A review on the occurrence of micropollutants in the aquatic environment and their fate and removal during wastewater treatment

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Abstract

Micropollutants are emerging as a new challenge to the scientific community. This review provides a summary of the recent occurrence of micropollutants in the aquatic environment including sewage, surface water, groundwater and drinking water. The discharge of treated effluent from WWTPs is a major pathway for the introduction of micropollutants to surface water. WWTPs act as primary barriers against the spread of micropollutants. WWTP removal efficiency of the selected micropollutants in 14 countries/regions depicts compound-specific variation in removal, ranging from 12.5 to 100%. Advanced treatment processes, such as activated carbon adsorption, advanced oxidation processes, nanofiltration, reverse osmosis, and membrane bioreactors can achieve higher and more consistent micropollutant removal.

However, regardless of what technology is employed, the removal of micropollutants depends on physico-chemical properties of micropollutants and treatment conditions. The evaluation of micropollutant removal from municipal wastewater should cover a series of aspects from sources to end uses. After the release of micropollutants, a better understanding and modeling of their fate in surface water is essential for effectively predicting their impacts on the receiving environment.

Abbreviations

AOP, advanced oxidation process; ASFBBR, aerated submerged fixed bed bioreactor; BAC, biological activated carbon; CAFO, concentrated animal feeding operation; CAS, conventional activate sludge; DBP, di-butyl phthalate; DEET, N,N-Diethyl-meta-toluamide; DEHP, di(2-ethylhexyl) phthalate; DMP, di-methyl phthalate; DOM, dissolved organic matter; EDC, endocrine disrupting compound; GAC, granule activated carbon; HRT, hydraulic retention time; IFAS, fixed film activated sludge; K_d, solid-water distribution coefficient; k_H, Henry's law constant; K_{OW}, octanol–water partition coefficient; MBBR, moving bed biofilm reactor; MBR, membrane bioreactor; MF, microfiltration; NF, nanofiltration; NOM, natural organic matter; NSAID, nonsteroidal anti-inflammatory drug; PAC, powdered activated carbon; PCP, personal care product; pK_a, acid dissociation constant; PNEC, predicted no effect concentration; PPCP, pharmaceutical and personal care product; RO, reverse osmosis; SAnMBR, submerged anaerobic membrane bioreactor; SBBGR, sequencing batch biofilter granular reactor; SRT, sludge retention time; TCEP, tris(2-chloroethyl) phosphate; TCPP, tris(1-chloro-2-propyl) phosphate; UF, ultrafiltration; WWTP, wastewater treatment plant

Keywords: Micropollutants; Occurrence; Fate; Removal; WWTP; Advanced treatment

1. Introduction

Over the last few decades, the occurrence of micropollutants in the aquatic environment has become a worldwide issue of increasing environmental concern. Micropollutants, also termed as emerging contaminants, consist of a vast and expanding array of anthropogenic as well as natural substances. These include pharmaceuticals, personal care products, steroid hormones, industrial chemicals, pesticides and many other emerging compounds. Micropollutants are commonly present in waters at trace concentrations, ranging from a few ng/L to several μg/L. The 'low concentration' and diversity of micropollutants not only complicate the associated detection and analysis procedures but also create challenges for water and wastewater treatment processes.

Current wastewater treatment plants (WWTPs) are not specifically designed to eliminate micropollutants. Thus, many of these micropollutants are able to pass through wastewater treatment processes by virtue of their persistency or/and the continuous introduction. In addition, precautions and monitoring actions for micropollutants have not been well established in most WWTPs (Bolong et al., 2009). Consequently, many of these compounds may end up in the aquatic environment, becoming threats to wildlife and spelling trouble for drinking water industry. The occurrence of micropollutants in the aquatic environment have been frequently associated with a number of negative effects, including short-term and long-term toxicity, endocrine disrupting effects and antibiotic resistance of microorganisms (Fent et al., 2006 and Pruden et al., 2006). To date, discharge guidelines and standards do not exist for most micropollutants. Some countries or regions have adopted regulations for a small number of micropollutants. For example, environmental quality standards for a minority of

micropollutants (e.g. nonylphenol, bisphenol A, DEHP and diuron) have been stipulated in Directive 2008/105/EC (European Parliament and The Council, 2008). Nonylphenol and nonylphenol ethoxylates have also been recognized as toxic substances by the Canadian government (Canadian Environmental Protection Act, 1999). Other micropollutants, such as pharmaceutical and personal care products (PPCPs) and steroid hormones, are not included in the list of regulated substances yet. To set regulatory limits for micropollutants, further research on biological responses to these compounds (both acute and chronic effects) is of particular importance. Furthermore, scientific community and regulatory agencies should gain insight into not only the impact of individual micropollutants, but also their synergistic, additive, and antagonistic effects.

Several review papers have been published with regard to the occurrence of micropollutants in different water bodies such as wastewater (Deblonde et al., 2011) and groundwater (Lapworth et al., 2012), as well as treatment methods for micropollutant removal (Bolong et al., 2009). In addition, Verlicchi et al. (2012) reviewed the pharmaceutical removal efficiency in conventional activated sludge systems and in MBR fed by municipal wastewater, while Liu et al. (2009) focused on the physical, chemical and biological removal of endocrine disrupting compounds (EDCs). However, no attempt has been made to provide a comprehensive summary of the occurrence of miscellaneous micropollutants in aquatic systems as well as the removal of micropollutants in conventional and advanced treatment processes. In this review, we systematically summarized the recent occurrence of various micropollutants in the aquatic environment and delineated the behavior and removal of micropollutants during conventional as well as advanced wastewater treatment processes.

2. Occurrence of micropollutants in the aquatic environment

Sources of micropollutants in the environment are diverse and many of these originate from mass-produced materials and commodities. Table 1 summarizes the sources of the major categories of micropollutants in the aquatic environment.

Table 1
Sources of micropollutants in the aquatic environment.

Category	Important subclasses	Major sources	
		Distinct	Nonexclusive
Pharmaceuticals	NSAIDs, lipid regulator, anticonvulsants, antibiotics, β-blockers, and stimulants	Domestic wastewater (from excretion) Hospital effluents Run-off from CAFOs[a] and aquaculture	Sources that are not exclusive to individual categories include: Industrial wastewater (from product manufacturing discharges) Landfill leachate (from improper disposal of used, defective or expired items)
Personal care products	Fragrances, disinfectants, UV filters, and insect repellents	Domestic wastewater (from bathing, shaving, spraying, swimming and etc.)	
Steroid hormones	Estrogens	Domestic wastewater (from excretion) Run-off from CAFOs and aquaculture	
Surfactants	Non-ionic surfactants	Domestic wastewater (from bathing, laundry, dishwashing and etc.) Industrial wastewater (from industrial cleaning discharges)	
Industrial chemicals	Plasticizers, fire retardants	Domestic wastewater (by leaching out of the material)	
Pesticides	Insecticides, insecticides, herbicides and fungicides	Domestic wastewater (from improper cleaning, run-off from gardens, lawns and roadways and etc.) Agricultural runoff	

[a] CAFOs: concentrated animal feeding operations.

The recent occurrence (2008 to date) of the micropollutants in the aquatic environment has been reviewed in terms of their aqueous concentrations in different types of waters, including wastewater, surface water, groundwater and drinking water. Of all aqueous media, WWTP influent and effluent are comprehensively reviewed. The collected data consist of the studies performed in a number of countries/regions, including Austria, China, EU-wide, France, Germany, Greece, Italy, Korea, Spain, Sweden, Switzerland, Western Balkan Region, UK and US. In general, the investigated micropollutants can be divided into six categories namely pharmaceuticals, personal care products, steroid hormones, surfactants, industrial chemicals and pesticides.

2.1. Occurrence of micropollutants in WWTPs

Occurrence data of micropollutants in WWTP influent and effluent from recent studies (2008–present) are summarized in Table 2. As can be noted from the table, the reported concentrations of micropollutants in WWTP influent and effluent reveal significant spatial and temporal variations, which are essentially due to a number of factors, including the rate of production, specific sales and practices, metabolism (excretion rate), water consumption per person and per day, the size of WWTPs, environmental persistence and elimination efficacy of wastewater treatment processes (Jelic et al., 2012 and Petrovic et al., 2009).

Table 2.
The concentrations and removals of the selected micropollutants in conventional WWPTs in different countries.

Categories	Selected compounds	Sampling sites	Influent (μg/L)	Effluent (μg/L)	Removal (%)[a]	References[b]
Pharmaceutical						
Analgesic and anti-inflammatory	Acetaminophen	Korea, Spain, WB[c]	1.57–56.9	ND[d]–0.03	98.7–100	2, 5, 8, 19, 25
	Diclofenac	EU-wide, Greece, Korea, Sweden, Switzerland, UK, WB	< 0.001–94.2	< 0.001–0.69	< 0–81.4	2, 8, 11, 14, 19, 21, 22, 25, 27, 28
	Ibuprofen	China, EU-wide, Greece, Korea, Sweden, UK, US, WB	< 0.004–603	ND–55	72–100	2, 8, 11, 14, 19, 20, 22, 25, 26, 28
	Ketoprofen	China, EU-wide, Korea, Spain, UK, WB	< 0.004–8.56	< 0.003–3.92	10.8–100	2, 8, 11, 14, 20, 25, 27
	Mefenamic acid	EU-wide, Korea, Spain, UK	< 0.017–1.27	< 0.005–0.39	< 0–70.2	2, 8, 11, 19
	Naproxen	Greece, Korea, Spain, Sweden, UK, WB	< 0.002–52.9	< 0.002–5.09	43.3–98.6	2, 8, 11, 19, 20, 22, 25, 28
	Salicylic acid	Greece, Spain, UK	0.58–63.7	ND–0.50	89.6–100	8, 11, 22
Anticonvulsant	Carbamazepine	China, EU-wide, Greece, Korea, Spain, UK, WB	< 0.04–3.78	< 0.005–4.60	< 0–62.3	2, 5, 11, 14, 19, 20, 25, 27
Lipid regulator	Bezafibrate	EU-wide, Spain, Korea, UK, WB	0.05–1.39	0.03–0.67	9.10–70.5	2, 8, 11, 14, 19, 25
	Clofibric acid	China, EU-wide, Greece, Korea, Spain, Sweden, UK, WB	0–0.74	ND–0.33	< 0–93.6	2, 11, 14, 19, 22, 25, 28
	Gemfibrozil	EU-wide, Greece, Korea, Spain, WB	0.10–17.1	< 0.0025–5.24	< 0–92.3	2, 8, 14, 19, 22, 25
	Erythromycin	China, Spain, UK, WB	0.14–10.0	0.02–2.84	< 0–82.5	8, 11, 19, 25, 27
Antibiotic	Sulfamethoxazole	EU-wide, France, Korea, Spain, Sweden, Switzerland, UK, WB	< 0.003–0.98	< 0.003–1.15	4–88.9	2, 5, 8, 11, 14, 15, 19, 21, 25
	Trimethoprim	China, EU-wide, Korea, Spain, UK	0.06–6.80	< 0.01–3.05	< 0–81.6	2, 5, 8, 11, 14, 19, 25, 27
β-Blocker	Atenolol	Korea, Spain, Switzerland, UK,WB	0.1–33.1	0.13–7.60	< 0–85.1	1, 2, 11, 19, 25

