research could investigate removal
652 of more PPCPs through sand filtration under different operational conditions.

653 A deeper understanding is required of the PPCP removal mechanisms through
654 biodegradation. As biodegradation is complex, the pathways (transformation or
655 accumulation) of PPCPs in microbial metabolism are worth investigating. Moreover,
656 although the *schmutzdecke* is responsible for most of the biodegradation in BSF and SSF,
657 microbial activity has also been observed with the increase in the sand bed depth (Huisman
658 and Wood, 1974; Nakhla and Farooq, 2003). It can be speculated that biodegradation of
659 different PPCP categories/groups occurs at various depths of the filter bed due to the
660 preference of the microbial community. This knowledge may help in filter design since
661 some PPCPs are removed more under anaerobic/oxic conditions (Suarez et al., 2010).

662 In the studies conducted by Pompei et al. (2022) and Li et al. (2019), spiked PPCPs
663 in the influent were found to affect the microbial community structure. As some
664 microbes are sensitive to the toxicity induced by spiked PPCPs, the composition of the
665 *schmutzdecke* and deeper microbial community may be negatively affected by high or
666 long-term input of PPCPs, thereby reducing the filter performance and deteriorating the
667 water quality. Therefore, the long-term filter performance during PPCP removal may
668 be an interesting subject of future research.

669 Considering the adsorption mechanism, the current findings indicate that PPCP
670 removal does not always correlate with log K_{OW}. Although adsorption is not the dominant
671 removal mechanism, the incorporation of GAC in sand-GAC filters significantly enhances
672 PPCP removal. But sorption is a dynamic adsorption/desorption process and competitive

673 adsorption may occur (Conkle et al., 2010; Rizzo et al., 2015; Xing et al., 2008; Zhang
674 and Zhou, 2005). Moreover, other factors (e.g., biodegradation and hydraulic conditions)
675 may also influence the adsorption process. Therefore, the factors and mechanisms
676 influencing PPCP adsorption during sand filtration require further investigation.

677 Besides GAC, other materials (e.g., woodchips and clay) have also been combined
678 with sand for water purification. Therefore, it is suggested to review their performance
679 for PPCP removal in the future. Theoretically, increasing the filtration media surface area
680 would enhance PPCP removal. The manufacture of advanced porous materials is thus an
681 option for future consideration. In addition, the combination of sand filtration with other
682 technologies, such as ozonation (Hollender et al., 2009), coagulation (Racar et al., 2019),
683 membrane filtration (Zahrim and Hilal, 2013), has been successfully implemented,
684 demonstrating the compatibility and flexibility of sand filtration. Therefore, future
685 investigations on the use of advanced materials in sand filtration and the combination of
686 sand filtration with other technologies for PPCP removal are suggested.

687 **7. Conclusion**

688 This paper reviewed the recent progress in sand and sand-GAC filtration
689 technologies for PPCP removal from water. Overall, SSF and BSF provide better PPCP
690 removal than RSF. Although some PPCPs are easier to be eliminated through sand
691 filtration (e.g., ibuprofen compared to carbamazepine), most PPCPs exhibit highly
692 variable removal. Differences in the influent water quality, experimental scale, initial
693 PPCP concentration and operational conditions limit detailed comparisons.

694 Biodegradation in the *schmutzdecke* and upper sand layer is the main PPCP

removal mechanism, and HRT is the key factor that influences biodegradation. Conversely, contact with PPCPs might affect the microbial community in the filter. Adsorption is generally excluded from the dominant mechanisms and PPCP removal does not always correlate with their hydrophilicity/hydrophobicity; van der Waals forces and electrostatic attraction contribute to the adsorption. Besides these two mechanisms, other processes such as bio-sorption may also contribute to PPCP removal. To enhance PPCP removal through adsorption, the porous material GAC has been combined with conventional sand as sand-GAC filtration technologies. Serial, dual, and sandwich filters provide significant PPCP removal improvements.

Further research is recommended along several relevant strands: explore the influence of different operational conditions on the removal of a broader range of PPCPs; attain a deeper understanding of different removal mechanisms; investigate filter performance over long-term operation; and evaluate the compatibility and compare the effectiveness of sand filtration with other water treatment technologies and materials for PPCP removal.

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1148 Figure captions:

1149 Figure 1. Schematic representations of typical slow/rapid sand filter (left) and household

1150 biosand filter (right).

1151 Figure 2. Removal of selected PPCPs reported in published sand filtration studies.

1152 Figure 3. Fitting graphs of PPCP log K_{OW} with removal based on reported data from Nakada et

1153 al. (2007).

1154 Figure 4. Schematic representations of typical serially connected sand-GAC filtration system

1155 (left) and dual-layer GAC-sand filter (right)

1156 Figure 5. Schematic representation of a typical GAC sandwich slow sand filter.

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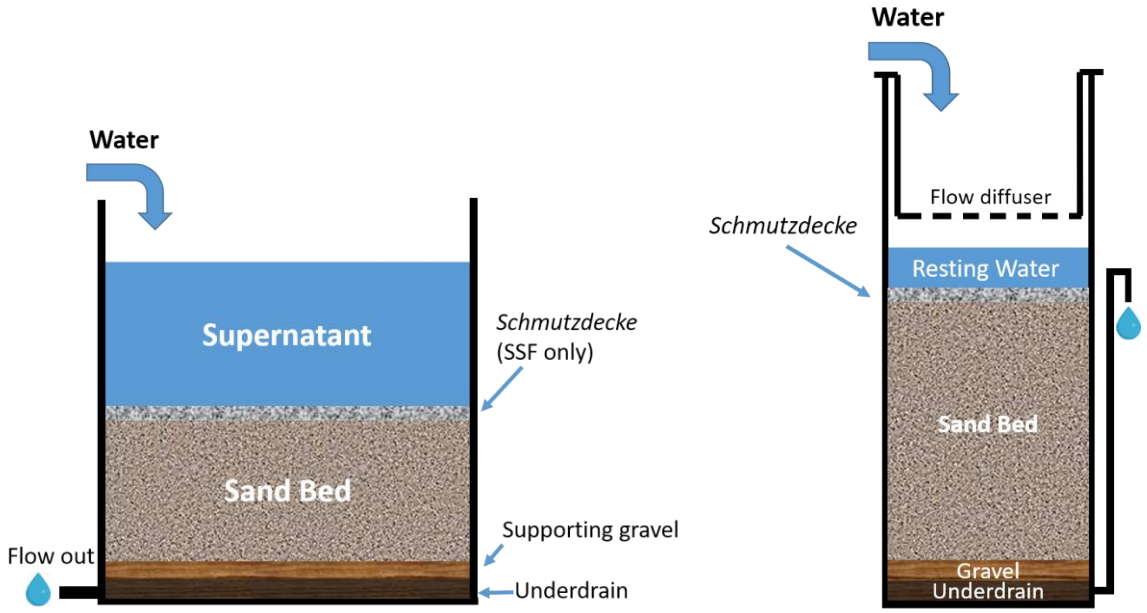

