Occurrences and removal of pharmaceuticals and personal care products (PPCPs) in drinking water and water/sewage treatment plants: a review

Yi Yang[1], Yong Sik Ok[2], Ki-Hyun Kim[3], Eilhann E. Kwon[4], Yiu Fai Tsang[1*]

[1]Department of Science and Environmental Studies, The Education University of Hong Kong, Tai Po, New Territories, Hong Kong

[2]Korea Biochar Research Center & School of Natural Resources and Environmental Science, Kangwon National University, Chuncheon 24341, Korea

[3]Department of Civil and Environmental Engineering, Hanyang University, Seoul 04763, Korea

[4]Department of Environment and Energy, Sejong University, Seoul 05006, Korea

*Tel: +852-2948-8122, Fax: +852-2948-7676; E-mail: tsangyf@eduhk.hk (Y.F. Tsang)

Highlights

- There is a large variation in PPCP removal in STPs and WTPs (-157-100%).
- PPCP removal is dependent on compound characteristics and process-specific factors.
- Advanced treatment technologies are effective for PPCP removal.

Abstract

In recent years, many of micropollutants have been widely detected because of continuous input of pharmaceuticals and personal care products (PPCPs) into the environment and newly developed state-of-the-art analytical methods. PPCP residues are frequently detected in drinking water sources, sewage treatment plants (STPs), and water treatment plants (WTPs) due to their universal consumption, low human metabolic capability, and improper disposal. When partially metabolized PPCPs are transferred into STPs, they elicit negative effects on biological treatment processes; therefore, conventional STPs are insufficient when it comes to PPCP removal. Furthermore, the excreted metabolites may become secondary pollutants and can be further modified in receiving water bodies. Several advanced treatment systems, including membrane filtration, granular activated carbon, and advanced oxidation processes, have been used for the effective removal of individual PPCPs. This review covers the occurrence patterns of PPCPs in water environments and the techniques adopted for their treatment in STP/WTP unit processes operating in various countries. The aim of this review is to provide a comprehensive summary of the removal and fate of PPCPs in different treatment facilities as well as the optimum methods for their elimination in STP and WTP systems.

Key words: Contaminants of emerging concern (CECs), Endocrine disrupting chemicals (EDCs), Removal efficiency, Water quality, PPCPs

Contents

1. Introduction

In recent decades, pharmaceuticals and personal care products (PPCPs) have been recognized as contaminants of emerging concern because of their persistent presence in aquatic environments. The term “PPCPs” broadly refers to any product with healthcare or medical purposes for humans and/or animals. Interest in the safety issue of PPCPs has been steadily increased over the past 30 years (Schumock et al., 2014). PPCPs are known to be released into aquatic environments through multiple pathways, including domestic wastewater, hospital discharges, improper manufacturer disposal, sewage treatment plants (STPs), and water treatment plants (WTPs) (Leung et al., 2012; Liu and Wong, 2013). Compared with domestic sewage, hospital effluents generally exhibit higher detection frequencies and concentrations of pharmaceuticals (Kosma et al., 2010; Oliveira et al., 2015). The excreted PPCPs may either retain their original concentrations and structures or be mobilized and converted into other active (or inactive) compounds during their lifespan in aquatic matrices.

PPCPs are generally present in surface water, groundwater, drinking water, and sewage at concentrations of parts-per-trillion (ng/L) to parts-per-billion (μg/L) (Dai et al., 2015). However, the removal efficiency of PPCPs in conventional STPs is low (Behera et al., 2011), because the most commonly used treatment system in secondary STPs (i.e., activated sludge

process (ASP)) is originally designed for the removal of organic matter (i.e., BOD) and suspended solids to meet the minimum discharge standard (Hua et al., 2008; Tsang, 2015). STPs have been identified as a primary source of PPCPs in the aquatic environment (Focazio et al., 2008; Padhye et al., 2014). Although the concentration of PPCPs in sewage influent is relatively low, PPCPs that are present as either individual molecules or as complexes may exert considerably toxic or inhibitory effects on activated sludge bacteria, resulting in deteriorated removal efficiency (Thomaidi et al., 2015; 2016).

Regulation of PPCPs has been strictly enforced and implemented to minimize their consumptions (Daughton, 2002). However, the use of these products is unlikely to be restricted because of their beneficial properties for humans and animals (Jones et al., 2005). Extensive profiling of PPCPs has been pursued in aquatic environments (Boxall et al., 2012). However, data on their metabolites, by-products, and degradation products are very limited (Miao et al., 2005; Borova, et al., 2014).The fates and removal mechanisms of PPCPs in STPs and WTPs have not been fully understood (Stasinakis et al., 2013; Blair et al., 2015). Thus, numerous analytical methods have been developed to assess the profiles and occurrence patterns of PPCPs during the last decade (Evgenidou et al., 2015).

Several review articles have reported the ecotoxicological effects of PPCPs (Brausch et al., 2011) and their occurrences in various water bodies, including groundwater (Lapworth et al., 2012), surface water and wastewater (Liu and Wong, 2013), and STPs (Feng et al., 2013; Evgenidou et al., 2015). However, such studies have generally been limited to single/few treatment plants and the removal performance of the corresponding unit process. This review initially focuses on the profiles of common PPCPs in both natural and artificial environments. It is then extended to discuss the performance of PPCP removal of different treatment systems employed at each unit process in STPs and WTPs in different regions, and describe the advanced treatment methods available for effective PPCP removal. Findings from over 200 studies of 219 STPs and WTPs in the US, Asia, and Europe are summarized and discussed (Tables 2-4). Considering that differences in the operational and experimental conditions of studies may influence the results, the detailed operating conditions of various STPs and WTPs and their relevant experimental information are presented in Supplementary Materials (Tables S1 and S2).

1.1 Classification of PPCPs

PPCPs can be classified into multiple groups according to their properties and purposes. Pharmaceuticals generally include antibiotics, hormones, analgesics, anti-inflammatory drugs, blood lipid regulators, β-blockers, and cytostatic drugs. Personal care products (PCPs) include preservatives, bactericides/disinfectants, insect repellents, fragrances, and sunscreen ultraviolet (UV) filters (Kosma et al., 2010; Liu and Wong, 2013). The typical classification of PPCPs and the representative compounds are summarized in Table 1 (Esplugas et al., 2007; Liu and Wong, 2013). To date, more than 3,000 PPCPs have been used for the medical treatment of both humans and animals and for the enhancement of their living standards (Muthanna and Plósz, 2008). Numerous drugs are hydrolyzed or metabolized to form water-stabilized metabolites (Reddersen et al., 2002). In most cases, the concentrations of

these metabolites are significantly lower than the concentration of the original drug, as they tend to be more effectively consumed. However, the concentrations of some substances, such as pharmaceutical excipients, may remain almost unchanged (Hirsch et al., 1999).

1.2 Pathways of PPCPs in the environment

PPCPs can enter the environment through several pathways (Fig. 1), including STPs, industrial services, hospitals, aquaculture facilities, runoff from fields into surface waters, and runoff into soil through animal farming and manure applications (Price et al. 2010; Boxall et al., 2012; Lambropoulou et al., 2014). Untreated household effluent and treated effluents from industries and hospital services containing some partially degraded and refractory PPCPs may directly discharge into various receiving water bodies without improper treatment. The occurrence of PPCPs in aquatic environments, including sewage, surface water, groundwater, and drinking water, was reviewed recently by Luo et al. (2014). PPCP residues can also enter the environment through natural hydrologic cycle (Petrović et al., 2003; Mompelat et al., 2009).

Domestic sewage is one of the major sources of PPCPs released into the environment. Drugs used by humans or animals can be directly or indirectly discharged into the environment. Some non-metabolized or dissolved pharmaceutical ingredients (e.g., methotrexate) are excreted from the body via feces and urine (Montforts, 1999; Kim et al., 2011) and finally discharged into the sewerage systems (Kimura et al., 2007). Personal care products (PCPs), including shampoos, body washes, toothpastes, sunscreens, cosmetics, and hand lotions, can be discharged into sewerage systems and surface water through the daily washing activities of human beings. Additionally, sloughing during swimming and other recreational activities can contribute to PCP discharge (Brausch and Rand, 2011).

Other PPCP exposure pathways include the disposal of unused medicines to landfills, runoff of veterinary medicines from hard surfaces in farmyards, disposal of the carcasses of treated animals, and irrigation using reclaimed water (Fick et al. 2009; Awad et al., 2014). Moreover, the management and use practices of PPCPs vary in different regions of the world. Hence, the significance of different exposure pathways also varies geographically. For example, the connectivity of the population to STPs is limited in several regions of the world. Thus, exposure modeling based on a specific region may not be widely applicable.

2. Environmental and health risks

The widespread occurrence of PPCPs in receiving water bodies is a growing concern because of its effects on environmental and human health. PPCPs exist widely in sewage, rivers, lakes, and groundwater. They can become detrimental to human and animal health because their residues can eventually enter and accumulate in the food chain through effluent discharge and the reuse of treated sewage and sludge for agricultural applications (Rajapaksha et al., 2014; Vithanage et al., 2014). Despite the low concentrations of PPCPs in WTPs, which range between ng/L and µg/L, PPCP residues may have serious adverse health impacts, and human exposure to these chemicals has unknown long-term effects (Boxall et al., 2012).

Many PPCPs rapidly dissipate in the environment, but their extensive use results in their pseudo-persistence in water environments and serious ecological impacts on aquatic organisms (Kostich et al., 2014). High-frequency detection of PPCPs in STPs, including sewage effluent and reclaimed water (Chen et al., 2013), is caused by their universal consumption (Liu and Wong, 2013), low human metabolic capability (Borova et al., 2014), improper disposal (Ternes et al., 2004), and biologically active structures (McClellan and Halden, 2010). PPCPs may be partially metabolized or incompletely biodegraded in artificial water matrices. Thus, the excreted metabolites can become secondary pollutants and be further modified in receiving water bodies (Cardinal et al., 2014). Several PPCPs are taken up by certain plant species when reclaimed water and organic manures from sewage sludge are used. The adverse effects of these PPCPs on health and the environment have been reported previously (Tanoue et al., 2012; Jiang et al., 2013; Rajapaksha et al., 2015).

PPCP residues have been found in the edible tissues of plants when bio-solids or manure-amended soils were used or when sewage was used for irrigation (Rajapaksha et al., 2014). Although most individual PPCPs represent a *de minimis* risk to human health, the additive effect of PPCPs can potentially be hazardous. The Environmental Working Group of the United States (EWG, 2008) found 1,4-dioxane, a known carcinogen, in 28% of 27,000 PCPs. In addition, they conducted a survey of 20 girls aged 14-19 years old. The EWG determined that 16 hazardous chemicals, including synthetic musk, 2-benzenedicarboxylic salt, and Triclosan (TCS), were present in the girls' bodies due to the use of cosmetic products. A study by the United States Environmental Protection Agency (USEPA) found some drug classes of concern in the US water sources, such as antibiotics, antimicrobials, estrogenic steroids, and antiepileptic drugs (EWG, 2009; USEPA, 2009).