Categories	Selected compounds	Sampling sites	Influent (μg/L)	Effluent (μg/L)	Removal (%)[a]	References[b]
	Metoprolol	China, Korea, Spain, Switzerland, UK	0.002–1.52	0.003–0.25	3–56.4	1, 2, 8, 11, 19
Nervous stimulant	Caffeine	China, EU-wide, Greek, Korea, Spain, UK	0.22–209	ND–43.50	49.9–99.6	2, 5, 14, 19, 20, 22, 26, 27
PCP						
Musk fragrance	Galaxolide	Spain, WB	0.03–25	< 0.06–2.77	87.8	19, 25
	Tonalide	Spain, WB	< 0.05–1.93	< 0.05–0.32	84.7	19, 25
Disinfectant	Triclosan	Spain, UK, US, Greece, Korea, France, EU-wide	0.03–23.9	0.01–6.88	71.3–99.2	2, 13, 14, 15, 17, 19, 22, 24, 26
Insect repellant	DEET	China, EU-wide	2.56–3.19	0.61–15.8	65.6–79.5	14, 25
UV-filter	Benzophenone-3	Korea, Spain	< 0.079–0.90	< 0.079–0.23	63.8–98.2	2, 19
Steroid hormone						
	Estrone	China, France, Germany, Italy, Korea, Sweden, US	0.01–0.17	< 0.001–0.08	74.8–90.6	2, 9, 16, 28
	Estradiol	China, France, Germany, Italy, Korea, Sweden, US	0.002–0.05	< 0.001–0.007	92.6–100	2, 9, 16, 28
	17α-Ethynylestradiol	China, France, Germany, Italy, Sweden, US	0.001–0.003	< 0.001–0.002	43.8–100	9, 16, 28
	Estriol	China, Korea	0.125–0.80	ND	100	2, 16
Surfactants						
	Nonylphenol	China, France, Germany, Greece, Italy, Spain, US, WB	< 0.03–101.6	< 0.03–7.8	21.7–99	4, 9, 15, 16, 17, 24, 25
	Octylphenol	China, France, Germany, Italy, Spain, UK, US	< 0.2–8.7	0.004–1.3	< 0–96.7	4, 9, 11, 15, 16, 17, 25
Industrial chemicals						
	Bisphenol A	China, France, Greece, US, WB	< 0.013–2.14	< 0.03–1.10	62.5–99.6	11, 15, 16, 17, 24, 25, 26
Plasticizers	DBP	Austria, China	ND–11.8	ND–4.13	73.6–75.5	6, 7
	DEHP	Austria, China, US	0.003–70.0	0.0001–54.0	25–97	6, 7, 26
	DMP	Austria, China	ND–6.49	ND–1.52	84.8–93.5	6, 7

Categories	Selected compounds	Sampling sites	Influent (μg/L)	Effluent (μg/L)	Removal (%)[a]	References[b]
Fire retardant	TCEP	EU-wide, Germany	0.06–0.50	0.06–2.40	< 0	14, 18
	TCPP	EU-wide, Germany	0.18–4	0.10–21	< 0	14, 18
Pesticide						
Herbicide	Atrazine	EU-wide, France, Spain, Switzerland, WB	0.02–28	0.004–0.73	< 0–25	3, 12, 14, 15, 21, 25
	Diuron	EU-wide, France, Spain, Switzerland	0.03–1.96	0.002–2.53	26.7–71.9	3, 12, 14, 15, 19, 21
Insectcide	Diazinon	EU-wide, Spain	< 0.684	0.0007–4.16	< 0	3, 12, 14
Fungicide	Clotrimazole	EU-wide, Greece	0.012–0.08	ND–0.005	84.5–93.6	10, 14, 23
	Tebuconazole	Greece, Spain	ND–1.89	0.0005–0.69	< 0–58.7	3, 10, 23

[a] When the removal efficiency was not presented in a study, it was calculated using the following equation, removal efficiency (%) = $(C_{inf} - C_{eff}) / C_{inf} \times 100$. ($C_{inf}$is the influent concentration of a compound and C_{eff} is the effluent concentration of a compound).

[b] 1. Alder et al. (2010); 2. Behera et al. (2011); 3. Campo et al. (2013); 4. Céspedes et al. (2008); 5.K. Choi et al. (2008); 6. Clara et al (2010); 7. Gao et al. (2014); 8. Gracia-Lor et al. (2012); 9. Janex-Habibi et al. (2009); 10. Kahle et al. (2008); 11. Kasprzyk-Hordern et al. (2009); 12. Köck-Schulmeyer et al. (2013); 13. Kumar et al. (2010); 14. Loos et al. (2013); 15. Martin et al. (2010); 16. Nie et al. (2012); 17. Pothitou and Voutsa (2008); 18. Reemtsma et al. (2008); 19. Santos et al. (2009); 20. Singer et al. (2010); 21. Stamatis and Konstantinou (2013); 22. Stamatis et al. (2010); 23. Stasinakis et al. (2008); 24.Rosal et al. (2010); 25. Terzić et al. (2008); 26. Yu and Chu (2009); 27. Zhou et al. (2010); and 28. Zorita et al. (2009).

[c] WB: Western Balkan Region (including Bosnia and Herzegovina, Croatia and Serbia).

[d] ND: not detected.

The local production and usage/consumption of products containing micropollutants determine the amount of micropoullutants reaching WWTPs. Studies suggested that PPCP concentrations in wastewater correlated well with their production amounts and usage/consumption patterns. K. Choi et al. (2008) reported that the occurrence concentrations of acetaminophen, carbamazepine, cimetidine, diltiazem, sulfamethoxazole and trimethoprim followed the same order (from highest to lowest) of their annual production amount in Korea. High concentrations (> 10 μg/L) of acetaminophen, tramadol, codeine, gabapentin and atenolol were detected at highest levels in raw wastewater in Wales, UK and this could be explained by the high quantities of these pharmaceuticals dispensed (Kasprzyk-Hordern et al.,

2009). As orally ingested products containing potential contaminants (e.g. pharmaceuticals) are metabolized in human body and are subsequently excreted via urine and feces, excretion rate plays a role in determining the introduction of pharmaceuticals into raw wastewater. Table 3 presents the excretion rates for some commonly encountered pharmaceuticals. It can be noted that pharmaceutical compounds with low excretion rates (e.g., ibuprofen, carbamazepine, sulfamethoxazole, diclofenac and primidione) are not necessarily present at low levels in the raw wastewater. This is possibly because the low excretion rates are offset by the massive use of these compounds. In addition, local common diseases can induce a higher consumption of specific pharmaceuticals in certain periods. Research showed climatic conditions could cause fluctuating micropollutant input (Kolpin et al., 2004). The use of pesticides can be seasonal due to the prevalence of pests in different climatic conditions. Another important factor is rainfall, as it affects the flow pattern of wastewater influent when a combined sewer system is employed. Kasprzyk-Hordern et al. (2009) found that the concentrations of most PPCPs in the raw wastewater were doubled when the flow was halved during dry weather conditions, suggesting that rainwater could dilute the concentrations of the compounds within the sewage. Other weather conditions, such as temperature and level of sunlight also can affect the discharge of micropollutants from WWTPs.

Table 3
Human excretion rates of some common pharmaceutical compounds in the aquatic environment.
Adapted from Alder, Hirsch et al. (1999), Huschek et al. (2004), Jjemba (2006) and Ternes (1998); and the range was selected according to Jjemba (2006).

Excretion rate	Pharmaceutical
Low (≤ 5%)	Aspirin (acetylsalicylic acid), carbamazepine, gemfibrozil, ibuprofen
Moderately low (6–39%)	Diclofenac, metroprolol, primidone, sulfamethoxazole
Relatively high (40–69%)	Bezafibrate, norfloxacin, trimethoprim
High (≥ 70%)	Amoxicillin, ciprofloxacin, tetracycline

Fig. 1 depicts the average occurrence levels reported for the selected compounds in WWTP influents and effluents. As can be seen in Fig. 1, most micropollutants occurred in WWTP influent in the concentration range between 0.1 and 10 μg/L, while some pharmaceutical compounds (acetaminophen, caffeine, ibuprofen, naproxen and salicylic acid), one biocide (triclosan), one surfactant (nonylphenol) and one industrial chemical (DEHP) exhibit relatively high occurrence concentrations. Generally, the compounds with the highest concentrations (mean values > 10 μg/L) in WWTP influent were ibuprofen, atenolol, caffeine and nonylphenol. For instance, ibuprofen was the most abundant compound detected in the influent of four WWTPs in Spain, with the concentration levels ranging from 3.73 to 603 μg/L (Santos et al., 2009). The particularly high levels could be explained by the high consumption and easy accessibility (over the counter drugs) of the compound. Caffeine was detected at the highest levels approaching 50 μg/L on average in the raw sewage of three WWTPs in China (Zhou et al., 2010). The abundant presence of caffeine is likely associated with the high consumption of coffee, tea and soft drinks as well as the disposal of these items. Steroid hormones and pesticides generally show lower detected concentrations (mostly less than 1 μg/L) as compared with compounds from other groups. The concentrations of most

micropollutants in effluent ranged from 0.001 to 1 μg/L, which were one to two orders of magnitude lower than those in influent. Some abundant compounds in influent were discharged at relatively high concentrations. For instance, atenolol, caffeine, DEHP, ibuprofen, naproxen, nonylphenol and triclosan were detected in the concentrations higher than 1 μg/L in treated effluent. In contrast, steroid hormones were found in wastewater at much lower levels (< 100 ng/L). However, their occurrence even at low concentrations is a concern because of their high estrogenic effect.

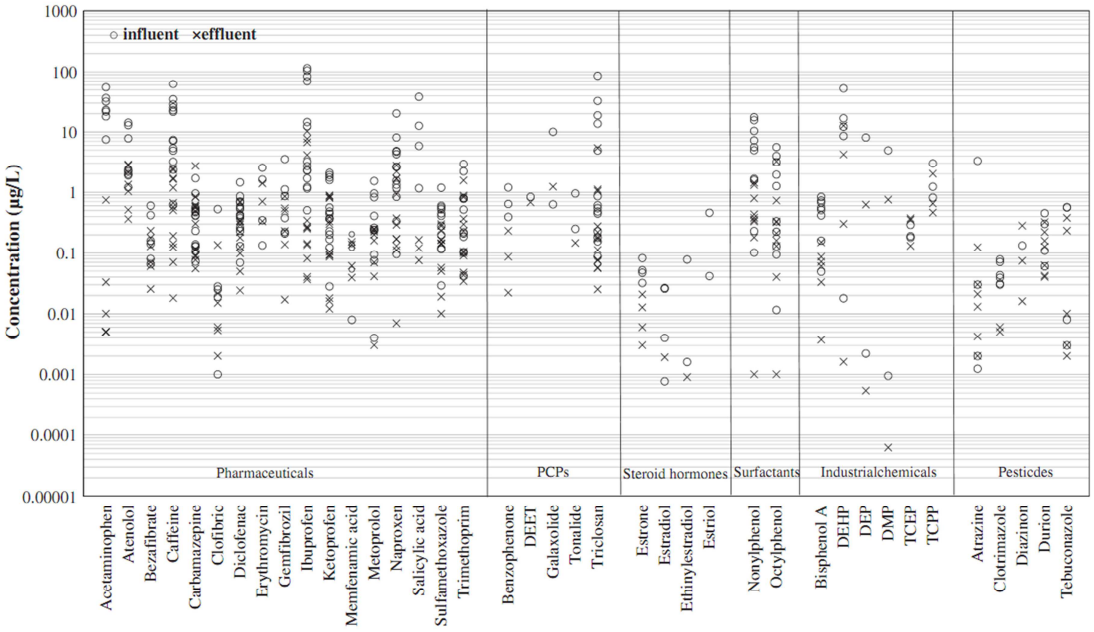

Fig. 1. Average concentrations (on logarithmic Y axis) reported for the selected micropollutants in WWPT influents and effluents (data from Table 2).