1159

1160 Figure 1. Schematic representations of slow/rapid sand filter (left) and household biosand filter

1161 (right).

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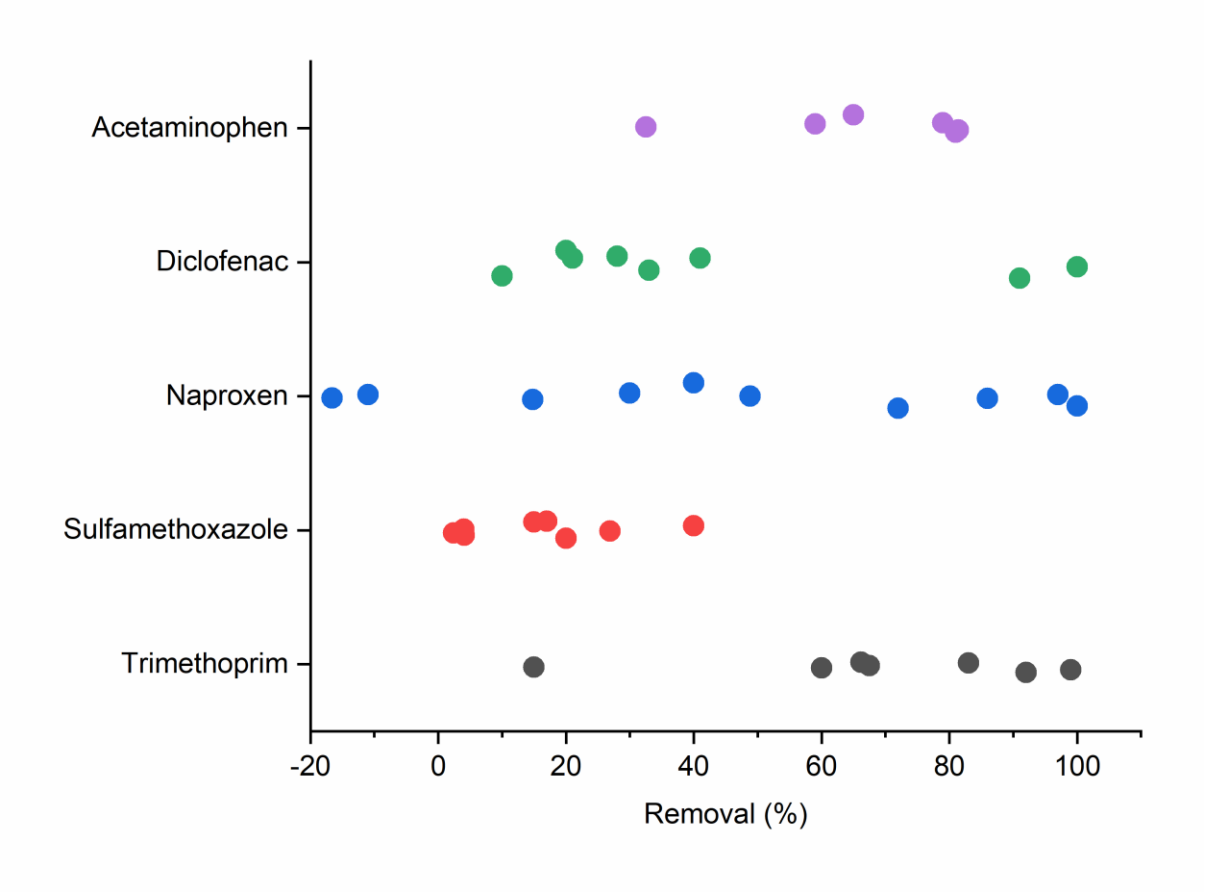
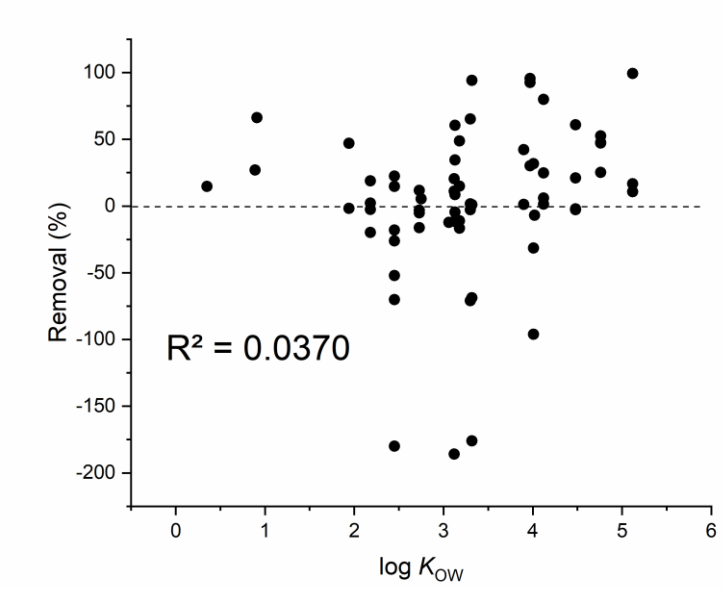