Certain PPCPs can lead to bioaccumulation in fish and other aquatic creatures, which triggers various unexpected interference on them. For example, chronic exposure to estrogenic pollutants in water can result in the enlargement of fish livers (Gunnarsson et al., 2009). Furthermore, single and mixed PPCP residues have been found to cause negative reproduction impacts and histopathological changes in zebrafish (Galus et al., 2013a; 2013b; Overturf et al., 2015). PPCPs also exhibit adverse cumulative effects on terrestrial and aquatic ecosystems (Hernando et al., 2004; 2006). The adverse effects of PPCPs on ecosystems are significant to human health because PPCP residues have been detected in our food chain, including fruits, vegetables, and drinking water (Hernando et al., 2006; Carmona et al., 2014; Awad et al., 2016).

3. Occurrence of PPCPs in water environments

3.1 PPCPs in surface water

Effluent from STPs is the predominant pathway through which PPCPs enter surface water in the UK (Roberts and Thomas, 2006; Gardner et al., 2012), the US (Spongberg et al., 2011), Italy (Meffe et al., 2014), and Africa (Wood et al., 2015) and accumulate in aquatic environments (Luo et al., 2014). Wang et al. (2015b) evaluated the occurrence of 36 PPCPs in urban river water samples collected from Beijing, Changzhou, and Shenzhen in China. The sum of 28 compounds, including sulfadimethoxine (164 ng/L), sulpiride (77.3 ng/L), atenolol

(52.9 ng/L), and indomethacin (50.9 ng/L), exhibited the highest median concentrations.

Overall, antibiotics comprise approximately half of PPCP contamination. The ratio of persistent PPCPs, such as sulpiride and carbamazepine, is useful for tracing contamination sources in rivers. Spongberg et al. (2011) analyzed 34 PPCPs present in 86 individual water samples collected from surface water and coastal locations in Costa Rica. The sampling sites included areas that received both treated and untreated sewage, and surface runoff. The four most frequently detected PPCPs were doxycycline (77%), sulfadimethoxine (43%), salicylic acid (41%), and TCS (34%). The PPCPs detected at higher concentrations were doxycycline (74 μg/L), ibuprofen (37 μg/L), gemfibrozil (17 μg/L), acetaminophen (13 μg/L), and ketoprofen (10 μg/L).

β-blockers are used for the treatment of bronchodilation, vasodilation, and the relaxation of visceral smooth muscles. They were found to occur frequently in the surface waters of Switzerland, with concentrations of up to ng/L (Alder et al., 2010). Most commonly found PPCPs had detection frequencies of 50-100% in Beijing, China, which is one of the most densely populated cities in the world (Dai et al., 2015). The median concentrations of the selected PPCPs were up to 4,200 ng/L. Wood et al. (2015) reported that the concentrations of antiretroviral drugs in South African surface waters were higher than in other countries. Kim et al. (2007) investigated the occurrences of 22 pharmaceuticals and 3 PCPs in 3 major rivers receiving effluents from secondary STPs located in industrialized areas in South Korea. The target PPCPs were found in all of the sampling sites in upstream and downstream, with detection frequencies from 17% to 53%. The concentrations of iopromide and caffeine were comparatively high (20-361 and 10-194 ng/L, respectively). In addition, several species, including tris(2-chloroethyl) phosphate (TCEP), iopromide, naproxen, carbamazepine, and caffeine, were frequently observed (>80%) in surface water samples.

Analgesic/anti-inflammatory drugs are one of the most common PPCP residues in sewage because of their high consumption. Moreover, these drugs, such as carbamazepine, are frequently detected in surface waters at relatively high concentrations (ng/L-μg/L) (Ashton et al., 2004; Hernando et al., 2006). These findings confirm that these stable PPCPs are difficult to remove in conventional STPs (Ashton et al., 2004; Ziylan and Ince, 2011). Therefore, they are expected to be present in similar concentrations in the influent, effluent, and downstream of the receiving water body (Feng et al., 2013).

3.2 PPCPs in groundwater

The frequencies and concentrations of PPCPs are lower in groundwater than in surface water (Vulliet and Cren-Olivé, 2011b). Of the 52 target PPCPs, erythromycin, sulfamethoxazole, fluconazole, salicylic acid, methyl paraben, TCS, and bisphenol were most frequently detected, at ng/L levels, in 70 groundwater samples collected nearby 2 multiple landfill sites in Guangzhou, China. Compared with groundwater, reservoirs were significantly more contaminated, exhibiting both higher detection frequencies and concentrations (Peng et al., 2014). Lapworth et al. (2012) discussed the five most commonly reported PPCPs in groundwater, based on studies conducted in 14 countries across Europe, the Middle East, North America, and Asia. The mean concentrations of carbamazepine, sulfamethoxazole,

ibuprofen, caffeine, and diclofenac were 5 μg/L (n=23), 252 ng/L (n=15), 1.5 μg/L (n=14), 9.8 μg/L (n=14), and 121 ng/L (n=11), respectively. In groundwater, PPCPs do not show significant trends or seasonal variations, whereas the PPCP concentrations in reservoirs are higher during spring than in other seasons. Lin et al. (2015) investigated the occurrences of contaminants of emerging concern and the correlation of their presence in groundwater with possible pollution sources in Taiwan. These authors detected most of the 50 target pharmaceuticals and perfluorinated chemicals at the ng/L level, except for 17α-ethinyl estradiol, sulfamethoxazole, and acetaminophen (i.e., 1,822, 1,820, and 1,036 ng/L, respectively). The results indicated that PPCPs with high detection frequencies and their corresponding concentrations in groundwater were consistent with the results obtained in other countries. Antibiotics, anti-inflammatories and analgesics, lipid regulators, and N, N-diethyl-m-toluamide (DEET) were frequently detected in groundwater (Sui et al., 2015). Sulfonamides are one of the most extensively studied classes of antibiotics and were found at high concentrations in several studies (García-Galán et al., 2010; Gottschall et al., 2012; Meffe and Bustamante et al., 2014).

The most commonly detected anti-inflammatories and analgesics in groundwater include ibuprofen, diclofenac, salicylic acid, carbamazepine, and paracetamol because they are widely and frequently consumed. Several pharmaceuticals and their metabolites, such as diclofenac, ibuprofen, and ketoprofen, have been found at concentrations of up to mg/L. Salicylic acid was found with a detection frequency of 98% in Guangzhou, China; its concentration ranged from 43.7 to 2,014.7 ng/L (Peng et al., 2014). Loos et al. (2010) reported that carbamazepine, which is a commonly used analgesic, was detected in 42% of groundwater samples collected from 164 locations in 23 European countries, with a maximum concentration of 390 ng/L. The detection frequencies of lipid regulators and metabolites in groundwater, such as bezafibrate (N.D.), gemfibrozil (N.D.), and clofibric acid (3%), were lower than those of antibiotics and anti-inflammatories (Peng et al., 2014). Results of a national survey of pharmaceuticals and organic pollutants in the US indicated that DEET (35%) and sulfamethoxazole (23%) were the most frequently detected PPCPs in 47 **groundwaters** across 18 states (Barnes et al., 2008). Holm et al. (1995) indicated that PPCP contamination and its subsequent ecological risks could be a serious concern for groundwater near landfill sites.

3.3 PPCPs in STPs

3.3.1 Fate of PPCPs in STPs

PPCPs are mainly released into aquatic environments through STPs before they reach the receiving soil, surface water, sediment, and groundwater (Leung et al., 2012; Liu and Wong, 2013). They are frequently detected at various concentrations in influent, effluent, reclaimed water, and receiving water bodies in Hong Kong (Li and Zhang, 2010), South China (Liu and Wong, 2013; Yin et al., 2012), Europe (Kosma et al., 2010; Jiang et al., 2013) and other regions of the world (Subedi et al., 2015a; Dotan et al., 2016; Wang et al., 2016). The potential fates of PPCPs in STPs (e.g., biodegradation/biotransformation, retention of solid/sludge, and release into receiving water bodies) are dependent on their original

chemical structures and the associated metabolites/transformation products (Jiang et al., 2013, Maia et al., 2014). Typical PPCP removal processes include ASP, tertiary treatment with nutrient removal, membrane bioreactors, and advanced oxidation processes (AOPs) (Miao et al., 2005; Tsang et al., 2007; Zhao et al., 2014). However, conventional STPs are usually inefficient in removing PPCPs because some PPCPs are specifically designed to achieve a biological response or are antimicrobial agents (McClellan and Halden, 2010; Parolini et al., 2013).

Most work has focused on investigating the occurrences and fates of PPCPs in sewage and STPs, and their elimination efficiency (Luo et al., 2014; Evgenidou et al., 2015). However, in-depth studies of the mass balance and removal mechanisms of PPCPs (e.g., biotransformation, sedimentation, adsorption, biodegradation, volatilization, and hydrolysis, etc.) in STPs, and their inhibitory effects on biological processes (e.g., ASP and nutrient removal) have not been fully established. (Carballa et al., 2007; Gao et al., 2012; Stasinakis et al., 2013; Blair et al., 2015). Therefore, it is necessary to further evaluate the effects of PPCPs on the performance of different treatment methods for different purposes (e.g., ultimate discharge or water reuse).

3.3.2 Pharmaceuticals

Given the volume of prescription, toxicity, and their presence in the environment, antibiotics, hormones, non-steroidal anti-inflammatory drugs (NSAIDs), β-blockers, blood lipid regulators, antiepileptics, analgesics and anti-inflammatories, and antidepressants are the most studied pharmaceutical groups (Miege et al., 2009; Jelić et al., 2012). Table 2a summarizes the influent and effluent concentrations of common pharmaceuticals detected in STPs in different countries.

Antibiotics are commonly used pharmaceuticals that protect humans and animals against diseases and infection caused by bacteria. Van et al. (2015) predicted that the global consumption of antibiotics in livestock would rise by 67%. Many antibiotics are released into the environment even without being metabolized. Sulfonamides, fluoroquinolones, and macrolides are persistent in sewage (Huang et al., 2011; Jelić et al., 2012). Among these antibiotics, sulfamethoxazole, ciprofloxacin, azithromycin, and tylosin are the most frequently detected species in STP effluent (Huang et al., 2011). Trimethoprim (TMP) and tetracycline (TET) exhibit high persistence in both influent and effluent, indicating low removal efficiencies in STPs (Brown et al., 2006; Watkinson et al., 2007; Leung et al., 2012).

β-blockers are common pharmaceuticals used for the treatment of cardiovascular diseases, such as angina and hypertension, and were observed in European waters in 1995 (Ternes, 1998). Subgroups, including atenolol, propranolol, and metoprolol, were also detected in the influents and effluents of STPs, demonstrating that these pharmaceuticals are not always removed effectively by STPs (Lee et al., 2007; Vieno et al., 2007a). For instance, the influent and effluent concentrations of metoprolol in Finnish STPs were 1,060 and 755 ng/L, respectively (Vieno et al., 2007a). In addition, a negative growth in the concentration of propranolol was observed in UK STPs from 60-638 ng/L to 93-288 ng/L in influent (Kasprzyk-Hordern et al., 2008; 2009; Gardner et al., 2012; 2013).