2.2. Occurrence of micropollutants in surface water

The release of WWTP effluent into surface water has been considered as a main cause of the presence of micropollutants in surface water in comparison to other sources (Kasprzyk-Hordern et al., 2009). Following treatment processes in WWTPs, micropollutants are subjected to varying degrees of natural attenuation (e.g., dilution in surface water, sorption onto suspended solids and sediments, direct and indirect photolysis and aerobic

biodegradation) (Pal et al., 2010). Due to river water dilution, pharmaceutical compounds may occur at levels at least one order of magnitude lower than effluent levels (Gros et al., 2007). Gómez et al. (2012) found that the natural attenuation of PCPs is more likely to result from river water dilution, or sorption to solids, than from degradation. Furthermore, river water dilution can be affected by rainfall. Consistent increase in micropollutant occurrence levels during dry weather conditions and marked reduction during wet weather conditions have been reported. Wang et al. (2011) indicated that pharmaceuticals in summer water samples showed lower occurrence levels than those in winter. This could be due to 1) promoted biodegradation of pharmaceuticals in warmer temperature, and 2) elevated dilution during wetter summer. However, rainfall did not always reduce the concentration levels of micropollutants released. In some cases, rainfall was identified as a contributor to the emission of micropollutants to surface water. Some studies revealed that the chemicals (e.g., bisphenol A and biocides) used in building material (e.g. pavement materials, facades and roof paintings) were able to leach during precipitation and accumulate to remarkable levels in roof runoff and subsequently ended up in surface water (Jungnickel et al., 2008, Sakamoto et al., 2007, Schoknecht et al., 2009 and Singer et al., 2010). In addition, rainfall events could intensify combined sewer overflows, resulting in a higher level of contaminant discharge. Regarding pesticides, the contamination of surface water by these compounds depends on crop type, soil properties, characteristics of the water bodies (depth and flow rate), features of the land close to the water bodies (soil use, slope, and distance from water bodies) and climatic conditions (temperature, rainfall, moisture and wind) (Bermúdez-Couso et al., 2013).

According to Table 4 showing common micropollutants in surface water from different countries, nonsteroidal anti-inflammatory drugs (NSAIDs), carbamazepine, sulfamethoxazole and triclosan were the most frequently reported compounds in surface water. The high

concentrations of micropollutants were found in Costa Rica, which mainly resulted from the discharge of hospital effluents and other highly contaminated waters (Spongberg et al., 2011). Notably, ibuprofen, ketoprofen, gemfibrozil and caffeine were detected at alarmingly high levels, with maximum concentrations of 36.8, 9.8, 17.0 and 1121.4 μg/L, respectively. Caffeine was also detected at relatively high concentrations in the US (224.8 ng/L) and Taiwan (1813 ng/L). Unlike Costa Rica, the reported caffeine concentrations in the US and Taiwan were far below the predicted no effect concentrations (PNECs). In general, the pollution of emerging contaminants in the natural water bodies of the densely populated regions are more severe because of the massive usage of these chemicals by the large population. For example, the concentrations of nonylphenol, bisphenol A and triclosan in a surface water in Guangzhou (one of the largest cities in China) were at rather high levels. Nonylphenol was also found at relatively high concentrations in a Greek river, with a maximum of 2704 ng/L. The maximum nonylphenol concentrations in China and Greece were well above the reported PNEC for nonylphenol. In addition to above mentioned factors, population aging has also been linked to the high occurrence levels of pharmaceuticals (Al-Rifai et al., 2007).

Table 4
Occurrence of some common micropollutants in surface waters in different countries.

Compound	Concentration (ng/L)											
	Canada [a] [nd]	China[c]	Costa Rica [and]	France[e]	Germany [and]	Greece[h]	Korea[i]	Spain [and]	Taiwan[k]	UK[l]	US[m]	PNEC[n]
Ibuprofen	0.98 (79)	ND–1417	5 (36,788)	ND–8	–	1–67	< 15–414	–	5–280	0.3–100	ND–77	5000
Naproxen	1 (87)	ND–328	–	ND–6.4	–	3–322	–	–	–	0.3–149	–	37,000
Ketoprofen	–	–	7 (9808)	ND–22.0	–	0.4–39.5	–	–	10–190	0.5–14	–	15.6×10^6
Diclofenac	–	–	14 (266)	ND–35.0	–	0.8–1043	–	–	–	0.5–261	–	10,000
Mefenamic acid	–	–	–	–	–	–	< 30–326	–	–	0.3–169	–	–
Carbamazepine	3 (749)	–	1 (82)	ND–31.6	102–1194	–	< 4–595	–	–	0.5–684	ND–9.6	25,000
Gemfibrozil	–	–	41 (17,036)	–	–	–	–	–	1.9–3.5	–	–	100,000
Atenolol	–	–	–	ND–34.0	–	–	< 100–690	–	–	1–560	–	10×10^6
Sulfamethoxazole	0.2 (284)	–	11 (56)	ND–5.1	–	–	–	–	0.3–60	0.5–4	ND–38	20,000
Trimethoprim	–	–	–	–	–	–	–	–	1–2.1	7–122	ND–9.1	1000
Triclosan	0.4 (25)	35–1023	11 (263)	–	124–220	3–39	ND[c]	–	–	5–95	ND–9.8	–
Galaxolide	–	–	–	–	35–1814	–	–	–	–	–	–	–
Tonalide	–	–	–	–	5–273	–	–	–	–	–	–	–
Estrone	–	ND–65	–	–	–	–	3.6–69.1	–	–	–	–	18
Estradiol	–	ND–2	–	–	–	–	1.1–10.1	–	–	–	–	–
Ethinylestradiol	–	ND–1	–	–	–	–	ND–	–	–	–	–	0.02

Compound	Concentration (ng/L)											
	Canada[a] and[nd]	China[c]	Costa Rica[and]	France[e]	Germany[and]	Greece[h]	Korea[i]	Spain[and]	Taiwan[k]	UK[l]	US[m]	PNEC[n]
Estriol	–	ND–1	–	–	–	–	1.9 –	–	–	–	–	149
Caffeine	–	–	24 (1,121,446)	–	–	–	–	–	1–1813	–	ND–225	10×10^5
Nonylphenol	–	36–33,231	–	–	–	558–2704	115–336	–	–	–	–	330
Bisphenol A	2.1 (87)	6–881	–	–	192–215	55–162	7.5–334	–	–	6–68	–	1000
TCEP	–	–	–	–	< 3–184	–	–	–	–	–	–	–
TCPP	–	–	–	–	< 4–379	–	–	–	–	–	–	–
Atrazine	–	–	–	–	–	–	–	11 (39)	–	–	–	2000
Diazinon	–	–	–	–	–	–	–	10(216)	–	–	–	–
Diuron	–	–	–	–	–	–	–	72(408)	–	–	–	1800

[a] Median concentration with maximum concentration in the brackets.

[b] Kleywegt et al. (2011).

[c] Peng et al. (2008).

[d] Spongberg et al. (2011).

[e] Vulliet et al. (2011).

[f] Regnery and Püttmann (2010).

[g] Reinstorf et al. (2008).

[h] Stasinakis et al. (2012).

[i] Kim et al. (2009c).

[j] Köck et al. (2010).

[k] Lin et al. (2011).

[l] Kasprzyk-Hordern et al. (2009).

[m] Wang et al. (2011).

[n] Data were derived from Fromme et al., 2002, Köck et al., 2010, Lin et al., 2008 and Loos et al., 2007.

2.3. Occurrence of micropollutants in groundwater

In comparison to surface water, ground water was found to be less contaminated with micropollutants (Loos et al., 2010 and Vulliet and Cren-Olivé, 2011). Hence, the presence of micropollutants in groundwater has been put far less emphasis on. Better characterization of micropollutants in groundwater has been only done regionally (mainly in some parts of Europe and North America). Micropollutant contamination of groundwater mainly results from landfill leachate, groundwater–surface water interaction, infiltration of contaminated water from agricultural land or seepage of septic tanks and sewer systems. Concentrations of micropollutants in landfill leachate and septic tank leakage generally range from 10 to 10^4 ng/L and 10 to 10^3 ng/L, respectively (Lapworth et al., 2012). Soil is the major pathway for groundwater pollution by some micropollutants (e.g. pesticides) (González-Rodríguez et al., 2011). Micropollutants can also be introduced in groundwater via bank filtration or artificial recharge using reclaimed water (Stepien et al., 2013). Generally, the processes governing subsurface flow and transport (such as dilution, adsorption to aquifer material, degradation and travel time) can decrease micropollutants' concentrations from the sources (e.g., landfill leachate and septic tank leakage) to groundwater (Teijon et al., 2010). The physicochemical properties of micropollutants are therefore important for the transfer of the compounds to groundwater. For example, octanol–water partition coefficient (K_{OW}) indicates contaminant mobility in the subsurface, where the compounds (e.g., trimethoprim and TCEP) with $K_{OW} < 1.5$ tend to stay in the dissolved phase (more mobility) and are more likely to occur in groundwater (Dougherty et al., 2010 and Karnjanapiboonwong et al., 2011). In a study conducted in the US, Fram and Belitz (2011) found good correlation of pharmaceutical levels in groundwater and presence of modern water (water recharged since 1953), occurrence of other synthetic contaminants (urban-use herbicides and insecticides and volatile organic compounds) and land application.

For selected countries (Table 5), most of the compounds were detected at less than 100 ng/L in groundwater. NASIDs, carbamazepine, sulfamethoxazole, caffeine and triclosan were of particular research interest. These compounds were also the most commonly detected ones in surface water and wastewater, evidencing a correlation of the presence of micropollutants in different aquatic systems. By comparing the occurrence concentrations of micropollutants with PNEC, most of the compounds were at levels without potential environmental significance. However, it is notable that these PNEC values were determined based on individual compounds rather than mixtures of contaminants such as encountered in the aquatic environments. Considerably high concentrations (2 or 3 orders of magnitude higher than PNEC) of steroid hormones were found in groundwater at a US land application site (Karnjanapiboonwong et al., 2011). The problem probably resulted from the application of wastewater effluent to a portion of the soil. Although the authors did not point out the adverse effects of the high-level steroid hormones, their occurrence would be of potential concern if the groundwater was utilized for direct or indirect potable water reuse.

Table 5
Occurrence of some common micropollutants in groundwater in different countries.