Figure 2. Removal of selected PPCPs reported in published sand filtration studies. (Data from (Escolà Casas et al., 2022; Escolà Casas and Bester, 2015; Hollender et al., 2009; Li et al., 2018; Nakada et al., 2007; Pompei et al., 2019, 2017; Rooklidge et al., 2005; van Gijn et al., 2021; Xu et al., 2021; Zearley and Summers, 2012); If the removal lies in a range, the average removal is used)



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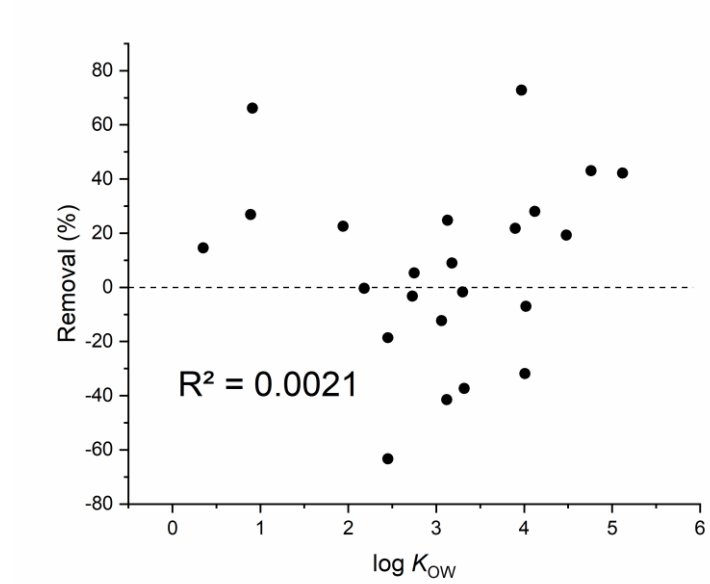
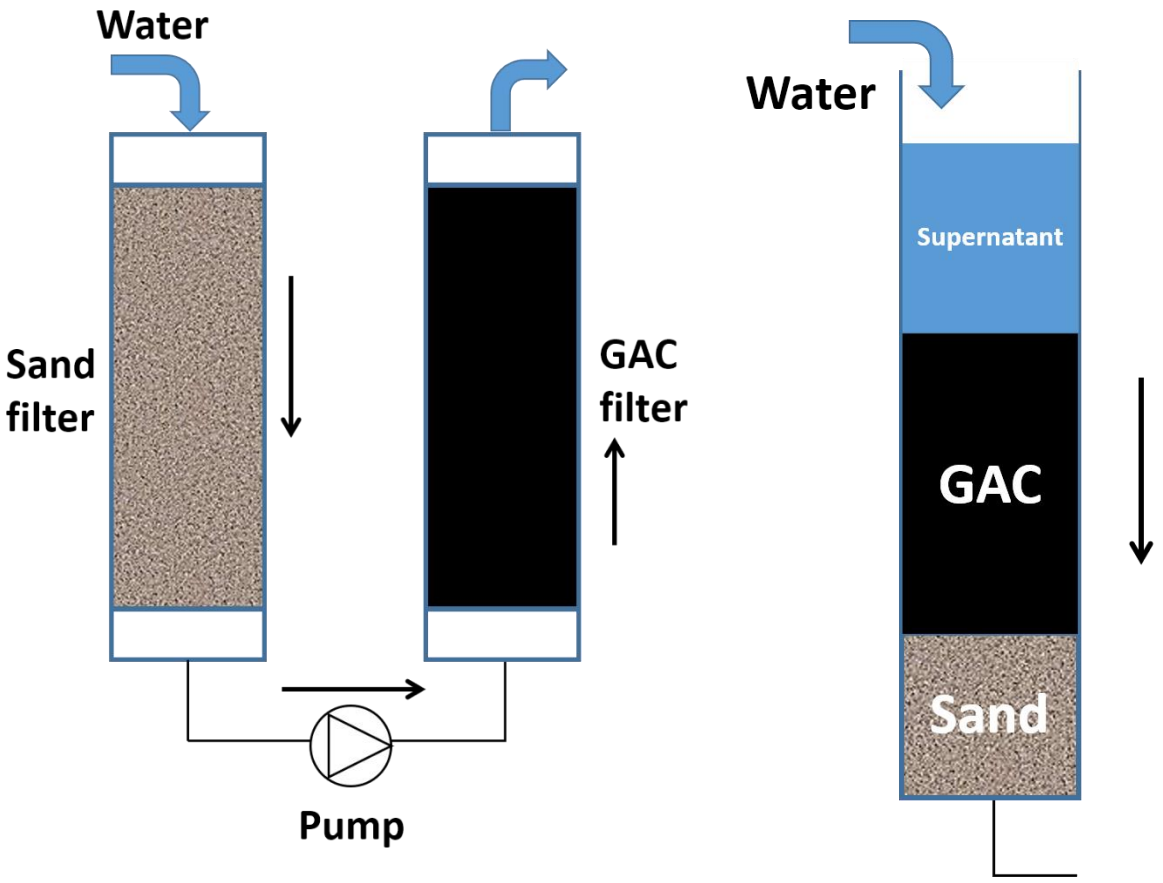


1172

1173 Figure 3. Fitting graphs of PPCP log K_{OW} with removal based on reported data from Nakada et
1174 al. (2007). (Top: fitting graph of removal from whole 4 campaigns; bottom: fitting graph of
1175 average removal. Data from the compounds of triclosan, thymol, naproxen, mefenamic acid,
1176 ketoprofen, fenoprofen, ibuprofen, DEET, crotamiton, carbamazepine, propyphenazone,
1177 sulfapyridine, sulfamethoxazole, trimethoprim, azithromycin, erythromycin, roxithromycin,
1178 nonylphenol, octylphenol, bisphenol A, E1, EE2 and E3. Removal of -737% of mefenamic acid

1179 was excluded)



1180

1181 Figure 4. Schematic representations of typical serially connected sand-GAC filtration system

1182 (left) and dual-layer GAC-sand filtration system (right)