Hormones are a class of signaling molecules produced by glands in multicellular organisms. They are transported by the circulatory system to distant target organs and regulate physiology and behavior. Contamination by the natural estrogens, estrone (E1), 17β-estradiol (E2), and estriol (E3), and the synthetic contraceptive 17α-ethinylestradiol (EE2) is of great concern (Desbrow et al., 1998). The concentrations of these species are relatively low compared with previous studies. For instance, the influent concentrations of E1, E2, E3, progesterone, and testosterone were 41, 8.6, 13, 10, and 7 ng/L, respectively, in Czech Republic. The corresponding effluent concentrations were <2.5, <1, <10, <0.5, and <0.5 ng/L, respectively (Vymazal et al., 2015). These results are similar to those obtained by Mailler et al. (2015). Biodegradation, discharge into the aquatic environment with secondary effluent, and discharge with excess sludge are three possible elimination pathways for hormones from different treatment units of STPs (Belhaj et al., 2015). In addition, biodeconjugation can be an effective method to remove natural hormones in STPs (Liu et al., 2015b).

NSAIDs are a pharmaceutical class that includes analgesic (pain-killing) and antipyretic (fever-reducing) drugs. Acetaminophen, diclofenac, ibuprofen, and naproxen are prominent NSAIDs available in most countries (Paxeus, 2004; Okuda et al., 2008; Zhang et al., 2008b). Previous studies determined relatively high concentrations of acetaminophen (up to 6,000 ng/L) in different countries (Jim et al., 2006; Roberts et al., 2006; Kostich et al., 2014). Diclofenac, ibuprofen, and naproxen showed relatively low concentrations but their removal (i.e., from 40% to 80%) was also found to be ineffective (Kasprzyk-Hordern et al., 2008; 2009; Jelic et al., 2011; Carmona et al., 2014; Fernández-López et al., 2016; Papageorgiou et al., 2016). As for hormones, biodegradation/biotransformation is an effective mechanism in removing NSAIDs (Samaras et al., 2013). Other pharmaceuticals, including antiepileptic drugs and blood lipid regulators, have also been detected in sewage at relatively low concentrations (Roberts et al., 2006; Kostich et al., 2014).

The concentration of pharmaceuticals in water environments varies from different regions and their properties significantly affect treatment performance. Carmona et al. (2014) investigated the occurrences of 21 pharmaceuticals in 3 STPs in Spain. Ibuprofen, tetrahydrocannabinol, and naproxen dominated in the STP influents, with concentrations of 4,374, 2,591, and 2,399 ng/L, respectively (n=21). Average removal efficiency higher than 90% can be achieved in most pharmaceuticals. However, 11 out of 21 tested pharmaceuticals, including tetrahydrocannabinol, triclocarban, gemfibrozil, and diclofenac, were still detected in the final effluents that are exceeding the regulation standard in these STPs. Moreover, some pharmaceuticals, such as diclofenac, flufenamic acid, and gemfibrozil, consistently exhibited higher concentrations in effluents than in the corresponding influents. This may be attributed to the deconjugation of metabolites, transformation products from hydrolysis, and desorption from suspended solids/sludge during the treatment processes. These findings were in accordance with those of other studies (Miege et al., 2009; Jelić et al., 2011; 2012; Gao et al., 2012; Gracia-Lor et al., 2012; Kosma et al., 2014).

3.3.3 Personal care products

PCPs typically refer to products used for the enhancement of living standards, including

preservatives (e.g., parabens), disinfectants (e.g., TCS), insect repellents (e.g., DEET), fragrances (e.g., musks), and sunscreen UV filters (e.g., 4-methyl-benzylidene-camphor (4-MBC)). To date, many studies have monitored the presence of PCPs in STPs; however, information on the effects of PCPs on the operation of wastewater treatment processes is still lacking (Zhou et al., 2009; Lee et al., 2010). Table 2b summarizes the influent and effluent concentrations of common PCPs detected in STPs in different countries.

Parabens are esters of para-hydroxybenzoic acid, containing either an alkyl or benzyl group. They are widely used as preservatives in cosmetics, foodstuffs, and pharmaceuticals (Guo and Kannan, 2013; Li et al., 2015). Methylparaben (MeP) and propylparaben (PrP) are the most abundant parabens in STP influents, with concentrations of up to 30 μg/L and 20 μg/L, respectively (Kasprzyk-Hordern et al., 2008; González et al., 2011; Carmona et al, 2014). As they are readily biodegradable under aerobic conditions and are effectively removed in ASP, the concentrations of parabens in effluents are generally on the order of several to several tens of ng/L (Hernández et al., 2010). Daily (Carmona et al., 2014) and seasonal (Pedrouzo et al., 2009) variations in the concentrations of parabens in raw sewage are always observed because of daily consumption patterns and their widespread use as preservatives. These variations may lead to an underestimation of paraben concentrations in STPs when using time-composite sampling methods (Guo and Kannan, 2013).

TCS is a bactericide, commonly used in healthcare products, such as cosmetics, deodorants, mouth rinses, shampoos, skin-care lotions, soaps, and toothpastes, at concentrations of 0.1-0.3% (w/w) (Thompson et al., 2005). TCS is frequently detected in STP influents and effluents in various countries, at concentrations of 0.2-16.6 μg/L and 0.08-2.7 μg/L, respectively (McAvoy et al., 2002; Behera et al., 2011; Yu et al., 2013; Subedi et al., 2015a). Dissociated TCS can be easily decomposed under sunlight, with a half-life of less than one hour; however, non-dissociated TCS and methyl TCS are relatively stable to photo-degradation. Approximately half of TCS is transformed into unknown metabolites or strongly bound residues (e.g., methyl TCS) through biological methylation and finally released into water environments through effluent discharge (Lindström et al., 2002; Bester, 2007). However, compared with TCS, there is a critical lack of information regarding triclocarban (TCC) in STPs.

DEET is the most commonly used active ingredient in insect repellents and is persistent in the aquatic environment. Although DEET has been detected globally in STPs, its concentrations in influents and effluents are relatively low. The level of DEET is significantly decreased in winter due to reduced consumption (Knepper, 2004; Costanzo et al., 2007; Sui et al., 2010; Wang et al., 2014). Brausch et al. (2011) reported that DEET was found in 95% of analyzed samples, with a median concentration of approximately 0.2 μg/L. However, DEET may not accumulate in aquatic organisms as indicated by its low bioconcentration factor (Glassmeyer et al., 2005).

Fragrances have been a widely investigated group of PCPs in STPs. Synthetic musks, namely nitro musks (e.g., musk ketone (MK) and musk xylene (MX)) and polycyclic musks (e.g., HHCB and AHTN), are the most frequently used fragrance ingredients in consumer products, including deodorants, soaps, and detergents (Daughton and Ternes, 1999). Among

these musks, HHCB and AHTN are regularly detected in STP influents with relatively high concentrations of 0.043-13.7 μg/L. The detected levels of polycyclic musks exceed their toxicity limits, indicating the importance of the removal of these fragrances in wastewater treatment processes. Sun et al. (2014) investigated the occurrences and statistical distribution of HHCB and AHTN in 40 STPs in the US. The mean concentrations of HHCB and AHTN were 1.86 μg/L (0.45-4.79 μg/L) and 0.18 μg/L (0.05-0.44 μg/L), respectively, across the US. Brausch and Rand (2011) found that MK and MX were present in 83-90% of STP effluents at comparatively low concentrations.

UV filters are commonly used in sunscreens, lotions, and cosmetics to protect skin against UV radiation. Their increased usage is the result of growing concerns regarding the adverse health effects of UV radiation. UV filters are released into water environments through water-based recreational activities and effluent discharge. Several studies conducted in Switzerland found that the concentration profiles of UV filters in STP influents were similar with the order of EHMC > MBC; BP-3 > OC. UV filters were also detected in the effluents of all of the tested STPs, but their concentrations were comparatively low and their profiles were significantly different (4-MBC > BP-3 > EHMC; OC) (Poiger et al., 2004; Balmer et al., 2005). A recent study found that the occurrences and removal of 12 widely used UV filters from five STPs in Hong Kong and South China (Tsui et al., 2014). 2,4-dihydroxybenzophenone (BP-1), benzophenone-3 (BP-3), benzophenone-4 (BP-4), and EHMC were detected with frequencies over 80% in the STP influents and effluents. The overall removal efficiency of these UV filters was around 50%. In addition, higher concentrations of UV-filters are generally found in wet and summer seasons (Bester, 2007; Kasprzyk-Hordern et al., 2008; Tsui et al., 2014).

3.4 PPCPs in WTPs

To date, most studies have reported on the monitoring of PPCPs in water and wastewater treatment systems. Additionally, considerably more research has focused on STPs than on WTPs. Considering that most WTPs do not have the capabilities required for routine PPCP analysis, PPCPs are possibly present in drinking water at the concentrations with unknown effects to humans (Padhye et al., 2014). However, the fates and removal of PPCPs in WTPs and the occurrences of PPCPs in tap water have not been extensively studied because of analytical difficulties (Mompelat et al., 2009). Tables 3a and 3b summarize the influent and effluent concentrations of selected pharmaceuticals and PCPs, respectively, in WTPs in different countries. In general, raw water is not highly polluted by PPCPs (Kim et al., 2007), while the concentrations of PPCPs in treated water are typically at trace levels or below their detection limits (Huerta-Fontela et al., 2011; Vulliet et al., 2011a; 2011b).

Kim et al. (2007) investigated the presence of 14 pharmaceuticals and 3 PCPs in two full-scale conventional WTPs in South Korea. In the Seoul WTP, only 6 target PPCPs were detected in raw water, at low concentrations (i.e., 2-143 ng/L). Moreover, their concentrations were below the detection limits in treated water (either < 1 or 10 ng/L). However, in Gwangju, only oxybenzone (sunscreen) was detected, at a very low level (i.e., 1.2 ng/L). Mompelat et al. (2009) summarized the occurrence of 90 PPCPs in reservoirs, treated water, and tap water in

Germany, Italy, Canada, France, Finland, and the US. Among 90 target PPCPs, bezafibrate, clofibric acid, diclofenac, gemfibrozil, ibuprofen, and TCS were detected, at concentrations of 2.5-734 ng/L. Vulliet et al. (2011a) found that 25 PPCPs were present in drinking water. Salicylic acid was most frequently detected, while carbamazepine and atenolol were detected in >30% of the contaminated water supplies but at low concentrations (< or = 2 ng/L). Carmona et al. (2014) reported that low concentrations of PPCPs (<100 ng/L) were found in tap and mineral waters, and parabens were present at relatively high concentrations in WTPs in Valencia, Spain. In tap water samples, naproxen and salicylic acid were frequently detected, while diclofenac, PrP, and ibuprofen exhibited the highest mean concentrations (1-39 ng/L). Methylparaben was detected in mineral water, at a concentration of 40 ng/L. Liu et al. (2015a) studied the fates and removal of six antibiotics in an industrial-scale WTP equipped with advanced treatment processes in China. The influent and effluent concentrations of these antibiotics ranged from 1 to 43 ng/L and from below the detection limit (BDL) to 6 ng/L, respectively. Currently, the information of occurrences and fates of PCPs in WTPs is very limited compared with equivalent data regarding pharmaceuticals.