Compound	Concentrations (ng/L)					
	Europe[a,b]	France[c,d]	Germany[e,f,g,h]	Spain[a,I,j,k]	US[a,c,l,m,n]	PNEC[o]
Ibuprofen	3 (395)	0	–	185 (185)	0, 3110	5000
Naproxen	–	1.2	–	204 (145–263)	–	37,000
Ketoprofen	26 (2886)	2.8	–	–	–	15.6×10^6
Diclofenac	0 (24)	9.7	3050	256 (35–477)	–	10,000
Carbamazepine	12 (390)	10.4	< 50, 2325	–	40 (420)	25,000
Gemfibrozil	–	–	–	165.3 (12–574)	–	100,000
Bezafibrate	–	0	112	–	–	–
Atenolol	–	5.5		60.8 (18–	–	10×10^6

Compound	Concentrations (ng/L)					
	Europe[a,b]	France[c,d]	Germany[e,f,g,h]	Spain[a,I,j,k]	US[a,c,l,m,n]	PNEC[o]
				106)		
Sulfamethoxazole	2 (38)	3.0	–	47.57 (2–117)	1110, 160 (170)	20,000
Trimethoprim	–	1.4	–	–	–	1000
Caffeine	13 (189)	–	–	63.56 (4–505)	130, 170 (290)	10×10^5
Triclosan	0 (9)	–	–	39.8 (2–118)	53	–
Nonylphenol	83 (3850)	–	–	–	–	330
Bisphenol A	79 (2299)	–	–	–	2550	1000
Estrone	0 (4)	0.7	–	–	79	18
Estradiol	–	0.4	–	–	147	–
Ethinylestradiol	–	1.2	–	–	230	0.02
Estriol	–	–	–	–	1661	149
TCEP	–	–	4–51	–	–	–
TCPP	–	–	14–355	–	–	–
Atrazine	–	–	–	36 (756)	–	2000
Diazinon	–	–	–	5.3 (30.8)	–	–
Diuron	–	–	–	8.8 (178)	–	1800

[a] Average concentration with maximum concentration in the brackets.

[b] Loos et al. (2010).

[c] Average concentration.

[d] Vulliet and Cren-Olivé (2011).

[e] Maximum concentration.

[f] Maeng et al. (2010).

[g] Müller et al. (2012).

[h] Stepien et al. (2013).

[i] Average concentration with minimum and maximum concentrations in the brackets.

[j] Postigo et al. (2010).

[k] Teijon et al. (2010).

[l] Barnes et al. (2008).

[m] Fram and Belitz (2011).

[n] Karnjanapiboonwong et al. (2011).

[o] Data were derived from Fromme et al. (2002), Köck et al. (2010), Lin et al. (2008), and Loos et al. (2007).

2.4. Occurrence of micropollutants in drinking water

A limited amount of publications are available with regard to the occurrence of micropollutants in drinking water (Vulliet et al., 2011). Some recent studies showed that most micropollutants in finished waters from drinking water treatment were below limit of quantitation or limit of detection (Benotti et al., 2008, Huerta-Fontela et al., 2011, Kleywegt et al., 2011 and Wang et al., 2011). Therefore, only the data of the most abundant compounds are presented in Fig. 2. To date, there has been a lack of guidelines for risk assessment for the presence of most micropollutants in drinking water. PNEC values were plotted to superficially describe the potential of negative effects (Fig. 2). The occurrence levels of micropollutants in drinking water are dependent on water sources and seasons, with winter water samples showing higher concentrations in comparing to summer water samples. Furthermore, drinking water treatment plays a significant role in eliminating micropollutants from drinking water and has therefore been comprehensively examined (Stackelberg et al., 2004 and Westerhoff et al., 2005).

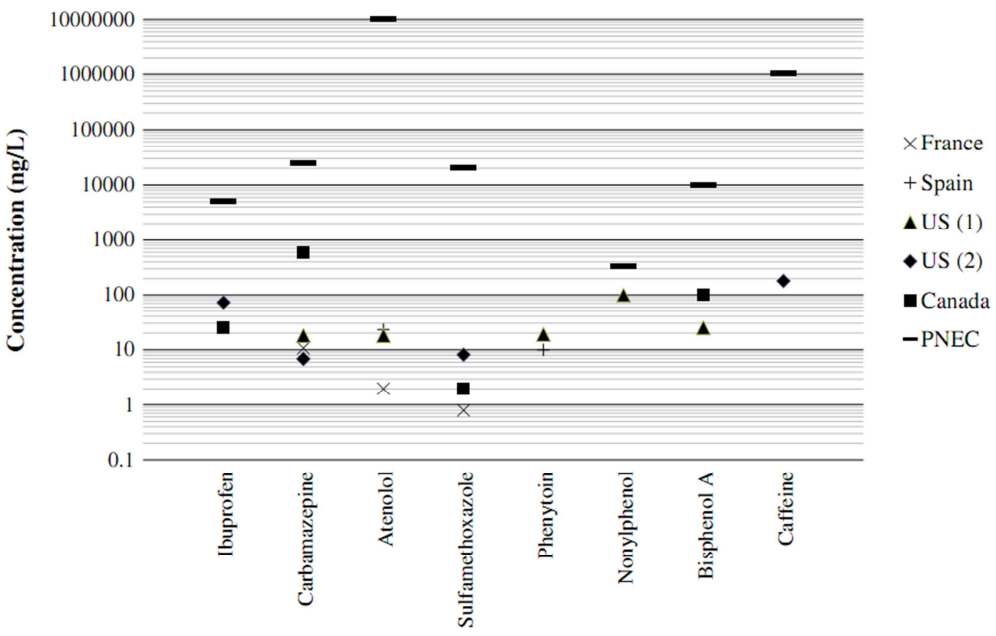

Fig. 2. Maximum occurrence concentrations of some most abundant micropollutants in drinking water (Benotti et al., 2008, Huerta-Fontela et al., 2011, Kleywegt et al., 2011, Vulliet et al., 2011 and Wang et al., 2011).

As can be seen in Fig. 2, the maximum occurrence concentrations of most micropollutants were reported to be below 100 ng/L, with the exception of carbamazepine and caffeine. Notably, carbamazepine was observed at a concentration exceeding 600 ng/L (a concentration more than 10 times higher than those of most other compounds) in the study conducted by Kleywegt et al. (2011). The high levels of carbamazepine could be explained by its high persistency. Even so, the occurrence level of carbamazepine was far below the PNEC (25,000 ng/L). It is also noteworthy that nonylphenol showed a maximum concentration (100 ng/L) most close to PNEC (330 ng/L, less than 1 order of magnitude). Other compounds were all at safe levels, since the PNEC values were 2 to 5 orders of magnitude higher than the their maximum concentrations. Overall, based on the studies reviewed here, these countries were all able to rule out the adverse impacts of selected micropollutants on drinking water. Nevertheless, since other compounds as well as transformation by-products, which can also pose adverse effects, were not monitored in these studies, the safety of the produced drinking water still needs to be under scrutiny.

3. The removal and fate of micropollutants in WWTPs

Municipal WWTPs are designed to control a wide range of substances, such as particulates, carbonaceous substances, nutrients and pathogens. While these substances can be efficiently and consistently eliminated, the removal of micropollutants is often insufficient. Hence, the evaluation of the fate and removal of micropollutants during wastewater treatment is imperative for the optimization of treatment processes, in order to prevent the release of these potentially harmful micropollutants.

3.1. The fate of micropollutants in WWTPs

Wastewater treatment plants generally employ a primary, a secondary and an optional tertiary treatment process. Tertiary treatment processes are commonly used to produce higher quality of discharged water for certain purposes (e.g. water reuse), and are always associated with high treatment cost. Thus, the requirement for tertiary treatment processes is generally based on public and environmental health objectives.

Primary treatment processes aim to remove suspended solids that enter WWTPs and are ineffective in removal of most micropollutants (Carballa et al., 2005). Micropollutants are removed mainly by sorption on primary sludge, as distribution of a compound into organic (lipophilic) layer is a predominant way of sorption (Ternes et al., 2004). Fragrances (galaxolide and tonalide) were found to be well removed (40%) during primary treatment (aerated grit chamber followed by circular sedimentation tank) due to their high partition coefficients between the solid and liquid phase (Carballa et al., 2004). Primary treatment (sedimentation tank) was also able to remove some EDCs moderately with removal efficiency ranging from 13% (nonylphenol monoethoxylate) to 43% (Bisphenol A) (Stasinakis et al., 2013). However, primary treatment using aerated grit chamber could cause significant increase of phenolic compounds, such as bisphenol A and nonylphenol, because the compounds originally attached to the grits could be peeled off due to air agitation in grit chamber (Nie et al., 2012). For pharmaceuticals and hormones, removal efficiency in primary treatment ranged up to only 28% (diclofenac and estriol), which suggested that adsorption of investigated compounds to sludge particles was rather limited (Behera et al., 2011). No considerable reduction was also reported for ibuprofen, naproxen, sulfamethoxazol and estrone (Carballa et al., 2004).

In secondary treatment, micropollutants are subjected to a range of processes, including dispersion, dilution, partition, biodegradation and abiotic transformation. The total removal during secondary treatment generally refers to the losses of a parent compound contributed by different mechanisms of chemical and physical transformation, biodegradation and sorption to solids (Jelic et al., 2011). Biodegradation/biotransformation and sorption are the two major removal mechanisms during biological treatment, while volatilization occurs to a minor degree (Verlicchi et al., 2012).

During secondary treatment, micropollutants are biologically degraded to various degrees, resulting in mineralization or incomplete degradation (production of by-products). Biodegradation of micropollutants can occur via different mechanisms: 1) single substrate growth of a small subset of specialist oligotrophic organisms, which is less common in WWTPs and more likely to occur in receiving water or sediment (Daughton and Ternes, 1999); 2) co-metabolism, in which micropollutants are decomposed by enzymes generated for other primary substation degradation (e.g. ammonia monooxygenase (AMO)) and are not used as carbon and energy source for microbial growth; and 3) mixed substrate growth, in which micropollutants are used as carbon and energy source and become mineralized (Vader et al., 2000). For pharmaceuticals, even if the compounds fall into the same therapeutical group, their biodegradability can show great variability. For example, Salgado et al. (2012) reported that, among NSAIDs, diclofenac exhibited low (< 25%) biodegradation, whereas ibuprofen and ketoprofen were biodegraded to a much higher extent (> 75%). Antibiotics are generally not readily biodegradable (Verlicchi et al., 2012). Regarding polycyclic musk, Clara et al. (2011) indicated that biological degradation serves as a minor removal pathway. 15% and 30% of galaxolide and tonalide were found to be eliminated via

biological transformation (Salgado et al., 2012). In contrast, Suárez et al. (2010) reported much higher biodegradation of tonalide and galaxolide (> 75%). As for steroid hormones, significant biodegradation (> 75%) was observed for estrone and estradiol (Suárez et al., 2010). Bisphenol A and triclosan were also found to be susceptible to biodegradation (up to 85% and 81% respectively), while nonylphenol was biologically transformed to a lesser degree (up to 56%) in two WWTPs using activated sludge (Samaras et al., 2013). In the case of pesticide, Stasinakis et al. (2009) found that almost 60% of diuron was biodegraded during an activated sludge process.

Sorption of micropollutants mainly occurs by (1) absorption, in which hydrophobic interactions occur between the aliphatic and aromatic groups of a compound and the lipophilic cell membrane of microorganisms as well as the fat fractions of sludge, and (2) adsorption, involving the electrostatic interactions of the positively charged groups with the negatively charged surfaces of the microorganisms and sludge (e.g. amino groups) (Ternes et al., 2004). Verlicchi et al. (2012) found that sorption onto solids is insignificant (< 5% in most cases) for most pharmaceuticals. In a study, mefenamic acid showed about 30% sorption (Jelic et al., 2011). In contrast, it was the major removal mechanism for some compounds, such as diclofenac, galaxolide and tonalide (Clara et al., 2011 and Salgado et al., 2012). Nonylphenol (35% to 51%) and triclosan (11% to 41%) were detected to be moderately removed via sorption to solids, while some acidic compounds (e.g., ibuprofen) could not be sorbed because of the charge repulsion between solids and compounds (Samaras et al., 2013). In general, the compounds that tend to be sorbed onto solids are expected to be better eliminated by activated sludge treatment than other low-cost secondary treatments (trickling filter beds, anaerobic lagoon and constructed wet lands) (Camacho-Muñoz et al., 2012). This can be due to the promoted biodegradation under forced aeration during the

conventional treatments, together with the enhanced sorption by large amounts of sludge generated in conventional treatment systems.