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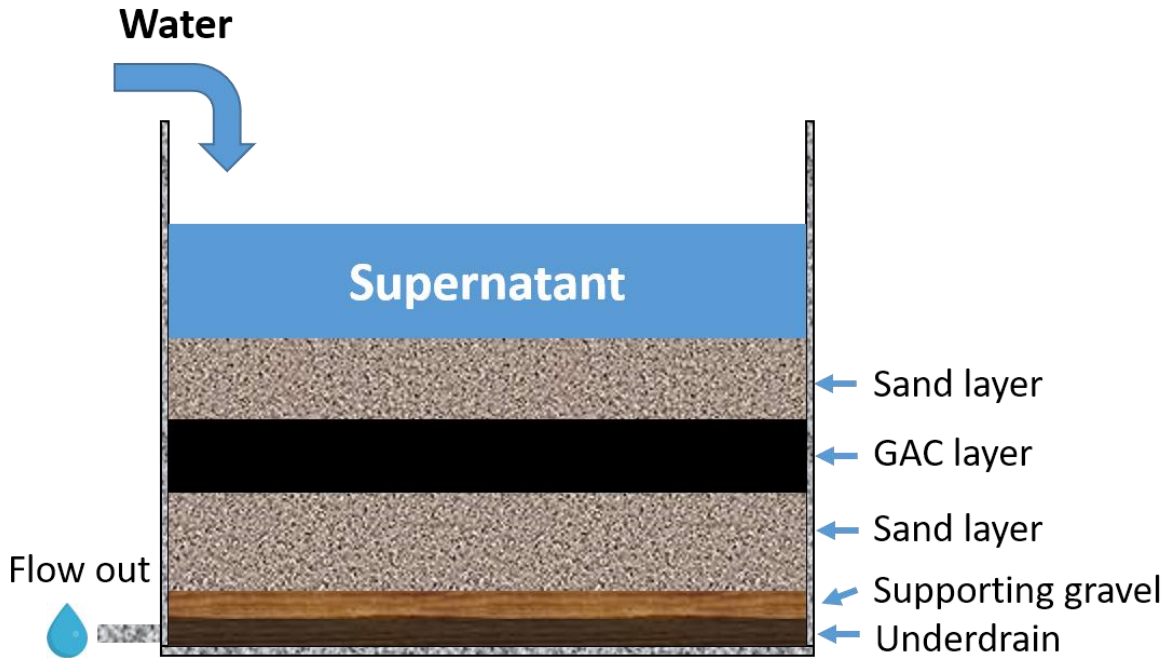

Figure 5. Schematic representation of a typical GAC sandwich slow sand filter.

Table captions:

Table 1. Typical design/operational parameters for SSF, BSF and RSF.

Table 2. Sand grain size/effective size (D_{10}), uniformity coefficient (D_{60}/D_{10}), feed water, experimental scale and filtration rate/volume used in sand filtration experiments.

Table 3. Typical PPCP removal from water using conventional sand filtration.

Table 1. Typical design/operational parameters for SSF, BSF and RSF.

Sand filter type	Supernatant water height	Sand media depth	Filtration rate/retention time	Operational mode	Cleaning strategy	Sand effective size (D_{10})	Sand uniformity coefficient (D_{60}/D_{10})
SSF	100~150 cm	1.6~1.2 m	0.1~0.3 m/h (2.4~7.2 m/d)	Continuous	Scraping/Replacement	0.1~0.3 mm	Around 3.0
BSF	5~20 cm	0.4~0.55 m	1~48 h	Intermittent	Scraping/Replacement	0.15~0.20 mm	1.5~2.5
RSF	150~200 cm	0.5~1.0 m	100~475 m/d	Continuous	Backwashing	>0.55 mm	<1.5

Table 2. Sand grain size/effective size (D_{10}), uniformity coefficient (D_{60}/D_{10}), feed water, experimental scale and filtration rate/volume used in sand filtration experiments.

Filter type	Sand effective/grain size	Uniformity coefficient	Feed water	Scale	Filtration rate/volume*	Reference
SSF	0.20 mm effective size, size range: 0.16~0.50 mm	1.82	Surface water	Lab	0.06 m/h (1.44 m/d)	(Xu et al., 2021)
BSF	0.18 mm effective size	1.64	Synthetic rainwater	Pilot	0.38 ± 0.13 m/d or 3 L/d	(Sabogal-Paz et al., 2020)
RSF	0.6 mm effective size	<1.3	WWTP sedimentation effluent	Full/lab	120 m/d	(Shirakawa et al., 2022)
SSF	0.60 mm effective size	1.40	Synthetic wastewater	Lab	5, 10, 20 cm/h (1.2, 2.4, 4.8 m/d)	(Li et al., 2018)
BSF	0.210 mm effective size	1.40	Surface water	Lab	24 L twice a week	(Pompei et al., 2017)
SSF	0.25 mm effective size	2~3	Reservoir water	Pilot	3 m/d	(Pompei et al., 2019)
SSF	0.210~0.297 mm		WWTP effluent	Lab	0.012 m/h (0.288 m/d)	(Escolà Casas and Bester, 2015)
SSF	0.38 mm effective size	2.78	Reservoir water	Full/lab	0.15 m/h (3.6 m/d)	(Haig et al., 2014)
BSF	0.7~1.0 mm		Surface water	Lab	0.3 L twice a day (0.6 L/d)	(Hwang et al., 2014)
BSF	0.19~0.22 mm effective size	3.5~4.0	Tap water added with sewage water	Lab	20 L/d	(Baig et al., 2011)
BSF	0.17mm effective size	2.06	Surface water added with microbes	Lab	2 L/d	(Young-Rojanschi and Madramootoo, 2014)
BSF	0.17 mm and 0.52 mm effective size both		Synthetic surface water	Lab	20 L/d	(Jenkins et al., 2011)
BSF	0.19~0.22 mm effective size	3.5~4.0	Reservoir and surface water	Lab	20, 40 L/d	(Elliott et al., 2008)
BSF	0.23 mm effective size	3.1	Tap water added with microbes; surface water	Lab	20, 40 L/d	(Ahammed and Davra, 2011)
BSF	0.27 mm effective size	1.4	Reservoir water	Lab	0.45 L/d	(Elliott et al., 2011)
SSF	0.3 mm effective size		Reservoir water	Full	0.15 m/h (3.6 m/d)	(Campos et al., 2002)
RSF	maximum grain size of 1.18mm, 0.6mm effective size	1.8	WWTP biological effluent	Lab	4.3~5.5 mL/min (6.2~7.9 L/d)	(Rizzo et al., 2015)
RSF	0.45 effective size	1.3	Tap water added with dissolved organic matter	Lab	1.2, 2.4 m/h (28.8, 57.6 m/d)	(Zearley and Summers, 2012)
SSF	0.55 mm effective size, D_{60}=3.1 mm	5.6	Constructed wetland-treated urban wastewater	Full	13~160 mm/d (0.013~0.16 m/d)	(Matamoros et al., 2007)