4. Removal of PPCPs in treatment plants

4.1 PPCP removal in STPs

Generally, conventional sewage treatment processes (Fig. 2a), including screening, degritting, primary sedimentation, aeration tanks, and final sedimentation, are ineffective in eliminating PPCPs (Carballa et al., 2004). Minus implies that the concentration of target PPCPs increases after wastewater treatment processes. Removal of PPCPs in STPs is a complicated process and depends on the chemical and biological properties of pollutants, such as hydrophilicity, solubility (Evgenidou et al., 2015), volatility, biodegradability (Jones et al., 2005), and the adsorption capability of the activated sludge (Liu and Wong, 2013). Some PPCPs (e.g., parabens) can be effectively eliminated in STPs, with an average removal rate of more than 90% (Jonkers et al., 2009; González et al., 2011). However, most PPCPs are only partially removed in conventional STPs equipped with primary and secondary treatment processes. Table 4a shows the removal efficiencies of PPCPs in different unit processes in STPs in different countries.

The capabilities of primary treatment processes (i.e., sedimentation) in removing PPCPs are very limited because of the hydrophilic nature of most PPCPs (Carballa et al., 2005; Luo et al., 2014). The removal efficiency of pharmaceuticals is comparatively lower than that of PCPs. For example, less than 28% of diclofenac and E3 was found to be removed in sedimentation tanks (Behera et al., 2011), and no considerable reduction was reported for estrone, ibuprofen, and sulfamethoxazole (Carballa et al., 2004; Gao et al., 2012). TCS removal by primary treatment varies significantly because the high water consumption rate results in short hydraulic retention time (HRT) in sedimentation tanks (McAvoy et al., 2002). Adsorption is one of the main mechanisms of PPCP removal in primary treatment processes (Suárez et al. 2008). Wang et al. (2014) investigated the removal of six PPCPs, namely caffeine, DEET, carbamazepine, metoprolol, TMP, and sulpiride in an STP. The overall removal efficiencies of these PPCPs in primary sedimentation tanks were less than 20%

owing to their hydrophilic characteristics (i.e., low water partition coefficient). Moreover, the specific size of sludge particles suitable for adsorption of PPCPs is extremely restricted (Luo et al., 2014). Therefore, primary treatment alone may be insufficient to remove PPCPs efficiently. However, up to 40% of fragrances (e.g., AHTN and HHCB) can be efficiently removed in primary treatment because of high partition coefficients between the liquid and solid phases (Stamatis and Konstantinou, 2013). Sun et al. (2014) found a strong correlation between the concentrations of AHTN and HHCB in STP effluent (r^2 = 0.71). The similar removal mechanisms of AHTN and HHCB, namely, sorption and volatilization, in STPs resulted from their similar physiochemical properties.

Secondary treatment mainly refers to biological process (e.g., ASP) and enables the removal of PPCPs through partition, adsorption, biotransformation, and biodegradation (Miao et al., 2005; McClellan and Halden, 2010; Jelić et al., 2011). The removal efficiency of PPCPs in ASP is highly dependent on the nature of PPCPs, HRT, sludge age, adsorption capacity on sludge, and reactor design (Lin et al., 2009; Bulloch et al., 2015; Evgenidou et al., 2015). Different PPCPs in the same class can exhibit significant variability in their biodegradability. McAvoy et al. (2002) reported that TCS was consistently eliminated, with a removal efficiency of >95% in ASP; however, poor and variable treatment performance was observed in biotrickling filters. No enrichment of the TCS biotransformation product, triclosan-OMe, was found in ASP, indicating that no persistent intermediates were formed. Federle et al. (2002) reported that over 80% of TCS was removed in ASP through biodegradation. Caffeine, ibuprofen, and ketoprofen were biodegraded by up to 75-87% but <25% of diclofenac was removed during secondary treatment (Salgado et al., 2012; Wang et al., 2014). Many studies have reported that the removal efficiency of DEET is around 40% in biological treatment systems (Costanzo et al. 2007; Sui et al. 2010; Zhou et al. 2009; Wang et al., 2014). Several PPCPs with low biodegradability, such as carbamazepine and TMP, are hardly biodegraded or incompletely removed in secondary treatment, regardless of the type of system used (Behera et al. 2011; Jelić et al. 2011; Wang et al., 2014). The ineffective removal of PPCPs in secondary treatment may be attributed to the transformation of PPCPs into by-products or metabolites (Miao et al., 2005) and the conjugation of target PPCPs (Carballa et al., 2004; Galán et al., 2012). Exposure to antibiotics (e.g., TMP), antibacterial agents (e.g., TCS), and β-blockers (e.g., metoprolol) can induce toxic or inhibitory effects on activated sludge bacteria (Göbel et al., 2005; Miege et al., 2009; Dann and Hontela, 2011) and alter the microbial community (Lubarsky et al., 2012; Drury et al., 2013), thereby resulting in low removal efficiency. For example, TCS is toxic to activated sludge bacteria because it inhibits the enzyme enoyl-ACP reductase, which is an essential component of the bacterial fatty acid biosynthetic pathway in bioreactors (Drury et al., 2013). Thus, ASP cannot be used to reduce PPCPs to an environmentally safe level in most of the existing secondary STPs in Hong Kong, China, and Europe (Muthanna and Plósz, 2008; Lin et al., 2009).

Several PPCPs are poorly eliminated by the secondary treatment processes; therefore, the use of the tertiary treatment processes in STPs, namely, sand filtration, AOPs, and membrane separation, is commonly necessitated to remove PPCPs prior to either chemical or UV disinfection. Transformation of PPCPs was also identified in chlorination process

(Gómez-Ramos et al., 2011).

Similar with sedimentation, sand filtration is generally ineffective for PPCP removal owing to the high hydrophilicities of most PPCPs. McAvoy et al. (2002) demonstrated that sand filter system in STPs was ineffective in removing TCS. Nakada et al. (2007) evaluated the treatment performance of 21 PPCPs in a full-scale STP equipped with sand filtration and ozonation in Tokyo. The results suggested that hydrophobicity was the controlling factor in PPCP removal. Low removal efficiencies (<50%) of PPCPs with a log/K_{ow} <3 were achieved during sand filtration, whereas PPCPs with a log/K_{ow} >3 exhibited over 80% removal in some cases. In addition, most of the target pharmaceuticals and all target antibiotics were effectively eliminated (i.e., >80%) ozonation. Several studies have suggested that oxidation is the major removal mechanism of ozonation and tertiary amino groups are susceptible to ozone attack (Huber et al., 2005; Dodd et al., 2006). These findings indicate that the removal of PPCPs via ozonation depends on their chemical structures. This may be attributed to the selective reaction of ozone with certain functional groups and the non-selective reactivity of hydroxyl radical (Papageorgiou et al., 2015). Removal of fenoprofen in ozonation system is ineffective because of the electron-withdrawing nature of the carboxylic group and the absence of electron-donating structures. The formation of by-products through the breakdown of ethoxylated and nonylphenolic compounds during ozonation is attributable to the poor removal of alkylphenol species (e.g., NP and OP) (Petrović et al., 2003).

It has been reported that effective removal of parabens (>90%) can be achieved in conventional STPs (Gorga et al., 2013; Haman et al., 2015). However, some parabens are still frequently detected in secondary effluents. For example, Li et al. (2015) investigated the fate and removal of 9 parabens and their derivatives in STPs using advanced treatment processes (i.e., ultrafiltration (UF) followed by ozonation). Only <1 to 10% of the target PPCPs were removed by UF, perhaps because these PPCP molecules are smaller than the membrane pores (Sahar et al., 2011). Several parabens were released from the membrane during backwashing or significant pH fluctuations of the influent (Caliman and Gavrilescu, 2009). However, ozonation exhibited outstanding performance (>98-100%) in removing most of the parabens, except for di-chlorinated compounds, because of the high oxidation potential of ozone. The findings were in agreement with a previous study, in which 99% of parabens were removed by ozonation with short HRTs (Tay et al., 2010). Several studies have demonstrated that UV radiation is effective in removing PPCPs. Moreover, the combination of biological processes and UV systems can considerably improve the overall treatment performance of PPCPs in STPs (Salgado et al., 2012; Wang et al., 2014).

Nakada et al. (2007) found that integration of ASP with sand filtration and ozonation is effective in removing (> 90%) most of the target PPCPs. Compared with the use of a single biological process, systems combining a bioreactor and AOPs (e.g., UV/O_3/H_2O_2) were found to noticeably enhance the removal efficiencies of cyclophosphamide (CP) and ifosfamide (IF) from 59% and 35%, respectively, to >99%. Although chemical processes, such as chlorination and ozonation, exhibit better treatment performance, the chemicals introduced in these processes may have certain levels of toxicity (Gerrity et al., 2011). In addition, chemical processes are not necessarily efficient in removing biologically active antibiotics

(e.g., clarithromycin) (Zhang et al., 2013) and light resistant UV filters (e.g., 4-MBC) (Brausch and Rand, 2011).

4.2 PPCP removal in WTPs

Compared with STPs, the performance and removal mechanisms of PPCPs in WTPs are less well characterized because they had been commonly investigated in lab-scale studies. Some authors have reported overall removal efficiencies based on the differences in concentration levels of raw and treated water (Ternes et al., 2002; Stackelberg et al., 2004; 2007). Table 4b summarizes the removal efficiencies of PPCPs in different unit processes in WTPs in different countries. Similar with STPs, PPCP removal in individual unit processes shows more significant variations than the overall PPCP removal in WTPs. The removal efficiency is mainly dependent on the specific processes used in WTPs (Boleda et al., 2011; Padhye et al., 2014). In general, conventional water treatment processes (Fig. 2b), including coagulation/flocculation, sedimentation, and sand filtration, are ineffective in removing PPCPs (<30%) (Huerta-Fontela et al., 2011; Diemert and Andrews, 2013). The removal efficiencies of antibiotics in conventional WTPs are even less than 10% (Liu et al., 2015a).