In WWTPs, there are circumstances where the effluent concentrations of some micropollutants exceed their influent concentrations. This can be explained by the presence of some substances, e.g. human metabolites and/or transformation products in the influent, which can subsequently be transformed back to parent compounds during biological treatment (e.g. diclofenac, carbamazepine, erythromycin, and sulfamethoxazole) (Göbel et al., 2007 and Kasprzyk-Hordern et al., 2009). In addition, some pharmaceuticals excreted with feces are probably partly enclosed in feces particles and released during biological treatment. The negative removal has also been ascribed to the daily concentration fluctuations during the sampling period, the analytical uncertainty, or desorption of molecules from sludge and suspended particulate matter (Clara et al., 2004 and Köck-Schulmeyer et al., 2013).

3.2. Overall removal of micropollutants in conventional WWTPs

The term “overall removal” generally refers to all the losses of micropollutant parent compounds from aqueous phase. Fig. 3 showing the WWTP removal efficiency of the most studied micropollutants in 14 countries/regions (data from Table 2) depicts compound-specific variation in removal (12.5 to 100%). Compounds even in the same usage class were removed to fairly different degrees. For individual compounds, large location-specific elimination disparities were also displayed. For example, diclofenac was significantly removed (81.4%) in a Korean WWTP (Behera et al., 2011) while it showed minor reduction (5%) in a Spanish WWTP (Rosal et al., 2010). Generally, the removal difference among different compounds in WWTPs could be ascribed to a number of factors such as micropollutant properties and operational conditions.

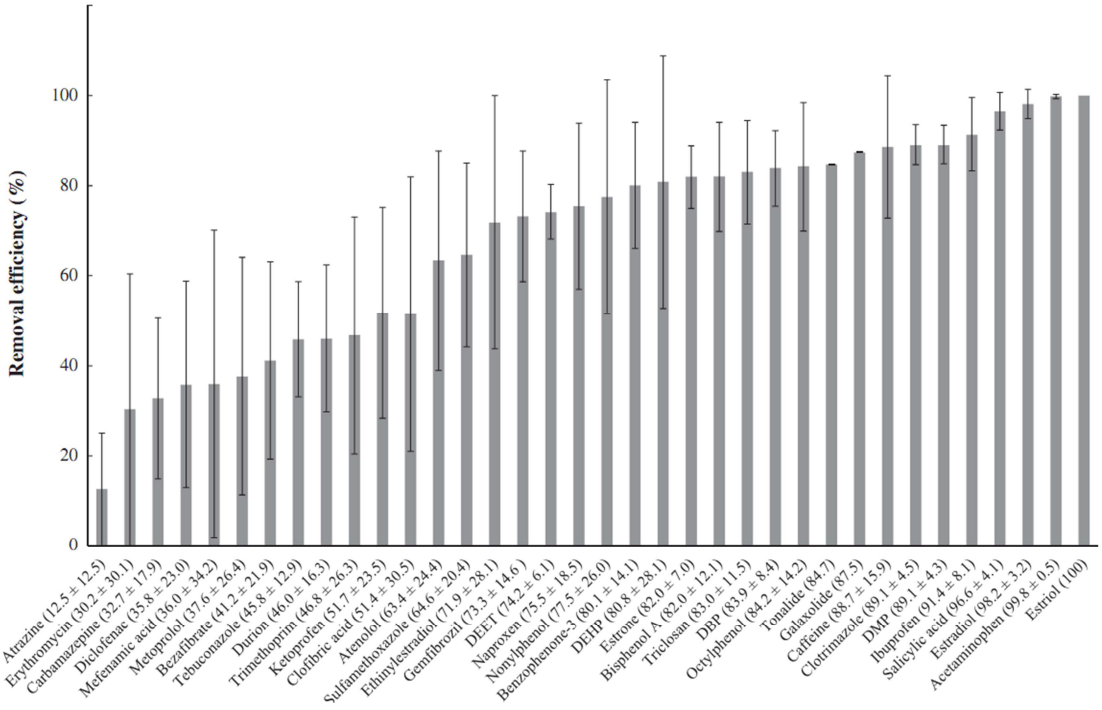

Fig. 3. Removals of the selected micropollutants in WWTPs (data from Table 2; negative removals not included). X-axis displays the selected compounds and their mean concentrations and standard deveations (in the brackets). Error bars represent the standard deviations of the data.

The most investigated micropollutants in WWTPs were NSAIDs. Ibuprofen, naproxen and ketoprofen exhibited moderate to high removal with average removal efficiency of 91.4%, 75.5% and 51.7%, respectively. In particular, the eliminations of ibuprofen were relatively consistent and commonly higher than 70%. As opposed to other NSAIDs, diclofenac experienced fairly inefficient (average 35.8%) and variable removals. The selected antibiotics showed low (erythromycin, 30.2%) to moderate removal (sulfamethoxazole, 64.6%). Lipid regulators and β-blockers were also not efficiently eliminated (37.6%–73.3%) in WWTPs. Anticonvulsant carbamazepine seemed to be the most persistent pharmaceutical and was averagely reduced by only 32.7%. Among all the reviewed studies, the highest removal of carbamazepine was observed by K. Choi et al. (2008), reaching 62.3%. As

mentioned above, caffeine was the most abundant compounds present in municipal wastewater. WWTPs proved to be effective in eliminating caffeine with an average removal efficiency of 88.7%. In the case of PCPs, relatively high reductions were exhibited, ranging between 74.2% (DEET) and 87.5% (galaxolide). As for steroid hormones, relatively stable and high removal efficiency was observed, which ranged from 71.9 to 100%. Two surfactants, nonylphenol and octylphenol, showed removals of 77.5% and 84.2%, respectively. Contradictory results have been reported for the elimination of nonylphenol, ranging from 21.7% (Stasinakis et al., 2008) to 99.0% (Janex-Habibi et al., 2009). The concentrations of bisphenol A were commonly considerably lowered (82%) during wastewater treatment. Other selected industrial chemicals also showed removal efficiencies exceeding 80%. Due to the fact that pesticides have been typically considered of agricultural rather than of urban origin, few studies have been performed at real plant scale and most of reported plants coincide in showing insufficient removal of pesticides (Köck-Schulmeyer et al., 2013). The selected pesticides, such as atrazine, fluconazole and tebuconazole, were particularly resistant in WWTPs.

It is difficult to draw a firm conclusion on the persistency of each compound, as many compounds showed significantly varied removals in different WWTP. However, a simple classification of these compounds is presented in Table 6.

Table 6
Simple classification of micropollutants based on removal efficiency.

Degree of removal	Compounds
Poorly removed (< 40%)	Atrazine, carbamazepine, diazinon, diclofenac, erythromycin, metoprolol, mefenamic acid, TCEP, TCPP
Moderately removed (40–70%)	Atenolol, bezafibrate, clofibric acid, durion, ketoprofen, nonylphenol, sulfamethoxzole, tebuconazole, trimethoprim
Highly removed (> 70%)	Acetaminophen, benzophenone-3, bisphenol A, caffeine, clotrimazole, DBP, DEET, DEHP, DMP, estradiol, estriol, estrone, ethinylestradiol, galaxolide, gemfibrozil, ibuprofen, naproxen, nonylphenol, octylphenol, salicylic acid, tonalide, triclosan

3.3. Factors governing the fate of micropollutants in WWTPs

The fate of micropollutants in WWTPs is under the control or influence of 'internal factors' and 'external factors'. Internal factors are micropollutant-related, including the characteristics of micropollutants (e.g. hydrophobicity, biodegradability, and volatility). In general, polar and non-volatile compounds are more likely to escape wastewater treatment processes. External factors are WWTP-specific, which are linked to the treatment conditions of wastewater treatment processes, the mixture of micropollutants that can act as competitors and nature of wastewater (pH and temperature).

3.3.1. Micropollutant-related factors

Sorption of a micropollutant to solids largely depends on the hydrophobicity of the compound. K_{OW} is frequently used to predict absorption of micropollutants on solids. Rogers (1996) provided a general rule of thumb for applying K_{OW} to the estimation of sorption: $\log K_{OW} < 2.5$ indicates low sorption potential, $2.5 < \log K_{OW} < 4$ indicates medium sorption potential, and $\log K_{OW} > 4$ indicates high sorption potential.

Acidity determined by the functional group of a compound can play an important role in chemisorption or/and electrostatic adsorption of micropollutants. Schäfer et al.

(2011) indicated that, at the pH above the acid dissociation constant (pK_a), the phenolic hydroxyl group of hormones dissociates and the compounds become negatively charged, facilitating the charge repulsion with the negatively charged membrane. Charge repulsion can also be expected to occur between negatively charged compounds and biomass in the activated sludge reactors, thereby impeding the removal of micropollutants.

In activated sludge processes, the solid-water distribution coefficient (K_d) is defined as the partition of a compound between the sludge and the water phase. Taking into consideration both K_{OW} and pK_a, K_d has been proposed as a relative accurate indicator of sorption behavior (Joss et al., 2005 and Ternes et al., 2004). For compounds having a K_d of below 300 L/kg ($\log K_d < 2.48$), the sorption onto secondary sludge can be considered to be insignificant. Additionally, Tadkaew et al. (2011) reported that the studied micropollutants with $\log K_d > 3.2$ (e.g. estrone and nonylphenol) were easily removed (> 85%).

As biodegradability of micropollutants depends on their bioavailability, the first phase of the biodegradation process is the uptake of micropollutants by cell, leading to by chance affinity of the compound with the bacterial enzymes (Siegrist et al., 2005). Compound structure also plays an important role in determining resistance of a micropollutant to biodegradation. The biodegradability of a compound intrinsically relies on the complexity of the compound (e.g. monocyclic or polycyclic) and its functional groups (e.g. halogen groups). In general, the easily degraded substances include 1) linear compounds with short side chains, 2) unsaturated aliphatic compounds, and 3) compounds possessing electron donating functional groups. On the other hand, the persistent micropollutants contain 1) compounds with long, highly branched side chains, 2) saturated or polycyclic compounds, and 3) compounds possessing sulfate, halogen or electron withdrawing functional groups (Jones et

al., 2005 and Tadkaew et al., 2011). Nevertheless, for some pharmaceutical compounds, there is no obvious relationship among chemical structure, functional groups and the removal. For example, two structurally similar compounds such as ibuprofen and ketoprofen could show different removals, with ibuprofen being eliminated more efficiently (Camacho-Muñoz et al., 2012).

Henry's law constant (k_H) is commonly used to characterize the volatility of a compound. The k_H ranging from 10^{-2} to 10^{-3} mol/($m^3 \cdot$ Pa) commonly indicates high tendency of volatilization (Stenstrom et al., 1989). According to Suárez et al. (2008), volatilization of micropollutants is totally negligible for pharmaceuticals and estrogens, nearly negligible for fragrance compounds tonalide and galaxolide and very significant for celestolide. Volatilization was found to account for up to 16% removal of celestolide (Suárez et al., 2010). Furthermore, in activated sludge processes, the volatilization behavior can be intensified due to the additional air supply.