* Both original and unified filtration rate/volume (m/d or L/d, if applicable) are displayed.

Table 3. Typical PPCP removal from water using sand filtration.

Compound	Class	Initial concentration	(Average) Removal (%)*	Filtration rate /volume**	Filter type	Filter mode***	Feed water	Scale	Reference
17α-ethinyl estradiol (EE2)	Steroid estrogen	5 mg/L	11.4 ± 11	20 L/d	BSF	I	Surface water	Lab	(Kennedy et al., 2013)
17β-estradiol (E2)	Steroid estrogen	50 μg/L	11~92	0.05 m/h (1.2 m/d)	SSF	C	Surface water (added with WWTP primary effluent)	Pilot	(D'Alessio et al., 2015)
		1.34~2.31 ng/L	-96.0~31.7	110 m/d	RSF	C	WWTP secondary effluent	Full	(Nakada et al., 2007)
		12 ng/L	34.58 (augmented filter) -66.66 (nonaugmented filter)	0.15 m/h (3.6 m/d)	SSF	C	Surface water	Lab	(Haig et al., 2016)
Acetaminophen (paracetamol)	Analgesic	306 ± 142 ng/L	59~79	1.2, 2.4 m/h (28.8, 57.6 m/d)	RSF	C	Tap water added with dissolved organic matter	Lab	(Zearley and Summers, 2012)
		25 μg/L	81.4	5, 10, 20 cm/h (1.2, 2.4, 4.8 m/d)	SSF	C	Synthetic wastewater	Lab	(Li et al., 2018)
		2 μg/L	81	3 m/d	BSF	C	Reservoir water	Pilot	(Pompei et al., 2019)
		2 μg/L	65.2	24 L twice a week	BSF	I	Surface water	Lab	(Pompei et al., 2017)
Amoxicillin	Antibiotic	5 μg/L	15~50	0.06 m/h (1.44 m/d)	SSF	C	Surface water	Lab	(Xu et al., 2021)
Atenolol	Hypotensor	0.2 μg/L	50	0.1 m/h (2.4 m/d)	SSF	C	Ground water	Lab	(Vu and Wu, 2022)
Bisphenol A	Plasticizer	311 ± 285 ng/L	64 ± 29	1.2, 2.4 m/h (28.8, 57.6 m/d)	RSF	C	Tap water added with dissolved organic matter	Lab	(Zearley and Summers, 2012)
		49.5~3480 ng/L	-176~94.1	110 m/d	RSF	C	WWTP secondary effluent	Full	(Nakada et al., 2007)
		2.35 ± 0.41 mg/L	-14 ± 16	0.38 ± 0.13 m/d	BSF	C	Synthetic rainwater	Pilot	(Sabogal-Paz et al., 2020)
		2.35 ± 0.41 mg/L	3 ± 8	3 L/d	BSF	I	Synthetic rainwater	Pilot	(Sabogal-Paz et al., 2020)
Benzophenone-3	Sun screener	2 μg/L	71	3 m/d	BSF	C	Reservoir water	Pilot	(Pompei et al., 2019)
		2 μg/L	0~100	24 L twice a week	BSF	I	Surface water	Lab	(Pompei et al., 2017)
Benzotriazole	Ultraviolet absorbent	100 μg/L	14 ± 8	288 mm/d (0.288 m/d)	SSF	C	Synthetic wastewater	Lab	(Escolà Casas et al., 2022)
		20 μg/L	≈10	125 mm/d (0.125 m/d)	SSF	C	Mix of 75% of secondary WWTP effluent with 25% nutritional solution	Lab	(Escolà Casas et al., 2022)
		2 μg/L	≈20	1 L/h (24 L/d)	SSF	C	WWTP secondary effluent	Lab	(van Gijn et al., 2021)
Caffeine	Psychomotor stimulant	188 ± 147 ng/L	67~80	1.2, 2.4 m/h (28.8, 57.6 m/d)	RSF	C	Tap water added with dissolved organic matter	Lab	(Zearley and Summers, 2012)
		25 μg/L	25.3	5, 10, 20 cm/h (1.2, 2.4, 4.8 m/d)	SSF	C	Synthetic wastewater	Lab	(Li et al., 2018)
		2 μg/L	≈60	1 L/h (24 L/d)	SSF	C	WWTP secondary effluent	Lab	(van Gijn et al., 2021)
		50 μg/L	23~100	0.05 m/h (1.2 m/d)	SSF	C	Surface water (added with WWTP primary effluent)	Pilot	(D'Alessio et al., 2015)
Carbamazepine	Antiepileptic	85 ± 49 ng/L	0.5~1.6	1.2, 2.4 m/h (28.8, 57.6 m/d)	RSF	C	Tap water added with dissolved organic matter	Lab	(Zearley and Summers, 2012)
		100 μg/L	9	288 mm/d (0.288 m/d)	SSF	C	Synthetic wastewater	Lab	(Escolà Casas et al., 2022)
		20 μg/L	0	125 mm/d (0.125 m/d)	SSF	C	Mix of 75% of secondary WWTP effluent with 25% nutritional solution	Lab	(Escolà Casas et al., 2022)
		2 μg/L	≈0~20	1 L/h (24 L/d)	SSF	C	WWTP secondary effluent	Lab	(van Gijn et al., 2021)
		50 μg/L	0	0.05 m/h (1.2 m/d)	SSF	C	Surface water (added with WWTP primary effluent)	Pilot	(D'Alessio et al., 2015)
		2.32~46.4 ng/L	-52.1~22.4	110 m/d	RSF	C	WWTP secondary effluent	Full	(Nakada et al., 2007)
Clarithromycin	Antibiotic	2 μg/L	<15	0.06 m/h (1.44 m/d)	SSF	C	Surface water	Lab	(Xu et al., 2021)
		2 μg/L	≈40	1 L/h (24 L/d)	SSF	C	WWTP secondary effluent	Lab	(van Gijn et al., 2021)
Clofibric acid	Lipid regulator	263 ± 70 ng/L	35~52	1.2, 2.4 m/h (28.8, 57.6 m/d)	RSF	C	Tap water added with dissolved organic matter	Lab	(Zearley and Summers,