Advanced treatment technologies, such as ozonation, activated carbon adsorption, and reverse osmosis (RO), are applicable to PPCP removal in WTPs (Heberer et al., 2002; Snyder et al., 2003; Lee et al., 2008; Huerta-Fontela et al., 2011). However, ozonation may produce unknown degradation products (Westerhoff et al., 2005). Zwiener and Frimmel (2000) suggested that AOPs can significantly enhance the removal of PPCPs, especially pharmaceuticals. However, their efficiencies are often limited by the radical scavenging capacity and ozone consumption of organic matter present in the water (Zwiener and Frimmel, 2000). Huerta-Fontela et al. (2011) reported that PPCPs with high hydrophobicities could be effectively eliminated by GAC filtration. Lin et al. (2016) investigated the occurrences and removal of 39 PPCPs in a WTP equipped with ozonation and GAC filtration. Most of the 14 PPCPs detected in raw water were completely removed by the advanced WTP. The removal efficiencies of caffeine, indomethacin, and sulfamethoxazole were 89.5%, 84.2%, and 92.2%, respectively. The results of principal component analysis also suggested that oxidation, coagulation combined with sedimentation, and filtration were the major removal mechanisms in the advanced WTP. These findings were consistent with a previous study, in which ozonation was found to be highly effective for PPCP removal (Hollender et al., 2009).

Biofilters, which can be simply fabricated by converting granular media filters, have been demonstrated to be effective for PPCP removal (Zuehlk et al., 2007; Meffe et al., 2010; Zearley and Summers, 2012). McKie et al. (2016) performed a pilot-scale study to evaluate the PPCP removal in WTPs equipped with biofilters. Compared with conventional dual-media filtration, biofiltration systems with and without coagulant addition successfully improved the PPCP removal from 13% to 39% and 70%, respectively. The treatment performance of biofilters for PPCP removal may be enhanced using low doses of in-line coagulant without adversely affecting headloss (Azzeh et al., 2015).

5. Control strategies for PPCP contamination

5.1 Membrane filtration

Membrane filtration processes, such as nanofiltration (NF) and RO, are promising alternatives for the elimination of PPCPs from wastewater (Nghiem et al., 2004; Yoon et al., 2006; Yoon et al., 2010). UF and microfiltration (MF) have been proven to remove PPCPs. However, their removal performances are relatively poor because membrane pore sizes are considerably larger than PPCP molecules. For comparison, pressure-driven membrane processes, NF and RO, were applied to the drinking water treatment (Watkinson et al., 2007). These processes generally show significant PPCP removal efficiencies; however these membranes are still slightly permeable to some relatively small pollutants (Schäfer et al., 2011).

The removal capabilities of two different types of submerged NF flat sheet modules for removal of pharmaceuticals from STPs were investigated (Röhricht et al., 2009). Approximately 60% of diclofenac and naproxen were retained by both types of membranes, whereas only a small proportion of carbamazepine was removed. Hence, diclofenac and naproxen may be obstructed by the negatively charged membrane surface, whereas carbamazepine may not (Nghiem et al., 2005).However, these removal efficiencies may not be sufficient to justify the use of such a system as an additional treatment step in STPs. For more polar compounds, the NF membrane showed higher removal efficiencies than the UF membrane. The removal of selected PPCPs by NF and RO has also been compared in previous studies (Yangali-Quintanilla et al., 2011). The average retention efficiency of NF is 82% for neutral pollutants and 97% for ionic contaminants, whereas RO can achieve 85% to 99%. Real et al. (2012) compared the efficiencies of different system configurations in the elimination of PPCPs from selected water sources. When ozonation was combined with NF, the removal efficiency was significantly affected by such variables as ozone dose and treatment sequences. For instance, NF followed by ozonation removed more than 97% of pollutants from natural water, with an ozone dose of 2.25 mg/L and more than 90% from secondary effluent, with an ozone dose of 3.75 mg/L. In contrast, a high removal efficiency (> 70% in the permeate stream) was achieved by ozonation with initial dose of 2.25 mg/L followed by NF in natural waters (Watkinson et al., 2007). Although NF and RO processes exhibit efficient PPCP removal, pollutants in a highly concentrated form remaining in the retentate require further treatment.

5.2 Granular activated carbon

Granular activated carbon (GAC) and powdered activated carbon (PAC) were investigated for the sorptive removal of PPCPs (Yang et al., 2011; Boehler et al., 2012; Margot et al., 2013). GAC is typically used in rapid filters, whereas PAC is an efficient method in removing seasonally occurring taste and odor in WTPs (Scheurer et al., 2010; Zoschke et al., 2011). In this review, we focus on GAC because it has been used widely in drinking water treatment and tertiary treatment in STPs. Stackelberg et al. (2007) found that GAC facilities in a conventional WTP accounted for 53% removal of the tested PPCPs, whereas disinfection and sedimentation accoutred for 32% and 15%, respectively.

669 In a study by Hernández-Leal et al. (2011), the removal efficiencies for tonalide and
670 nonylphenol ranged from 50% to >90% (galaxolide). Contact time was found to markedly
671 affect the extent of carbon adsorption. Short contact times resulted in low removal
672 efficiencies. Correspondingly, long contact times increase surface loading and the number of
673 accessible adsorption sites (Bolong et al., 2009; Meinel et al., 2015). In general, adsorption
674 by activated carbon has greater potential for removal of antibiotics than coagulation and
675 flocculation processes (Choi et al., 2008).
676 Activated carbon has also demonstrated as an effective advanced treatment process in
677 removing PPCP residues from treated effluents. Ek et al. (2014) conducted a pilot-scale study
678 to evaluate the performance of activated carbon in removing pharmaceutical residues from
679 treated wastewater. The results suggested that activated carbon beds with 90-98% PPCP
680 removal may be a competitive alternative to treatment with ozone. Similar conclusions were
681 drawn by Grover et al. (2011), who studied the removal of pharmaceuticals from sewage
682 effluent in a full-scale STP. 43-64% of steroidal estrogens were successfully removed by
683 GAC. The elimination rates varied for different types of pharmaceuticals; for example, the
684 removal efficiencies of mebeverine and diclofenac were 84%-99%. In contrast,
685 carbamazepine and propranolol exhibited relatively low removal rates of 17%-23%.
686 Paredes et al. (2016) assessed the treatment of secondary effluents using sand and GAC
687 biofilters. Several reactors were used to determine the contributions of adsorption and
688 biotransformation to the removal of several PPCPs. The PPCP removal mechanisms were
689 classified into three different categories: (I) biotransformation and high adsorption on GAC
690 and sand (e.g., galaxolide, tonalide, celestolide, and TCS), (II) biotransformation and high
691 adsorption on GAC, but either low or null adsorption on sand (e.g., ibuprofen, naproxen,
692 fluoxetine, erythromycin, roxythromycim, sulfamethoxazole, TMP, bisphenol A, E1, E2, and
693 EE2), and (III) adsorption on GAC alone (e.g., carbamazepine, diazepam, and diclofenac).
694 When choosing the most appropriate PPCP treatment process, the high operating cost, the
695 clogging problem, and the associated hydraulic capacity limits should be considered (Ek et al.,
696 2014).
697
698 **5.3 Advanced oxidation processes**
699 AOPs, such as ozonation, UV, photocatalysis, and Fenton reaction, have been used for
700 drinking water treatment (e.g., odor/taste control and disinfection) and to lesser extent in
701 wastewater disinfection (Huber et al., 2003; Klavarioti et al., 2009; Gerrity et al., 2010).
702 AOPs may change the polarity and functional groups of the target PPCPs (McMonagle, 2013;
703 Papageorgiou et al., 2014). Thus, AOPs are suitable for water reuse purposes that involve
704 direct human contact, such as household wastewater reuse applications (Hernández-Leal et al.,
705 2011). It has been reported that WTPs equipped with AOPs further eliminated PPCPs.
706 Compounds, such as caffeine, indomethacin, and sulfamethoxazole, were removed at
707 efficiencies of 89.5%, 84.2%, and 92.2%, respectively (Lin et al., 2016).
708 A study on pilot-scale experiments in a WTP was conducted by Borikar et al. (2015).
709 The results indicated that conventional WTPs equipped with either ozone/H_2O_2 or UV/H_2O_2
710 greatly improved PPCP removal from 26% to 97% or 92%, respectively. Among the tested

PPCPs, carbamazepine, fluoxetine, naproxen, gemfibrozil, and TCS showed near complete removal. Diclofenac and ibuprofen were also removed by up to 97% and 98%, respectively. However, pharmaceuticals demonstrated some resistance in that, the highest removal of atorvastatin was only 88%. Fast (2015) conducted a holistic analysis, including a ranking system, to determine the performance of several AOPs. The findings indicated that H_2O_2/ozone presented the highest average ranking in reducing PPCPs. In addition, performance improved significantly when oxidation was combined with other unit processes. Česen et al. (2015) demonstrated that removal rates of 99% for CP and 94% for IF were be achieved using a UV/O_3/H_2O_2 system with 5 g/L of H_2O_2 for 120 min. By coupling this AOP with a biological treatment, the removal rates of CP and IF could be further enhanced >99%. Real et al. (2012) demonstrated that a combined process using UV radiation (254 nm; for 30 min) and NF was very effective, with removal rates of >80% in the majority of the experiments. However, some recent reports have expressed substantial concern regarding the application of AOPs for PPCP removal. For example, Huang et al. (2015) indicated that ibuprofen oxidation products generated a higher risk of acute toxicity than their parent chemical. Yang et al. (2016) evaluated the performance of UV/chlorine and UV/H_2O_2 processes in water purification to degrade PPCP residues after sand filtration. The results showed that UV/chlorine exhibited superior PPCP removal and disinfection byproducts (DBPs) were formed after chlorination.

6. Conclusion

In recent years, PPCPs in water environments have been recognized as an important environmental issue. Many studies have focused on the occurrences and fates of PPCPs in STPs and WTPs, in which trace concentrations ranging from nanograms to micrograms per liter have been detected. Surface water and groundwater polluted by PPCPs may be attributed to sewage discharge and limited water treatment intended to reduce direct discharge. In some areas, PPCPs have also been detected in drinking water or treated water from WTPs. To protect the water sources from contamination is the first priority in future development in water supply services. Conventional STPs are originally intended to remove organic matter and suspended solids. Consequently, high concentrations of PPCPs in sewage effluent, excess sludge, and reclaimed water could be ultimately introduced into aquatic environments and the food chain. However, there is limited information about the removal mechanisms of PPCPs in STPs and WTPs and their corresponding inhibitory effects in biological processes. Recently, some advanced technologies, namely membrane filtration, carbon adsorption, and AOPs, have been widely adopted for PPCP removal. However, the performance and cost of different unit processes vary by case. Therefore, it is necessary to evaluate the effects of PPCPs on treatment performance, process stability, and microbial community structure of biological processes in STPs and WTPs. The results could provide a theoretical basis for the optimization of existing treatment systems with varying design and could significantly contribute to protecting the receiving water bodies and promoting the use of reclaimed water.