3.3.2. WWTP-specific factors

Sludge retention time (SRT) controls the size and diversity of a microbial community. Enhanced elimination of micropollutants can be achieved if the treatment processes have extended SRTs, which facilitate the buildup of slowly growing bacteria, such as nitrifying bacteria. In nitrifying conditions, co-metabolism using ammonium monooxygenase enzyme is a possible degradation pathway for micropollutants. Nitrifying biomass have been found to have positive effects on the removal of a range of micropollutants such as ibuprofen, naproxen, trimethoprim, erythromycin, galaxolide, tonalide, ethinylestradiol, bisphenol A and nonylphenol (Fernandez-Fontaina et al., 2012 and Suárez et al., 2010).

Clara et al. (2005) suggested that the SRTs allowing nitrogen removal (nitrification and denitrification) above 10 days can enhance the elimination of some biodegradable compounds (e.g. ibuprofen, bezafibrate, natural estrogens and bisphenol A). In a study, the activated sludge treatment with an elevated SRT of 18 days could achieve considerably higher removal of beta blockers and psycho-activate drugs in comparison with the same treatment with shorter SRT of 0.5 day (Wick et al., 2009). Suárez et al. (2010) identified 10% higher removal efficiency for fluoxetine, citalopram and ethinylestradiol when prolonged SRT was applied. Enhanced biodegradation was found for 4-n-nonylphenol and triclosan at SRT of 20 days (compared with 3 days and 10 days) (Stasinakis et al., 2010). However, high SRT does not necessarily mean better removal performance. Joss et al. (2005) suggested that variation of the sludge age between 10 and 60–80 days showed no noticeable effects on removal efficiency of the investigated pharmaceuticals. High SRT (20 days) also seemed not to appreciably affect the biodegradation of bisphenol A (Stasinakis et al., 2010). Santos et al. (2009) indicated that application of low SRTs (1.5–5.1 days) had minor effects on the removal of some pharmaceutical compounds (e.g., ibuprofen, diclofenac, naproxen, and carbamazepine).

Hydraulic retention time (HRT) is the amount of time that allows for biodegradation and sorption. The micropollutants having slow/intermediate kinetics such as fluoxetine or some antibiotics will experience less effective biodegradation at shorter HRTs or increasing loading rates (Fernandez-Fontaina et al., 2012). Huang et al. (2008) indicated that HRT in the range from 5 to 14 h achieved minor removal of DEHP, while higher HRT increased DEHP accumulation in the system and DEHP retention in the waste sludge.

Redox conditions may cause the observed differences by having an effect on certain wastewater or sludge characteristics as wells as on the biodiversity of the microbial flora present (Göbel et al., 2007). Qiang et al. (2013) indicated that unfavorable redox conditions (anaerobic conditions) could result in inefficient biodegradation of some micropollutants. In another study, naproxen, ethinylestradiol, roxithromycin and erythromycin were found only considerably eliminated under aerobic condition and anoxic removal was much less effective (Suárez et al., 2010). Zwiener and Frimmel (2003) compared short-term biodegradation of clofibric acid, ibuprofen, and diclofenac in oxic and anoxic (denitrification conditions, absence of oxygen while presence of nitrate) biofilm reactor. In the oxic biofilm reactor, clofibric acid and diclofenac were not eliminated, with only 1–4% loss of their initial concentration being observed. Ibuprofen was reduced by 64–70%. By contrast, the anoxic biofilm reactor achieved much lower removal of ibuprofen (17–21%) and higher removal of diclofenac (34%–38%) and clofibric acid (26–30%). Goel et al. (2003)reported that removal of the nonylphenol ethoxylate surfactant was higher in the oxic reactors (50 to 70%) compared to the anoxic reactors (30 to 50%). Similarly, DEHP were removed by 15%, 19% and 62% in anaerobic, anoxic and aerobic reactors (Huang et al., 2008). Anoxic redox conditions were not necessarily less favorable environments for micropollutant removal. For instance, anoxic conditions could lead to improved elimination of iodinated X-ray contrast media, while aerobic environments witnessed minor removal (Drewes et al., 2001). Some persistent substances, diclofenac, sulfamethoxazole, trimethoprim and carbamazepine showed minor removals (< 25%) by the biological treatment with either nitrifying (oxic) or denitrifying bacteria (anoxic) (Suárez et al., 2010).

Wastewater characteristics, such as pH and temperature, may have effects on micropollutant removal. The acidity or alkalinity of an aqueous environment can vary the

elimination of micropollutants from wastewater by influencing both the physiology of microorganisms (pH optima of microbial enzyme activities) and the solubility of micropollutants present in wastewater (Cirja et al., 2008). Kimura et al. (2010) found that modest pH variation had significant effects on the removal of acidic pharmaceuticals (clofibric acid, ibuprofen, ketoprofen, naproxen and mefenamic acid) by the biosolids, which was presumably ascribed to activation of enzymes involved or enhancement of affinity between the biosolids and pharmaceuticals due to protonation of acidic pharmaceuticals. Seasonal variation of temperature may have impact on micropollutant removal in WWTPs. Temperature variation can affect biodegradation and partition (sorption and volatilization) of micropollutants. To eliminate the seasonal effect, alteration of operation parameters can be taken into consideration. For example, a possible strategy to improve EDC removal in the cold temperature is to increase the mixed liquor suspended solids (MLSS) concentration by raising the SRT (Nie et al., 2012). Generally, enhanced micropollutant removal can be achieved at warmer temperature due to promoted microbial activities (Nie et al., 2012 and Qiang et al., 2013). Nevertheless,Hai et al. (2011) found that operation at high temperature levels (45 °C) could lead to lower micropollutant removal. Some other studies showed that micropollutant elimination was independent of temperature fluctuation (Suárez et al., 2010).

4. Overview of treatment alternatives for micropollutant removal

No specific treatment is now available to assure the complete removal of various micropollutants due to their diverse properties. Reliable processes that are able to eliminate both bulk substances as well as micropollutants are yet to be developed. An overview of the current treatment options is present in the following sections to reveal the performance of each technique for micropollutant removal and to identify the need for improvement.

4.1. Coagulation–flocculation

Coagulation–flocculation is used for removing particulate matter, colloids as well as some dissolved substances. Table 7 presents some recent literature data regarding the removal of the most studied micropollutants from wastewater by coagulation–flocculation processes. In general, coagulation–flocculation processes yield ineffective elimination of most micropollutants. Matamoros and Salvadó (2013) evaluated the micropollutant removal in a coagulation/flocculation–lamellar clarifier for treating secondary effluent. The removals ranged from imperceptible elimination to 50%, among which the relatively high removals (20–50%) were observed for the compounds with $K_{OW} > 4$ at pH = 7–8 (e.g. galaxolide, tonalide, and octylphenol). Suárez et al. (2009) reported significant reduction (around 80%) of musks (e.g. galaxolide and tonalide) during coagulation–flocculation treatment of hospital wastewater. The other compounds that showed identifiable elimination were diclofenac (max. 46%), naproxen (max. 42%) and ibuprofen (max. 23%). Since landfill leachate has been considered as an important source of some EDCs, Asakura and Matsuto (2009) pointed out that treatment of landfill leachate by coagulation and sedimentation was not able to remove biphenol A but achieved much higher removals for DEHP and nonylphenol (70% and 90% respectively).

Table 7.
Removals of some micropollutants during coagulation–flocculation processes.

Coagulant	Dosage with pH value presented in the parentheses	Compound	Removal (%)	References
$FeCl_3$/$Al_2(SO_4)_3$	25, 50 ppm (7)	Ibuprofen	12.0 ± 4.8	Suárez et al. (2009)
		Diclofenac	21.6 ± 19.4	
		Naproxen	31.8 ± 10.2	
		Carbamazepine	6.3 ± 15.9	
		Sulfamethoxazole	6.0 ± 9.5	
		Tonalide	83.4 ± 14.3	
		Galaxolide	79.2 ± 9.9	
$FeCl_3$	100, 200 mg/L (4, 7, 9)	Bisphenol A	20	Asakura and Matsuto (2009)
		DEHP	70	
		Nonylphenol	90	
$Al_2(SO_4)_3$	200 mg/L (7)	Aldrin	46	Thuy et al. (2008)
	100 mg/L (7)	Bentazon	15	
Not mentioned	Not mentioned	Ibuprofen	4	Matamoros and Salvadó (2013)
		Ketoprofen	4	
		Carbamazepine	2	
		Tonalide	24	
		Galaxolide	16	
		Celestolide	50	
		Triclosan	24	
		DMP	19	
		Octylphenol	50	

As a whole, most micropollutants, as shown above, have been reported to be poorly removed during coagulation–flocculation processes. Exceptions were some musks, a few pharmaceuticals (e.g. diclofenac) and nonylphenol due to their high K_{OW} (4–6). Besides, neither coagulant dose nor operation temperature influenced the removal of pesticides significantly (Thuy et al., 2008). Despite the minor differences among different types of coagulants at different doses, Suárez et al. (2009) reported that the addition of 25 mg/L $FeCl_3$ achieved optimal results in most cases. Huerta-Fontela et al. (2011)demonstrated that aluminum sulfate was effective in eliminating some hydrophobic pharmaceutical compounds. Composition of wastewater can exert either positive or negative effects on micropollutant removal during coagulation–flocculation treatment. For example, high fat content in water

source was reported to improve the removal of hydrophobic compounds (Suárez et al., 2009). Dissolved humic acid could also enhance the elimination of some pharmaceutical compounds, such as diclofenac, ibuprofen and bezafibrate (Vieno et al., 2006). On the contrary, the presence of dissolved organic matters (DOM), especially low-molecular-weight fractions, can possibly inhibit the micropollutant removal due to the preferential removal of DOM through coagulation. Negatively charged DOM could react with positively charged aluminum hydrolysis species, leading to a less amount of coagulant available for elimination of the compounds (K.-J. Choi et al., 2008). In addition, the performance of coagulation–flocculation processes can be also governed by several operating conditions including mixing conditions, pH, alkalinity, temperature as well as the presence of divalent cations and concentrations of destabilizing anions (e.g. bicarbonate, chloride, and sulfate) (Alexander et al., 2012).

4.2. Activated carbon adsorption

Adsorption by activated carbons (ACs) is commonly employed for controlling taste and odor in drinking water. This technique has also great potential for treatment of secondary effluent and has proved to be more effective in removing micropollutants in comparison with coagulation–flocculation process (K.-J. Choi et al., 2008). Both powdered activated carbon (PAC) and granular activated carbon (GAC) have been widely used in adsorption processes (Table 8), which can be affected by the properties of both adsorbate (K_{OW}, pK_a, molecular size, aromaticity versus aliphaticity, and presence of specific functional groups) and adsorbent (surface area, pore size and texture, surface chemistry, and mineral matter content) (Kovalova et al., 2013).

Table 8
Removals of some micropollutants during adsorption process.