									2012)
Crotamiton	Antipruritic	656~950 ng/L	-5.2~16.3	110 m/d	RSF	C	WWTP secondary effluent	Full	(Nakada et al., 2007)
DEET	Mosquito repellent	25 μg/L	25.7	5, 10, 20 cm/h (1.2, 2.4, 4.8 m/d)	SSF	C	Synthetic wastewater	Lab	(Li et al., 2018)
		16.9~198 ng/L	-19.8~18.9	110 m/d	RSF	C	WWTP secondary effluent	Full	(Nakada et al., 2007)
Diclofenac	Analgesic	252 ± 90 ng/L	21~28	1.2, 2.4 m/h (28.8, 57.6 m/d)	RSF	C	Tap water added with dissolved organic matter	Lab	(Zearley and Summers, 2012)
		2 μg/L	91	3 m/d	BSF	C	Reservoir water	Pilot	(Pompei et al., 2019)
		100 μg/L	33 ± 12	288 mm/d (0.288 m/d)	SSF	C	Synthetic wastewater	Lab	(Escolà Casas et al., 2022)
		20 μg/L	≈20	125 mm/d (0.125 m/d)	SSF	C	Mix of 75% of secondary WWTP effluent with 25% nutritional solution	Lab	(Escolà Casas et al., 2022)
		2 μg/L	≈0~20	1 L/h (24 L/d)	SSF	C	WWTP secondary effluent	Lab	(van Gijn et al., 2021)
		2 μg/L	100	24 L twice a week	BSF	I	Surface water	Lab	(Pompei et al., 2017)
		0.24 ± 0.047 μg/L	41 ± 2	0.012 m/h (0.288 m/d)	SSF	C	WWTP effluent	Lab	(Escolà Casas and Bester, 2015)
		Data not shown	20	14.4 m/h (345.6 m/d)	RSF	C	WWTP ozonation unit effluent	Full	(Hollender et al., 2009)
Erythromycin	Antibiotic	104 ± 77 ng/L	15~27	1.2, 2.4 m/h (28.8, 57.6 m/d)	RSF	C	Tap water added with dissolved organic matter	Lab	(Zearley and Summers, 2012)
		91.8 ng/L	-12.3	110 m/d	RSF	C	WWTP secondary effluent	Full	(Nakada et al., 2007)
Estriol (E3)	Steroid estrogen	5 mg/L	15.6 ± 12	20 L/d	BSF	I	Surface water	Lab	(Kennedy et al., 2013)
		0.11~0.72 ng/L	-180~>14.7	110 m/d	RSF	C	WWTP secondary effluent	Full	(Nakada et al., 2007)
		39 ng/L	11.66 (augmented filter) -11.60 (nonaugmented filter)	0.15 m/h (3.6 m/d)	SSF	C	Surface water	Lab	(Haig et al., 2016)
Estrone (E1)	Steroid estrogen	50 μg/L	-165~31	0.05 m/h (1.2 m/d)	SSF	C	Surface water (added with WWTP primary effluent)	Pilot	(D'Alessio et al., 2015)
		5 mg/L	14.4 ± 12	20 L/d	BSF	I	Surface water	Lab	(Kennedy et al., 2013)
		19.6~40.6 ng/L	-4.59~60.5	110 m/d	RSF	C	WWTP secondary effluent	Full	(Nakada et al., 2007)
		50 ng/L	79.46 (augmented filter) 2.08 (nonaugmented filter)	0.15 m/h (3.6 m/d)	SSF	C	Surface water	Lab	(Haig et al., 2016)
Gemfibrozil	Lipid regulator	228 ± 49 ng/L	70~94	1.2, 2.4 m/h (28.8, 57.6 m/d)	RSF	C	Tap water added with dissolved organic matter	Lab	(Zearley and Summers, 2012)
		50 μg/L	3~8	0.05 m/h (1.2 m/d)	SSF	C	Surface water (added with WWTP primary effluent)	Pilot	(D'Alessio et al., 2015)
Ibuprofen	Analgesic	276 ± 176 ng/L	≥95	1.2~2.4 m/h (28.8, 57.6 m/d)	RSF	C	Tap water added with dissolved organic matter	Lab	(Zearley and Summers, 2012)
		2 μg/L	99	3 m/d	BSF	C	Reservoir water	Pilot	(Pompei et al., 2019)
		2 μg/L	100	24 L twice a week	BSF	I	Surface water	Lab	(Pompei et al., 2017)
		4.26~15.1 ng/L	30.1~95.6	110 m/d	RSF	C	WWTP secondary effluent	Full	(Nakada et al., 2007)
Iohexol	X-ray contrast agent	3.28 ± 1.3 μg/L	57 ± 3	0.012 m/h (0.288 m/d)	SSF	C	WWTP effluent	Lab	(Escolà Casas and Bester, 2015)
Iomeprol	X-ray contrast agent	20.8 ± 11 μg/L	85 ± 0.2	0.012 m/h (0.288 m/d)	SSF	C	WWTP effluent	Lab	(Escolà Casas and Bester, 2015)
Iopromide	X-ray contrast agent	556 ± 168 ng/L	3~13	1.2, 2.4 m/h (28.8, 57.6 m/d)	RSF	C	Tap water added with dissolved organic matter	Lab	(Zearley and Summers, 2012)
		2.9 ± 0.83 μg/L	58 ± 0.3	0.012 m/h (0.288 m/d)	SSF	C	WWTP effluent	Lab	(Escolà Casas and Bester, 2015)
Ketoprofen	Analgesic	95.5~299 ng/L	-186~20.5	110 m/d	RSF	C	WWTP secondary effluent	Full	(Nakada et al., 2007)
Lincomycin	Antibiotic	0.2 mg/L	<25	0.15 m/h (3.6 m/d)	SSF	C	Surface water	Pilot	(Rooklidge et al., 2005)
Methylparaben	Fungicide	2 μg/L	70	3 m/d	BSF	C	Reservoir water	Pilot	(Pompei et al., 2019)