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Table 1. Typical classes of PPCPs and their representative compounds

Typical classes	**Representative compounds**
A. Pharmaceuticals	
A1. Broad-spectrum antibiotics	
1a	Levofloxacin
1b	Penicillin
A2. Hormones	
2a	17-β-estradiol (E2)
2b	Estriol (E3)
2c	Estrone (E1)
A3. Non-steroidal anti-inflammatory drugs (NSAIDs)	
3a	Diclofenac
3b	Ibuprofen
3c	Naproxen
A4. β-blockers	
4a	Metoprolol
4b	Propranolol
A5. Blood lipid regulators	
5a	Clofibric acid
5b	Gemfibrozil
B. Personal care products	
B1. Preservatives	
6a	Parabens
B2. Bactericides/disinfectants	
7a	Methyltriclosan
7b	Triclocarban (TCC)
7c	Triclosan (TCS)
B3. Insect repellents	
8a	N, N-diethyl-m-toluamide (DEET)

B4. Fragrances	
9a	Galaxolide fragrance (HHCB)
9b	Toxalide fragrance (AHTN)
B5. Sunscreen UV filters	
10a	2-ethyl-hexyl-4-trimethoxycinnamate (EHMC)
10b	4-methyl-benzylidene-camphor (4-MBC)
10c	Octyl-methoxycinnamate (OMC)
10d	Octyl-triazone (OC)

Table 2a. The concentrations and removal (%) of selected pharmaceuticals in conventional STPs in different countries

Order	**Representative compounds**	**Influent (ng/L)**	**Final effluent (ng/L)**	**Overall removal (%)**	**Sludge (ng/kg)**	**Location**	**References**
A. Antibiotics							
1a	Amoxicillin (AMOX)	ND	ND	NA		Hong Kong, Stonecutters	Leung et al. (2012)
		261±3	66±2	74		Hong Kong, Tai Po	Leung et al. (2012)
		ND	ND	NA		Hong Kong, Sha Tin	Leung et al. (2012)
1b	Ampicillin	ND-1805	ND-498	72		Greece, Volos	Papageorgiou et al. (2016)
1c	Cefalexin (CFX)	ND	ND	NA		Hong Kong, Stonecutters	Leung et al. (2012)
		ND	ND	NA		Hong Kong, Tai Po	Leung et al. (2012)
		40±5	ND	>90		Hong Kong, Sha Tin	Leung et al. (2012)
1d	Chloramphenicol (CAP)	206±56	234±63	-14		Hong Kong, Stonecutters	Leung et al. (2012)
		11	ND			Spain, Valencia	Carmona et al. (2014)
		28±3	3.3±0.6	88		Hong Kong, Tai Po	Leung et al. (2012)
		109±53	ND	>99		Hong Kong, Sha Tin	Leung et al. (2012)
1e	Ciprofloxacin		67	NA		USA (20 states)	Kostich et al. (2014)
1f	ERY-H_2O	460±224	455±194	1		Hong Kong, Stonecutters	Leung et al. (2012)
		315±3	533±24	13		Hong Kong, Tai Po	Leung et al. (2012)
		707±35	708±274	~~0~~		Hong Kong, Sha Tin	Leung et al. (2012)
1g	Erythromycin (ERY)	ND-320	ND			Greece, Volos	Papageorgiou et al. (2016)
1h	Levofloxacin	180	10	50-80	210	UK (160 STPs)	Gardner et al. (2012; 2013)
1i	Norfloxacin (NOR)	680±181	364±159	46		Hong Kong,	Leung et al. (2012)

						Stonecutters	
		48±19	33	31		Hong Kong, Tai Po	Leung et al. (2012)
		275±11	77±0.4	72		Hong Kong, Sha Tin	Leung et al. (2012)
			160			USA (20 states)	Kostich et al. (2014)
1j	Ofloxacin (OFX)	1020±243	980±240	4		Hong Kong, Stonecutters	Leung et al. (2012)
		220±71	202±134	8		Hong Kong, Tai Po	Leung et al. (2012)
		275±11	707±35	-157		Hong Kong, Sha Tin	Leung et al. (2012)
1k	Roxithromycin (ROX)	120	120	0		Hong Kong, Stonecutters	Leung et al. (2012)
		ND	ND	NA		Hong Kong, Tai Po	Leung et al. (2012)
		126±0.4	142±5	-13		Hong Kong, Sha Tin	Leung et al. (2012)
1l	Sulfamethazine (SMX)	110±45	110±36	0		Hong Kong, Stonecutters	Leung et al. (2012)
		140±3	37±6	74		Hong Kong, Tai Po	Leung et al. (2012)
		39±0.7	8±3	79		Hong Kong, Sha Tin	Leung et al. (2012)
		ND-507	ND-80	84		Greece, Volos	Papageorgiou et al. (2016)
			12			USA (20 cities)	Kostich et al (2014)
1m	Tetracycline (TET)	257±176	152±59	44		Hong Kong, Stonecutters	Leung et al. (2012)
		77±24	ND	>90		Hong Kong, Tai Po	Leung et al. (2012)
		25±8	14±3	44		Hong Kong, Sha Tin	Leung et al. (2012)
1n	Trimethoprim (TMP)	95±23	91±28	4		Hong Kong, Stonecutters	Leung et al. (2012)
		114±5	68±4	40		Hong Kong, Tai Po	Leung et al. (2012)
		124±12	68±38	45		Hong Kong, Sha	Leung et al. (2012)

						Tin	
B. Antiepileptic drugs							
2a	Carbamazepine		97	NA		USA (20 cities)	Kostich et al. (2014)
		15780	7570	52		Spain, Murcia	Fernández-López et al. (2016)
C. Blood lipid regulators							
3a	Gemfibrozil		420	NA		USA (20 cities)	Kostich et al (2014)
D. β-Blockers							
4a	Propanolol	60-638	93-388	-50-44	170	UK (162 STPs); UK, South Wales	Gardner et al. (2012;2013);
E. Hormones							
5a	Estrone (E1)	7	9	-28		France	Mailler et al. (2015)
		41	<2.5	>94		Czech Republic	Vymazal et al. (2015)
5b	17β-estradiol (E2)			1.1-1.2		USA	Belhaj et al. (2015)
		8.6	<1	>88		Czech Republic	Vymazal et al. (2015)
	Estriol (E3)	13	<10	>23		Czech Republic	Vymazal et al. (2015)
F. NSAIDs							
6a	Acetaminophen		300	NA		USA (20 cities)	Kostich et al. (2014)
6b	Diclofenac	1660	430	74		Spain, Murcia	Fernández-López et al. (2016)
		400-1500		NA		Spain, Catalonia	Jelić et al. (2011)
		ND-4869	ND-2668	45		Greece, Volos	Papageorgiou et al. (2016)
6c	Ibuprofen	1681-33,764	143-4239	>80	380	UK, Bath	Petrie et al. (2015)
			460	NA		USA (20 cities)	Kostich et al. (2014)
		4374	ND	>99		Spain, Valencia	Carmona et al. (2014)
		2800	720	72		Spain, Murica	Fernández-López et al. (2016)
		1100-2300	400-1000	64-56		Spain, Catalonia	Jelić et al. (2011)
		ND-793	ND-220	72		Greece, Volos	Papageorgiou et al. (2016)

6d	Naproxen	1180	190	84		Spain, Murcia	Fernández-López et al. (2016)
		4200-7200		NA		Spain, Catalonia	Jelić et al. (2011)
		2399	102	>90		Spain, Valencia	Carmona et al. (2014)

ND = Not detectable; NA = Not available

Table 2b. The concentrations and removal (%) of selected PCPs in conventional STPs in different countries

Order	**Representative compounds**	**Influent (ng/L)**	**Final effluent (ng/L)**	**Overall removal (%)**	**Sludge (ng/kg)**	**Location**	**References**
A. Bactericides/Disinfectants							
1a	Triclosan (TCS)	892	202	77	645	India (2 states)	Subedi et al. (2015a)
		2300	48	>90		USA, California	Yu et al. (2013)
		547	112	79		Korea, Ulsan	Behera et al. (2011)
		300	NA	55		USA	Blair et al. (2015)
1b	Triclocarban (TCC)	1150	49	>80	5570	India (2 states)	Subedi et al. (2015a)
		540	NA	11		USA	Blair et al. (2015)
B. Fragrances							
2a	Calaxilid Fragrance (HHCB)	2560-4520	NA	61->99		Korea, Busan	Lee et al. (2010)
2b	Toxalide Fragrance (AHTN)	550-1210		NA		Korea, Busan	Lee et al. (2010)
C. Insect repellents							
3a	DEET	600-1200	60-624	69±21		China, Beijing	Sui et al. (2010)
		66	40	40		China, Shanghai	Wang et al. (2014)
D. Preservatives							
4a	Butylparaben (BuP)	15-27	3	>80		China,Guangzhou	Yu et al. (2011)
		160-170	1	>99		China, Guangzhou	Yu et al. (2011)
4b	Methylparaben (MeP)	290-10000	6-50	>90		Spain (northwest)	González et al. (2011)
		334	11	96		Spain, Valencia	Carmona et al. (2014)
		36.8; 97.9	0.14; 0.14	99.7; 99	41.6; 58.5	New York, USA (2 STPs)	Wang et al., (2016)
4c	Propylparaben (PrP)	520-2800	2-210	>90		Spain (northwest)	González et al. (2011)

		1630	<5	99		Spain, Valencia	Carmona et al. (2014)
E. Sunscreen UV filters							
5a	4-methyl-benzilidine-camphor (4MBC)	169	43	12 (n=60)	49	Hong Kong (5 regions)	Tsui et al. (2014)
5b	2-ethyl-hexyl-4-trimethoxycinnamate (EHMC)	462	150	93 (n=60)	68	Hong Kong (5 regions)	Tsui et al. (2014)
		309	126	59		Hong Kong (5 regions)	Tsui et al. (2014)
		601	347	42		Hong Kong (5 regions)	Tsui et al. (2014)
5c	Butyl methoxydibenzoylmethane (BMDM)	289	147	49		Hong Kong (5 regions)	Tsui et al. (2014)
5d	Ethylhexyl salicylate (EHS)	93	8	91		Hong Kong (5 regions)	Tsui et al. (2014)
5e	Homosalate (HMS)	151	31	79		Hong Kong (5 regions)	Tsui et al. (2014)
5f	Isoamyl p-methoxycinnamate (IAMC)	43	24	44		Hong Kong (5 regions)	Tsui et al. (2014)
5g	Octyl dimethyl-p-aminobenzoic acid (ODPABA)	138	56	17		Hong Kong (5 regions)	Tsui et al. (2014)
5h	Octocrylene (OC)	8	0	>99		Hong Kong (5 regions)	Tsui et al. (2014)
5i	Oxycodone (OXB)	ND	41.2	1.53		India (2 states)	Subedi et al. (2015a)

ND = Not detectable; NA = Not available

Table 3a. The concentrations and removal (%) of selected pharmaceuticals in conventional WTPs in different countries