Adsorbent	Dosage	Compound	Removal (%)	References
PAC	8, 23, 43 mg/L	Diclofenac	96, 98, 99	Kovalova et al. (2013)
		Carbamazepine	98, 99, 100	
		Propranolol	> 91, > 94, > 94	
		Sulfamethoxazole	2, 33, 62	
GAC	Full scale	Diclofenac	> 98	Grover et al. (2011)
		Carbamazepine	23	
		Propranolol	17	
		Estrone	64	
		17β-Estradiol	> 43	
		17α-Ethinylestradiol	> 43	
	29 g/70.6 mL bed volume	Galaxolide	79	Hernández-Leal et al. (2011)
		Tonalide	67	
		Bisphenol A	66	
		Nonylphenol	84	
		Triclosan	95	
	Full scale, empty bed contact time:15 min	Diclofenac	~ 100	Yang et al. (2011)
		Trimethoprim	90	
		Carbamazepine	75	
		Caffeine	45	
		Primidone	30	
		DEET	15	

4.2.1. PAC

PAC has been considered as an effective adsorbent for treating persistent/non-biodegradable organic compounds. An advantage of employing PAC is that it can provide fresh carbon continuously or can be used seasonally or occasionally when risk of trace organics is present at a high level (Snyder et al., 2007).Kovalova et al. (2013) investigated the elimination of micropollutants from a MBR-treated hospital effluent using PAC treatment at a retention time of two days. With PAC doses of 8, 23 and 43 mg/L and retention time of 2 days, the PAC reactor achieved efficient elimination for most of the micropollutants (pharmaceuticals, metabolites and industrial chemicals). The reduction of total load of selected pharmaceuticals and metabolites was around 86%. Batch tests performed by Hernández-Leal et al. (2011)also demonstrated marked removal (> 94%) of various

micropollutants (personal care products, bisphenol A and nonylphenol) during PAC treatment with initial compound concentrations of 100–1600 μg/L at a dose of 1.25 g/L and a contact time of 5 min.

PAC addition in activated sludge tank or post treatment configurations is a major application of PAC in the full-scale municipal WWTPs. A study was carried out to assess the efficiency of micropollutant removal by addition of PAC in different flow schemes in municipal wastewater treatment (Boehler et al., 2012). It was found that counter-current use of PAC by recycling waste PAC from post-treatment tank to biological treatment tank could enhance micropollutant removal by 10 to 50% in comparison with the application without recycling. PAC addition in WWTPs was shown to be able to reduce micropollutant levels by more than 80%. The PAC dosage for adequate treatment of secondary effluent with dissolved organic carbon (DOC) of 5–10 mg/L was 10–20 mg/L, while a higher amount (30–40 g/m^3 influent) was required to achieve similar results if direct PAC addition was employed in biology tank.

The performance of PAC in eliminating micropollutants depends upon PAC dose and contact time, the molecular structure and behavior of the targeted compound, as well as the water/wastewater composition (Boehler et al., 2012 and Snyder et al., 2007). Either higher dose or longer contact time can probably result in greater removal of micropollutants. Westerhoff et al. (2005) revealed that micropollutant removal was improved with higher PAC dosages (20 mg/L) and independent of the initial compound concentrations. Water/wastewater composition also affects the adsorption of micropollutants. The sorption efficiency of PAC could be reduced as the DOC content increases (Boehler et al., 2012). Despite the influence of other contaminants in wastewater, the efficacy of applying PAC to

wastewater for micropollutant removal is comparable with that of ozonation. Thus, PAC addition appears an attractive method for upgrading municipal WWTPs for improved micropollutant removal (Bolong et al., 2009).

4.2.2. GAC

Rossner et al. (2009) suggested that GAC dosage typically applied to taste and odor control in drinking water (< 10 mg/L) was sufficient to provide a 2-log removal for most of various compounds in a lake water.Hernández-Leal et al. (2011) evaluated the effectiveness of GAC in treating two wastewaters: (1) spiked (0.1–10 μg/L) aerobic effluent in a GAC column operated at low flow and (2) aerobic effluent with real concentrations (40 ng/L to 7.9 μg/L) of micropollutants in a GAC column. In the first case, removals for all the compounds were generally high (> 67%), particularly for ethyl-, propyl- and butylparaben, triclosan and caffeine (> 90%). In the second case, most compounds were also effectively eliminated. Specifically, the removal efficiency ranged from 50% (tonalide and nonylphenol) to more than 90% (galaxolide).

A full-scale granular activated carbon plant treating a WWTP effluent was assessed in terms of the removal efficiency of steroidal estrogens and pharmaceuticals (Grover et al., 2011). Considerable removals of steroidal estrogens from sewage effluent were observed during the GAC tertiary treatment. By comparison, the reduction of pharmaceutical concentrations was more variable. For example, higher removals (84–99%) were observed for mebeverine, indomethacine, and diclofenac, while some compounds (e.g. carbamazepine and propranolol) displayed much less removals (17–23%). In spite of the efficient treatment of sewage effluent, GAC-based removal technology should be carefully operated, as the efficiency will decrease over time due to the saturation of adsorption site.

Similar to PAC, the contact time is a major factor that affects the degree of adsorption. Short contact time is likely to lead to significantly lowered adsorption efficiency. As the elimination of the trace contaminants depends largely upon particle–contaminant interactions, the competition for adsorption sites and/or pore blocking (by particle solids) can reduce the removal efficiency of activated carbon (Bolong et al., 2009). Thus, GAC tends to perform poorly if wastewaters are highly contaminated. Snyder et al. (2007) suggested that a steam-treated GAC could be employed to overcome the drawbacks of GAC due to its greater absorption capacity. Regular regeneration of GAC also seemed of vital importance to maintain minimal breakthrough of micropollutants. Furthermore, pore shape/size and volumes of activated carbons, carbon type, surface charge of compounds and operation year were noted to have influence on the removal performances (Choi et al., 2008b and Rossner et al., 2009): 1) Broader micropore size distribution of the GAC led to more efficient adsorption of micropollutants with different shapes and sizes; 2) Pore volume was more important to adsorption capacity than specific area and larger pore volume was commonly associated with greater removal efficiency; 3) Negatively charged micropollutants were likely to be poorly adsorbed by the negatively charged carbon and well adsorbed by the positively charged carbon; and 4) Adsorption capacity reduced with operation year.

From the aforementioned studies, GAC and PAC appear to be attractive methods for micropollutant removal. In general, efficient removal is potentially achievable when the compounds have non-polar characteristics ($K_{OW} > 2$) as well as matching pore size/shape requirements (Rossner et al., 2009 and Verlicchi et al., 2010b). However, activated carbon efficacy might be significantly lowered by the presence of natural organic matter (NOM) which competes for binding sides, thereby resulting in blocked pores. Besides, PAC dose,

GAC regeneration as well as contact time play important roles in efficient removal of micropollutants.

4.3. Ozonation and advanced oxidation processes (AOPs)

Due to the refractory nature of some micropollutants, conventional physicochemical and biological treatments are not able to provide adequate elimination of these compounds. To overcome the problem, ozonation and AOPs can be considered. Performance of these processes in micropollutant removal is reported in Table 9. Ozonation and AOPs are efficient redox technologies which demonstrate some superiority over conventional treatments, such as high degradation rates and non-selectivity. Moreover, these processes have disinfecting effects, which are essential for reuse applications that involve direct human contact, e.g., household reuse applications (Hernández-Leal et al., 2011). Ozone can degrade contaminants directly and indirectly (mainly via formation of stronger and less selective oxidizing agent, **.** OH). Some micropollutants are susceptible to both ozone and AOPs (e.g., naproxen and carbamazepine), whereas some are only subject to **.**OH (e.g. atrazine and meprobamate) and some are resistant to both forms of oxidation (e.g. TCEP and TCPP) (Gerrity et al., 2011). The generation of **.**OH can be promoted with the presence of H_2O_2, Fenton reagent and ultraviolet.

Table 9
Removals of some micropollutants during ozonation and AOPs.

Treatment	Compound	Removal (%)
O_3 (5 mg/L): 15 min (Sui et al., 2010)	Carbamazepine	> 90
	Diclofenac	> 90
	Metoprolol	80–90
	Bezafibrate	0–50
	Trimethoprim	> 90
	DEET	50–80
O_3 (15 mg/L) (Hernández-Leal et al., 2011)	Tonalide	79
	Galaxolide	> 87
	Nonylphenol	> 79
O_3 (5 mg/L) + H_2O_2 (3.5 mg/L) (Gerrity et al., 2011)	Ibuprofen	83
	Diclofenac	> 99
	Carbamazepine	> 99
	Sulfamethoxazole	98
	Triclosan	> 99
	Bisphenol A	> 78
	Estradiol	> 83
	Estrone	> 98
	Atrazine	69
UV_{254}: 10 min (De la Cruz et al., 2012)	Ibuprofen	34
	Diclofenac	100
	Carbamazepine	23
	Sulfamethoxazole	51
	Atrazine	69
UV_{254} + H_2O_2 (50 mg/L): 10 min, 30 min (De la Cruz et al., 2012)	Ibuprofen	100 (10 min), 100 (30 min)
	Diclofenac	100 (10 min), 100 (30 min)
	Carbamazepine	75 (10 min), 100 (30 min)
	Sulfamethoxazole	98 (10 min), 100 (30 min)
	Atrazine	100 (10 min), 100 (30 min

Ozonation is a promising technique to considerably decrease the micropollutant load of full-scale WWTPs (Hollender et al., 2009). Hernández-Leal et al. (2011) examined the efficiency of ozonation for the removal of a wide range of micropollutants (UV-filter, fragrance, biocide and surfactant) from biologically treated grey water. In general, all the compounds were significantly removed (> 79%) from the biologically treated effluent at an applied ozone dose of 15 mg/L. In another study, lower ozone dose of 5 mg/L also showed high removal efficiency for most of the targeted micropollutants (Sui et al., 2010). The

concentrations of carbamazepine, diclofenac, indomethacin, sulpiride and trimethoprim were considerably reduced by more than 95%. The reductions of DEET and metoprolol were modest. By contrast, bezafibrate was very resistant to ozonation and was removed by only 14%.

A study conducted by Gerrity et al. (2011) focused on the application of O_3/H_2O_2 for removing a suite of micropollutants (PPCPs and steroid hormones) during water reclamation. The process showed considerable removal efficiency (> 90%) for almost all of the target contaminants, except TCEP (13%), TCPP (26%), atrazine (69%), meprobamate (80%), and ibuprofen (83%). They indicated that micropollutants which exhibited the highest levels of oxidation were characterized by high ozone and **.**OH rate constants associated with their electron-rich moieties (e.g., phenols, anilines, olefins and activated aromatic). Although the formation of **.**OH was enhanced under alkaline conditions, Zhang et al. (2012)reported lower pH was beneficial for EDCs removal by ozone when treating synthetic secondary effluent. This is because ozone was less reactive to the inorganic and organic matters (non-target compounds) in the synthetic secondary effluent as compared to **.**OH (generated at high pH) and a greater amount of O_3could thereby be preserved for the reactions with target compounds. Furthermore, in spite of the fact that suspended sludge particles could lead to higher O_3 consumption, which might reduce the efficiency of ozonation for micropollutant removal, this effect was not significant and had only a minor impact on ozonation as well as oxidation by **.**OH at low O_3 dosages (Hernández-Leal et al., 2011 and Huber et al., 2003).