		2 μg/L	100	24 L twice a week	BSF	I	Surface water	Lab	(Pompei et al., 2017)
Metoprolol	Hypotensor	2 μg/L	≈60~80	1 L/h (24 L/d)	SSF	C	WWTP secondary effluent	Lab	(van Gijn et al., 2021)
Naproxen	Analgesic	170 ± 101 ng/L	72~86	1.2, 2.4 m/h (28.8, 57.6 m/d)	RSF	C	Tap water added with dissolved organic matter	Lab	(Zearley and Summers, 2012)
		2 μg/L	≈20~60	1 L/h (24 L/d)	SSF	C	WWTP secondary effluent	Lab	(van Gijn et al., 2021)
		2 μg/L	97	3 m/d	BSF	C	Reservoir water	Pilot	(Pompei et al., 2019)
		2 μg/L	100	24 L twice a week	BSF	I	Surface water	Lab	(Pompei et al., 2017)
		33.0~84.9 ng/L	-11.0~58.8	110 m/d	RSF	C	WWTP secondary effluent	Full	(Nakada et al., 2007)
		Data not shown	30	14.4 m/h (345.6 m/d)	RSF	C	WWTP ozonation unit effluent	Full	(Hollender et al., 2009)
Oxytetracycline	Antibiotic	2 μg/L	15~50	0.06 m/h (1.44 m/d)	SSF	C	Surface water	Lab	(Xu et al., 2021)
p-TSA	Plasticizer	<0.05~41μg/L	93 (median)	2~6 m/h	RSF	C	Ground water	Lab	(Richter et al., 2008)
Phenazone	Analgesic	50 μg/L	0	0.05 m/h	SSF	C	Surface water (added with WWTP primary effluent)	Pilot	(D'Alessio et al., 2015)
Propranolol	Hypotensor	0.055 ± 0.015 μg/L	94 ± 2	0.012 m/h (0.288 m/d)	SSF	C	WWTP effluent	Lab	(Escolà Casas and Bester, 2015)
		2 μg/L	≈60~80	1 L/h (24 L/d)	SSF	C	WWTP secondary effluent	Lab	(van Gijn et al., 2021)
Sulfamethazine	Antibiotic	0.2 mg/L	<4	0.15 m/h (3.6 m/d)	SSF	C	Surface water	Pilot	(Rooklidge et al., 2005)
Sulfamethoxazole	Antibiotic	230 ± 33 ng/L	2.4~4.1	1.2, 2.4 m/h (28.8, 57.6 m/d)	RSF	C	Tap water added with dissolved organic matter	Lab	(Zearley and Summers, 2012)
		100 μg/L	20	288 mm/d (0.288 m/d)	SSF	C	Synthetic wastewater	Lab	(Escolà Casas et al., 2022)
		20 μg/L	17	125 mm/d (0.125 m/d)	SSF	C	Mix of 75% of secondary WWTP effluent with 25% nutritional solution	Lab	(Escolà Casas et al., 2022)
		39.9 ng/L	26.9	110 m/d	RSF	C	WWTP secondary effluent	Full	(Nakada et al., 2007)
		2 μg/L	≈20~60	1 L/h (24 L/d)	SSF	C	WWTP secondary effluent	Lab	(van Gijn et al., 2021)
		0.2 mg/L	<4	0.15 m/h (3.6 m/d)	SSF	C	Surface water	Pilot	(Rooklidge et al., 2005)
		2 μg/L	<15	0.06 m/h (1.44 m/d)	SSF	C	Surface water	Lab	(Xu et al., 2021)
Tylosin	Antibiotic	0.2 mg/L	>99	0.15 m/h (3.6 m/d)	SSF	C	Surface water	Pilot	(Rooklidge et al., 2005)
Triclosan	Antiseptic	190 ± 42 ng/L	≥90	1.2, 2.4 m/h (28.8, 57.6 m/d)	RSF	C	Tap water added with dissolved organic matter	Lab	(Zearley and Summers, 2012)
		25 μg/L	74.2	5, 10, 20 cm/h (1.2, 2.4, 4.8 m/d)	SSF	C	Synthetic wastewater	Lab	(Li et al., 2018)
		158~360 ng/L	25.2~52.5	110 m/d	RSF	C	WWTP secondary effluent	Full	(Nakada et al., 2007)
Trimethoprim	Antibiotic	175 ± 98 ng/L	83~92	1.2, 2.4 m/h (28.8, 57.6 m/d)	RSF	C	Tap water added with dissolved organic matter	Lab	(Zearley and Summers, 2012)
		16.3 ng/L	66.2	110 m/d	RSF	C	WWTP secondary effluent	Full	(Nakada et al., 2007)
		2 μg/L	≈60	1 L/h (24 L/d)	SSF	C	WWTP secondary effluent	Lab	(van Gijn et al., 2021)
		Data not shown	15	14.4 m/h (345.6 m/d)	RSF	C	WWTP ozonation unit effluent	Full	(Hollender et al., 2009)
		0.2 mg/L	>99	0.15 m/h (3.6 m/d)	SSF	C	Surface water	Pilot	(Rooklidge et al., 2005)
		2 μg/L	50~85	0.06 m/h (1.44 m/d)	SSF	C	Surface water	Lab	(Xu et al., 2021)
Warfarin	Blood anticoagulant	268 ± 24 ng/L	39~68	1.2, 2.4 m/h (28.8, 57.6 m/d)	RSF	C	Tap water added with dissolved organic matter	Lab	(Zearley and Summers, 2012)

* Removal values were summarized if different removal were found for one compound in a study.
** Both original and unified filtration rate/volume (m/d or L/d, if applicable) are displayed.
*** Filter mode. C, continuous; I, intermittent.