Order	Representative compounds	Raw water (ng/L)	Treated water (ng/L)	Overall removal (%)	Mineral waters (ng/L)	Tap waters (ng/L)	Location	References
A. Antibiotics								
1a	Clarithromycin	40.1-54.4	ND	>99			Spain (Northeast)	Boleda et al. (2011)
1b	Chloramphenicol				1	2	Spain, Valencia	Carmona et al. (2014)
1c	Erythromycin	21-33	1.3-2.0	>90			Spain (Northeast)	Boleda et al. (2011)
1d	Sulfamethoxazole	57-149	ND	>99			Spain (Northeast)	Boleda et al. (2011)
			0.41	NA		0.37	USA, New York	Subedi et al. (2015b)
		4	ND	>99			France (8 WTPs)	Vulliet et al. (2011a)
1e	Sulfadimethoxine	ND-8.3	ND	>99			Spain (Northeast)	Boleda et al. (2011)
B. Antiepileptic drugs								
2a	Carbamazepine	144-215	1.0-1.4	>99			Spain (one city in south-eastern)	Azzouz et al. (2013)
						43.2	France, Marseilles	Togola and Budzinski (2008)
		33	8	75			France (8 WTPs)	Vulliet et al. (2011a)
C. Analgesics and anti-inflammatory drugs								
3b	Acetylsalicylic acid	21-54	<0.1	>99			Spain (one city in south-eastern)	Azzouz et al. (2013)
3c	Diclofenac	9	ND	>99			France (8 WTPs)	Vulliet et al. (2011a)
3d	Ibuprofen				12	39	Spain, Valencia	Carmona et al. (2014)

		6.6	1.3	80			France (8 WTPs)	Vulliet et al. (2011a)
D. Blood lipid regulators								
4a	Clofibric acid	11.5-20	ND	>99			Spain (Northeast)	Boleda et al. (2011)
		68	<0.1	>99			Spain (one city in south-eastern)	Azzouz et al. (2013)
4b	Gemfibrozil	187-326	ND	>99			Spain (Northeast)	Boleda et al. (2011)
E. β-Blockers								
5a	Metoprolol	2	ND	>99			France (8 WTPs)	Vulliet et al. (2011a)
F. Hormones								
7a	Estrone (E1)	77-120	<0.15				Spain (one city in south-eastern)	Azzouz et al. (2013)
7b	17-β-estradiol (E2)	35-101	<0.15				Spain (one city in south-eastern)	Azzouz et al. (2013)
7c	Ethinylestradiol (EE2)	10-97	<0.2				Spain (one city in south-eastern)	Azzouz et al. (2013)
G. Non-steroidal anti-inflammatory drugs (NSAIDs)								
8a	Acetaminophen	163-260	3-16				Spain (Northeast)	Boleda et al. (2011)
						210	France, Marseilles	Togola and Budzinski (2008)
		7-37.1	ND-6.4	>90			China, Taihu	Lin et al. (2016)
8b	Diclofenac				25	18	Spain, Valencia	Carmona et al. (2014)
		210-316	45-68				Spain (one city in south-eastern)	Azzouz et al. (2013)
						2.5	France, Marseilles	Togola and Budzinski (2008)

8c	Ibuprofen				12	39	Spain, Valencia	Carmona et al. (2014)
		81-230	ND				Spain (Northeast)	Boleda et al. (2011)
		257-357	74-102				Spain (one city in south-eastern)	Azzouz et al. (2013)
						0.6	France, Marseilles	Togola and Budzinski (2008)
8d	Ketoprofen	133-250	20-37				Spain (one city in south-eastern)	Azzouz et al. (2013)
						3	France, Marseilles	Togola and Budzinski (2008)
8e	Naproxen	0.9	ND				USA (17 WTPs)	Benotti et al. (2008)
		3.1	ND				France (8 WTPs)	Vulliet et al. (2011a)
		99-152	ND				Spain (Northeast)	Boleda et al. (2011)
		71-321	0.5-2.4				Spain (one city in south-eastern)	Azzouz et al. (2013)
					25	11	Spain, Valencia	Carmona et al. (2014)
H. Additional pharmaceuticals								
9a	Flofeniol	24-111	<0.04				Spain (one city in south-eastern)	Azzouz et al. (2013)
9b	Flunixin	69-145	<0.03				Spain (one city in south-eastern)	Azzouz et al. (2013)
9c	phenylbutazone	67-98	<0.15				Spain (one city in south-eastern)	Azzouz et al. (2013)
9d	pyrimethamine	21-57	<0.15				Spain (one city in south-eastern)	Azzouz et al. (2013)

NA = Not available; ND = Not detectable

Table 3b. The concentrations and removal (%) of selected PCPs in conventional WTPs in different countries

Order	Representative compounds	Raw water (ng/L)	Treated water (ng/L)	Overall removal (%)	Mineral waters (ng/L)	Tap waters (ng/L)	Location	References
A. Parabens								
1a	Butylparaben				36	28	Spain, Valencia	Carmona et al. (2014)
1b	Ethylparaben				2	ND	Spain, Valencia	Carmona et al. (2014)
1c	Methylparaben				40	12	Spain, Valencia	Carmona et al. (2014)
1d	Propylparaben				23	9	Spain, Valencia	Carmona et al. (2014)
1e	Methyltriclosan	74	ND	>99			China (polit)	Zhao et al. (2014)
B. Bactericides/Disinfectants								
2a	Triclosan (TCS)				4	ND	Spain, Valencia	Carmona et al. (2014)
		ND				ND	USA, New York	Subedi et al. (2015b)
		3	ND	>99			USA (17 WTPs)	Benotti et al. (2008)
		74-102	<0.1	>99			Spain (one city in south-eastern)	Azzouz et al. (2013)
		1230	100	92			Israel and Palestin	Dotan et al. (2016)
2b	Triclocarban (TCC)				12	13	Spain, Valencia	Carmona et al. (2014)
		7.18				5.4	USA, New York	Subedi et al. (2015b)
C. Insect repellents								

3a	DEET	85	49	42			USA (17 WTPs)	Benotti et al. (2008)
		19.8-78.4	ND	>99			China, Taihu	Lin et al. (2016)
D. Sunscreen UV filters								
4a	Oxybenzone	19.4				1.39	USA, New York	Subedi et al. (2015b)

NA = Not available; ND = Not detectable

Table 4a. The removal (%) of PPCPs in different unit processes in STPs in different countries

Order	Representative compounds	Preliminary treatment (%)	Primary treatment (%)	Secondary treatment (%)	Tertiary treatment (%)	Location	References
A. Bactericides/disinfectants							
1a	Triclosan (TCS)			77	18	Greece, Agrinio	Stamatis and Konstantinou (2013)
			42	97		Australia, Canberra	Roberts et al. (2016)
			-20	75	-25	Korea, Ulsan	Behera et al. (2011)
B. Broad-spectrum antibiotics							
2a	Acetaminophen (AMP)	6	8	>99	>99	USA, Michigan	Gao et al. (2012)
2b	Carbamazepine (CBZ)		-19	-42	-41	USA, Michigan	Gao et al. (2012)
			-36	22	NA	Australia, Canberra	Roberts et al. (2016)
2c	Codeine			53-83.2		China (Southern and Northern)	Zhao et al. (2013)
2d	Chlortetracycline (CTC)	5	32	>99	>99	USA, Michigan	Gao et al. (2012)
2e	Doxycycline (DOC)	35	40	64	50	USA, Michigan	Gao et al. (2012)
2f	Lincomycin (LCM)		-31	2	40	USA, Michigan	Gao et al. (2012)
2g	Oxytetracycline (OTC)	28	8	64	39	USA, Michigan	Gao et al. (2012)
2h	Sulfadiazine (SDZ)	1	2	22	27	USA, Michigan	Gao et al. (2012)
2i	Sulfamerazine (SMR)	>99	NA	>99	>99	USA, Michigan	Gao et al. (2012)
2j	Sulfamethoxazole (SMX)	18	17	69	89	USA, Michigan	Gao et al. (2012)
2k	Sulpiride (SP)		8	-33	5	China, Shangha	Wang et al. (2014)
2l	Tetracycline (TC)	59	50	>99	>99	USA, Michigan	Gao et al. (2012)
2m	Trimethoprim (TMP)		12	10	9	China, Shangha	Wang et al. (2014)

C. Hormones							
3a	Estradiol		NA	93±14	>99	Korea, Ulsan	Behera et al. (2011)
3b	Estriol		45	90±11	>99	Korea, Ulsan	Behera et al. (2011)
D. Insect repellents							
4a	DEET		-22	43	15	China, Shangha	Wang et al. (2014)
			5	93	92	China, Beijing	Gao et al. (2016)
E. Nonsteroidal anti-inflammatory drugs (NSAIDs)							
5a	Ibuprofen			90	25	Greece	Stamatis and Konstantinou (2013)
F. Preservatives							
6a	Mathylparaben		71.6			China (Southern and Northern)	Zhao et al. (2013)

NA = Not available; ND = Not detectable

Table 4b. The removal (%) of PPCPs in different unit processes in WTPs in different countries

Order	**Representative compounds**	**Step 1**	**Step 2**				
		Coagulation and flocculation (%)	**Sand filtration and chlorination (%)**	**Advanced treatment processes**		**Location**	**References**
				GAC (%)	**Ultrafiltration and reverse osmosis (%)**		
A. Bactericides/ disinfectants							
1a	Triclosan	89	86.6			Spain (one city in south-eastern)	Azzouz et al. (2013)
B. Blood lipid regulators							
2a	Metoprolol	8	11	>95		Finland, Helsinki	Vieno et al. (2007b)
2b	Pravastatin			91	91	Spain (Northeast)	Boleda et al. (2011)
C. Broad-spectrum antibiotics							
3a	Acetaminophen			96	99	Spain (Northeast)	Boleda et al. (2011)
		46	86			Canada, Ontario	McKie et al. (2016)
3b	Acetylsalicylic acid	83	NA			Spain (one city in south-eastern)	Azzouz et al. (2013)
			8			Finland, Helsinki	Vieno et al. (2007b)
3c	Azithromycin			99	99	Spain (Northeast)	Boleda et al. (2011)
3d	Carbamazepine		7			Finland, Helsinki	Vieno et al. (2007b)

		35	25			Canada, Ontario	McKie et al. (2016)
			46	NA		Spain, Llobregat	Huerta-Fontela et al. (2011)
3e	Chlorhexidine			>99	>99	Spain (Northeast)	Boleda et al. (2011)
3f	Clarythromycin			>99	>99	Spain (Northeast)	Boleda et al. (2011)
3g	Diclofenac			99	>99	Spain (Northeast)	Boleda et al. (2011)
		78.3	76.8			Spain (one city in south-eastern)	Azzouz et al. (2013)
			8			Finland, Helsinki	Vieno et al. (2007b)
		36	27			Canada, Ontario	McKie et al. (2016)
3h	Erythromycin			95	99	Spain (Northeast)	Boleda et al. (2011)
3i	Lincomycin			99	>99	Spain (Northeast)	Boleda et al. (2011)
3j	OH-omeprazole			97	99	Spain (Northeast)	Boleda et al. (2011)
3k	Omeprazole			93	95	Spain (Northeast)	Boleda et al. (2011)
3l	Paracetamol	81.3	80.7			Spain (one city in south-eastern)	Azzouz et al. (2013)
3m	Phenylbutazone	79.3	95.5			Spain (one city in south-eastern)	Azzouz et al. (2013)
3n	Sulfadimetoxine			97	91	Spain	Boleda et al.