Supplementary Information

For

Sand and Sand-GAC Filtration Technologies in Removing PPCPs: A Review

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Captions

Figure S1. Fitting graphs of PPCP log K_{OW} with removal based on reported data of each campaign (Nakada et al., 2007).

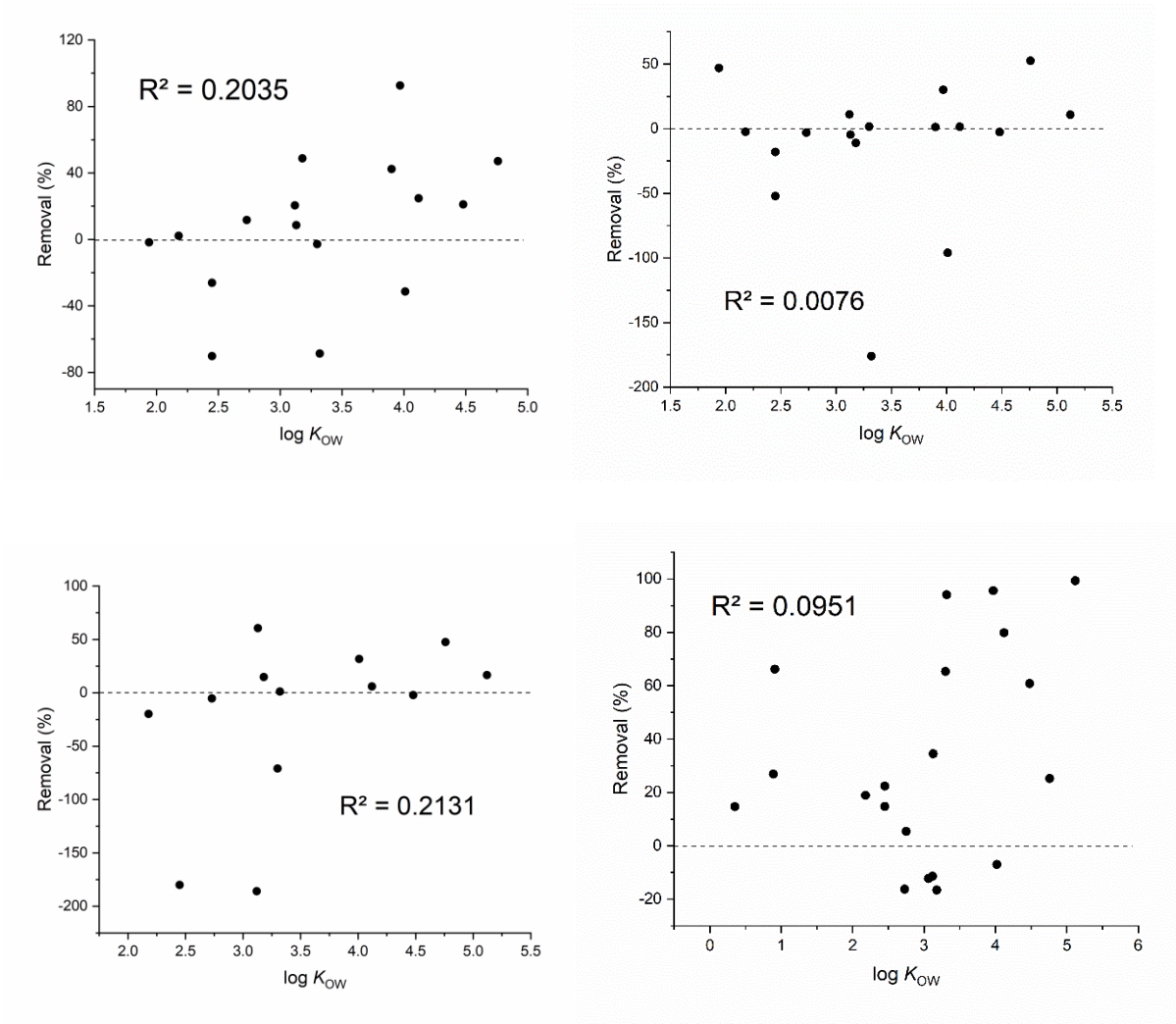

Figure. Fitting graphs of PPCP log K_{OW} with removal based on reported data of each campaign (Nakada et al., 2007). (Top left: campaign 1 (July 2013); top right: campaign 2 (November 2013); bottom left: campaign 3 (June 2004); bottom right: campaign 4 (October 2005); Data from the compounds of triclosan, thymol, naproxen, mefenamic acid, ketoprofen, fenoprofen, ibuprofen, DEET, crotamiton, carbamazepine, propyphenazone, sulfapyridine, sulfamethoxazole, trimethoprim, azithromycin, erythromycin, roxithromycin, nonylphenol, octylphenol, bisphenol A, E1, EE2 and E3. Removal of -737% of mefenamic acid was excluded from campaign 1)

Nakada, N., Shinohara, H., Murata, A., Kiri, K., Managaki, S., Sato, N., Takada, H., 2007. Removal of selected pharmaceuticals and personal care products (PPCPs) and endocrine-disrupting chemicals (EDCs) during sand filtration and ozonation at a municipal sewage treatment plant. Water Res. 41, 4373–4382. https://doi.org/10.1016/j.watres.2007.06.038