						(Northeast)	(2011)
3o	Sulfamethazine			99	91	Spain (Northeast)	Boleda et al. (2011)
3p	Sulfamethoxazole			>99	>99	Spain (Northeast)	Boleda et al. (2011)
3q	Trimethoprim			99	>99	Spain (Northeast)	Boleda et al. (2011)
3r	Tylosin			94	99	Spain (Northeast)	Boleda et al. (2011)
D. β-Blockers							
4a	Atenolol		12			Finland, Helsinki	Vieno et al. (2007b)
4b	Salicylic acid			84	85	Spain (Northeast)	Boleda et al. (2011)
4c	Sotalol	<1	5	>96		Finland, Helsinki	Vieno et al. (2007b)
E. Hormones							
5a	Bezafibrate			98	>99	Spain (Northeast)	Boleda et al. (2011)
		17	27	>77		Finland, Helsinki	Vieno et al. (2007b)
5b	Estrone	73	93.8			Spain (one city in south-eastern)	Azzouz et al. (2013)
		33	87			Canada, Ontario	McKie et al. (2016)
5c	17β-Estradiol	73.1	95.2			Spain (one city in south-eastern)	Azzouz et al. (2013)
5d	17α-Ethinylestradiol	75	90			Spain (one city in south-eastern)	Azzouz et al. (2013)

F. Nonsteroidal anti-inflammatory drugs (NSAIDs)							
6a	Clofibric acid			87	97	Spain (Northeast)	Boleda et al. (2011)
		52	39			Canada, Ontario	McKie et al. (2016)
		83.8	NA			Spain (one city in south-eastern)	Azzouz et al. (2013)
6b	Flunixin	84	96			Spain (one city in south-eastern)	Azzouz et al. (2013)
6c	Gemfibrozil			>99	>99	Spain (Northeast)	Boleda et al. (2011)
		29	NA			Canada, Ontario	McKie et al. (2016)
6d	Ibuprofen			98	>99	Spain (Northeast)	Boleda et al. (2011)
		71.3	88.9			Spain (one city in south-eastern)	Azzouz et al. (2013)
		9	12	92		Finland, Helsinki	Vieno et al. (2007b)
6e	Ketoprofen	85.1	80			Spain (one city in south-eastern)	Azzouz et al. (2013)
		38	28			Canada, Ontario	McKie et al. (2016)
			13			Finland, Helsinki	Vieno et al. (2007b)
6f	Naproxen			99	>99	Spain (Northeast)	Boleda et al. (2011)
		52	12			Canada,	McKie et al.

						Ontario	(2016)
		80.4	77.9			Spain (one city in south-eastern)	Azzouz et al. (2013)
			10			Finland, Helsinki	Vieno et al. (2007b)

NA = Not available; ND = Not detectable

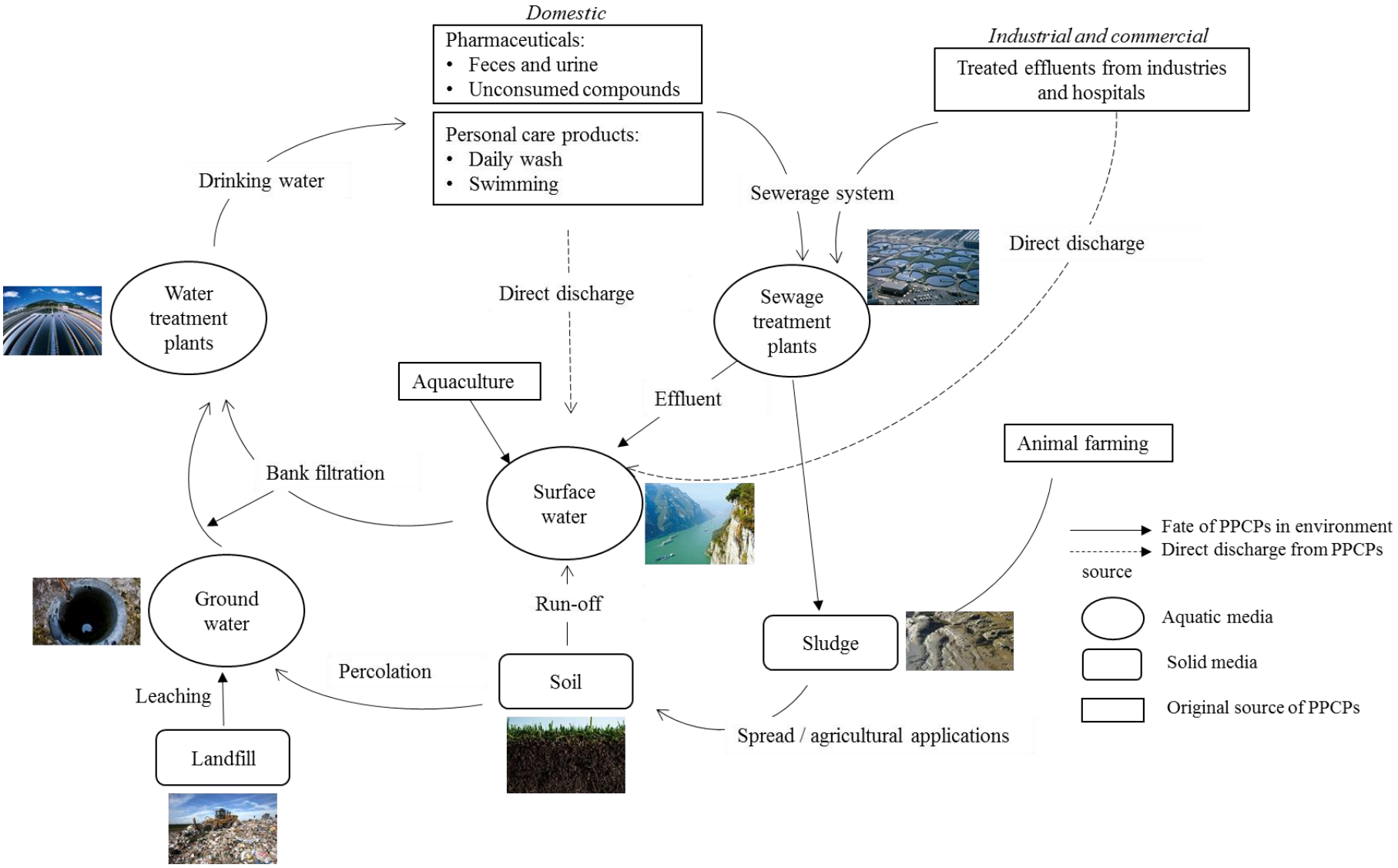

Figure 1. Sources and pathways of PPCPs (modified from Petrović et al., 2003; Mompelat et al., 2009

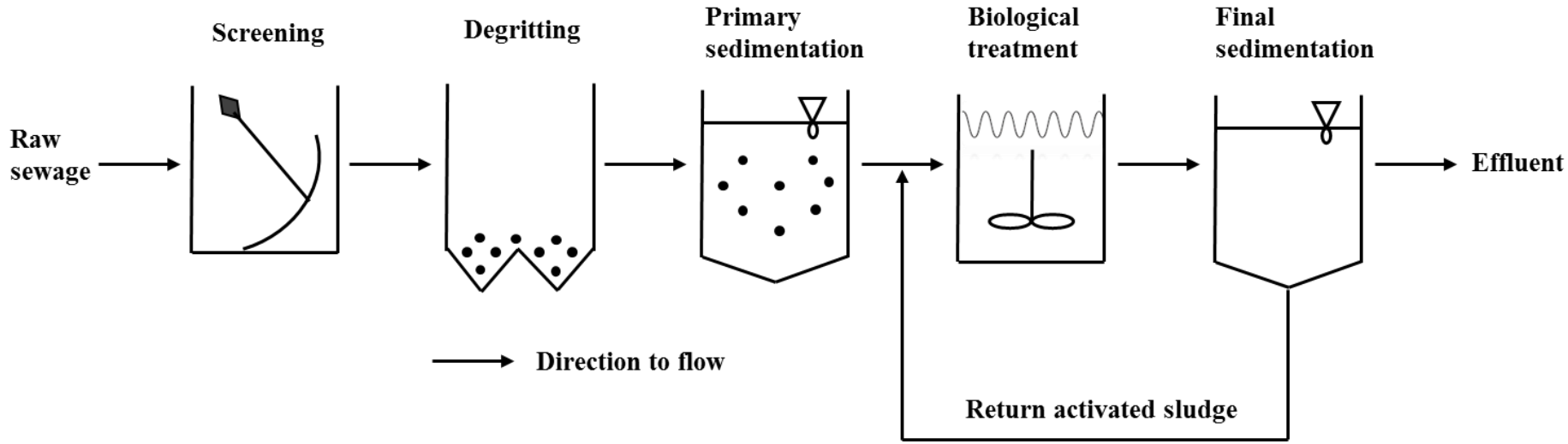

Figure 2a. Flow diagram of conventional STPs (modified from Carballa et al., 2004; Metcalf and Eddy, 2014)

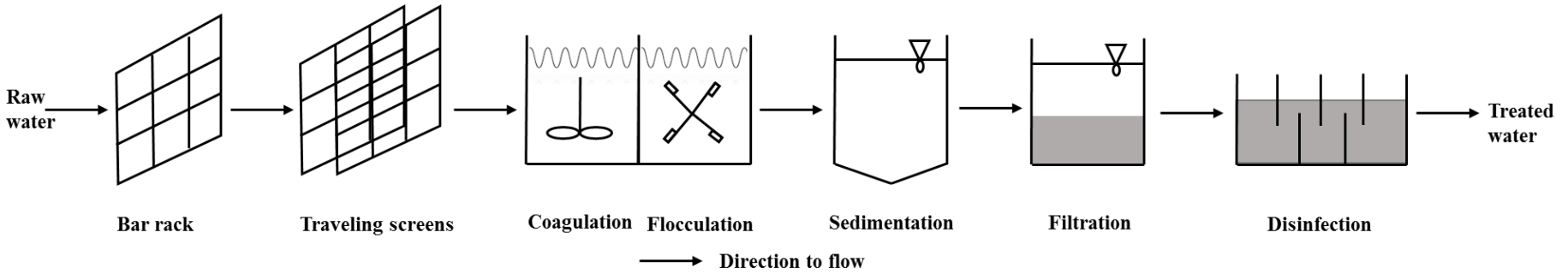

Figure 2b. Flow diagram of conventional WTPs (modified from Metcalf and Eddy, 2014; Stackelberg et al., 2004)