Kim et al. (2009b) examined the effectiveness of UV (wave length: 254 mm)-based processes (UV and UV/H_2O_2) for the elimination of 41 pharmaceutical compounds. UV alone could significantly remove (> 90%) only a few compounds (e.g. ketoprofen, diclofenac and

antipyrine) while ineffective removals (24%–34%) were observed for macrolides. By contrast, with the addition of H_2O_2 (7.8 mg/L), the process considerably improved its efficacy and removal efficiency increased up to 90% for 39 out of 41 compounds. Treatment of 32 selected micropollutants (pharmaceuticals, corrosion inhibitors and biocides/pesticides) in an effluent coming from a municipal activated sludge WWTP was also investigated using UV (wavelength: 254 nm), UV/H_2O_2, Fenton ($Fe^{2+,3+}/H_2O_2$) and photo-Fenton ($Fe^{2+,3+}/H_2O_2$/UV and $Fe^{2+,3+}/H_2O_2$/simulated sunlight) (De la Cruz et al., 2012). The process with only UV irradiation yielded a global degradation of 46% for the micropollutants after 10 min. Four compounds (diclofenac, ketoprofen, mefenamic acid and diuron) were completed removed during the process. In contrast, the concentrations of gabapentin, trimethoprim, metformin, primidone, azithromycin and clarithromycin were unaltered or only slightly reduced (< 10%). Comparing with UV treatment alone, UV and H_2O_2 (50 mg/L) exhibited elevated transformation (a total degradation of 81%) of the micropollutants. After 30 min of UV/H_2O_2, the transformation increased further up to 97%. Fenton process (5 mg/L $Fe^{2+,3+}$/50 mg/L H_2O_2) achieved 31% degradation. It was able to completely eliminate only one of the micropollutants, norfloxacin, after 30 min, and the concentrations of ten compounds were reduced by less than 15%. When UV was applied to the Fenton process (under the same conditions mentioned above), significantly increased global degradation (97%) was observed. For the photo-Fenton process, either increased H_2O_2 dosage or extended reaction time was found to have positive impact on the global degradation. Fenton/UV_{254} (100% degradation after 90 min) displayed much higher degradation efficiency compared with Fenton/sunlight (47% degradation after 90 min). In addition, the presence of dissolved organic matter in the wastewater seemed to enhance the micropollutant removal during all the processes. In another study, Klamerth et al. (2010) reported much higher efficiency of photo-Fenton with solar light for treatment of 52 micropollutants (PPCPs and

pesticides) in a WWTP effluent. The process was able to reduce 48 compounds to below their limit of detection.

Since oxidation processes do not commonly result in complete mineralization of micropollutants, the major concern of applying these processes is the formation of oxidation by-products (or transformation products) from micropollutants. Research data indicated that the by-products generally have low concentration levels as well as insignificant estrogenic and antimicrobial activity compared to the parent compounds (Hollender et al., 2009 and Reungoat et al., 2010). To further reduce parent compounds and oxidation by-products, biological post-filtration (sand filtration or activated carbon filtration) can be considered.

4.4. Membrane processes

Table 10 presents some recent research data concerning the effectiveness of membrane technology in eliminating micropollutants. The retention of micropollutants in membrane processes can generally achieved by size exclusion, adsorption onto membrane, and charge repulsion. These removal mechanisms are largely dependent on a number of factors, such as membrane process type, membrane characteristics, operating conditions, specific micropollutant characteristics and membrane fouling (Schäfer et al., 2011).

Table 10
Removals of some micropollutants during membrane processes.

Membrane	Water type	Membrane conditions	Compound	Removal (%)	References
UF	Synthetic water	PES[a] flat-sheet, 100 kDa; TMP = 0.5 ± 0.01 bar	Ibuprofen	7	Jermann et al. (2009)
		RC4[b] flat-sheet; TMP = 0.5 ± 0.01 bar		Minor	
		PES flat-sheet, 100 kDa; TMP = 0.5 ± 0.01 bar	Estradiol	Up to 80	
		RC4 flat-sheet; TMP = 0.5 ± 0.01 bar		Up to 25	
NF	WWTP effluent	Flat-sheet, area 3.5 m^2; TMP = 0.3 or 0.7 bar	Diclofenac	60	Röhricht et al. (2009), Yangali-Quintanilla et al. (2011)
		Flat-sheet, area 3.5 m^2; TMP = 0.3 or 0.7 bar	Naproxen	60	
		Flat-sheet, area 3.5 m^2; TMP = 0.3 or 0.7 bar	Carbamazepine	Minor	
		Filmtec NF90; TMP = 345 kPa		91	
		Filmtec NF200; TMP = 483 kPa	Acetaminophen	23	
		Filmtec NF200; TMP = 483 kPa	Ethynilestradiol	90	
		Filmtec NF90; TMP = 345 kPa	Atrazine	97	
RO	WWTP effluent	–	Ibuprofen	99	Sahar et al. (2011), Yangali-Quintanilla et al. (2011)
	Secondary effluent	Filmtec TW30; TMP = 9.5–10.2 bar	Ibuprofen	> 99	
		Filmtec TW30; TMP = 9.5–10.2 bar	Sulfonamides	> 93	
		Filmtec TW30; TMP = 9.5–10.2 bar	Diclofenac	95	
		Filmtec TW30; TMP = 9.5–10.2 bar	Macrolides	> 99	
		Filmtec TW30; TMP = 9.5–10.2 bar	Bisphenol A	> 99	

[a] PES: polyethersulfone.
[b] RC: regenerated cellulose.

Although microfiltration (MF) and ultrafiltration (UF) are proved processes to efficiently eliminate turbidity, micropollutants are generally poorly removed during UF and MF, as the membrane pore sizes are much larger than the molecular sizes of micropollutants. However, micropollutants can be removed via adsorption on to membrane polymers, as well as interaction with NOM in wastewater. Jermann et al. (2009) examined the fate of ibuprofen and estradiol during an UF process and the effects of fouling by NOM. Without NOM, UF with hydrophilic membrane showed insignificant removal for ibuprofen and low (8%) removal for estradiol, while hydrophobic membrane retained much larger amount of estradiol (up to 80%) and ibuprofen (up to 25%). The higher retention of estradiol was due to the higher Carbon–Water Partitioning Coefficient (Koc) value of the compound. As for the effect of NOM, NOM substances of high molecular weight such as alginate and Aldrich humic acid showed a greater effect than the lower molecular weight Nordic aquatic humic acid on enhancing micropollutant removal. Due to the low removal efficiency, MF or UF alone is not feasible for micropollutant removal. Hence, the combination of MF or UF with other processes (e.g. NF or RO) is essential for enhanced elimination of different micropollutants. Garcia et al. (2013) combined MF with RO to remove micropollutants for municipal wastewater reuse. MF was found to be able to reduce the concentrations of some compounds, such as DEHP, by more than 50%. With the incorporation of RO, the removal efficiency was significantly improved, ranging from 65% to 90% for most micropollutants (except ibuprofen and nonylphenol). Similarly, a tertiary MF/RO treatment process exhibited very efficient retention (> 95%) of most of the studied PPCPs, except mefenamic acid and caffeine (Sui et al., 2010).

In comparison with MF and UF, nanofiltration (NF) and reverse osmosis (RO) have much ‘tighter’ structures. NF and RO are widely used in water reuse industry due to their

high contaminant removal efficiency. However, NF and RO membranes are still somewhat permeable to some relatively small micropollutants (Steinle-Darling et al., 2010).

Röhricht et al. (2009) investigated two different types of submerged NF flat sheet modules for the removal of pharmaceuticals from WWTP effluent. Naproxen and diclofenac (60%) were retained to a greater extent compared with carbamazepine (slight removal). At pH 7 and 8, naproxen and diclofenac (with pK_a values of 4.2 and 4.15, respectively) were deprotonated, while carbamazepine (pK_a = 13.9) was not. Hence, naproxen and diclofenac could be rejected by the negatively charged membrane surface, whereas carbamazepine could not be removed. This was in accordance with the viewpoint indicated by Schäfer et al. (2003) and Nghiem et al. (2005): the speciation of pharmaceuticals may result in a significant change in rejection as a function of pH, with much greater retention occurring for ionized, negatively charged pharmaceuticals. For uncharged pharmaceuticals, intrinsic physicochemical properties of the pharmaceutical molecules play a role in their retention. Apart from electrostatic repulsion, adsorption can serve as the overriding removal mechanism in some cases. This was demonstrated in a study evaluating the removal of a variety of EDC/PPCPs using UF or NF (Yoon et al., 2006). For more polar compounds, the NF membrane (44–93% removals except naproxen of no rejection) was more efficient than the UF membrane with typical removals of less than 40% except a few compounds (triclosan, 87%; oxybenzone, 77%; progesterone, 56%). By contrast, for the less polar compounds, many permeate EDC/PPCP concentrations (14 out of the 25 compounds) were below detection, suggesting high removal efficiency by both NF and UF membranes. Better performance was also observed for NF.

RO generally shows great potential to partially or significantly remove micropollutants. Sahar et al. (2011)applied RO after CAS-UF and MBR processes and assessed its efficiency in eliminating micropollutants. The two processes, CAS-UF/RO and MBR/RO, showed relatively similar and high elimination efficiencies: > 99% for macrolides, pharmaceuticals, cholesterol and bisphenol A, 95% for diclofenac, 97% for sulfamethoxazole, and > 93% for both sulfamethazine and trimethoprim. Despite the highly effective RO treatment, 28–223 ng/L residuals of ibuprofen, diclofenac, salicylic acid, cholesterol, and bisphenol A were detected in the permeates from both units. This elucidated that RO was not an absolute barrier for micropollutants and complementary treatment processes should be considered to aid the RO to achieve complete elimination of micropollutants. Yangali-Quintanilla et al. (2011) compared the various micropollutants (pharmaceuticals, pesticides, endocrine disruptors and others) removal by NF and RO. The elimination efficiency of NF membranes was very close to that achieved by RO membranes. The average retention efficiency by tight NF was 82% for neutral contaminants and 97% for ionic contaminants, while RO was able to achieve 85% removal of neutral contaminants and 99% removal of ionic contaminants.

4.5. Membrane bioreactor

Membrane bioreactor (MBR) process combine activated sludge biological treatment and membrane filtration (MF and UF). MBRs possess the following advantages over conventional wastewater treatment in the following aspects (Ngo et al., 2012) such as high effluent quality, excellent microbial separation ability, absolute control of SRTs and HRTs, high biomass content and less sludge bulking problem, low-rate sludge production, small footprint and limited space requirement, and possibilities for a flexible and phased extension of existing wastewater treatment plants (WWTPs).

MBRs are able to effectively remove a wide spectrum of micropollutants including compounds that are resistant to activate sludge processes (Radjenovic et al., 2009). This is because 1) They are able to retain sludge to which many compounds are adhered; 2) The membrane surface can also intercept the compounds; and 3) The longer SRT in MBRs may promote microbial degradation of the compounds (Spring et al., 2007). Table 11 summarizes some recent studies involving MBR processes. The removal of micropollutants in MBR can be affected by a number of factors, such as sludge age and concentration, existence of anoxic and anaerobic compartments, composition of the wastewater, operating temperatures, pH and conductivity (Kovalova et al., 2012).

Trinh et al. (2012) investigated the micropollutant removal efficiency of a full-scale MBR. High elimination (> 90%) was observed for most of the micropollutants. Nevertheless, some compounds were incompletely removed (24–68%), including amitriptyline, carbamazepine, diazepam, diclofenac, fluoxetine, gemfibrozil, omeprazole, sulfamethoxazole and trimethoprim. Hence, these compounds were considered as potential indicators for evaluating the micropollutant removal using MBR processes. Generally, hospitals are the major source of many pharmaceuticals released into the environment (Verlicchi et al., 2010a). A pilot-scale MBR was employed for on-site treatment of hospital effluent (Kovalova et al., 2012). The overall reduction of all pharmaceuticals and metabolites was only 22%, as a large fraction (80%) of the feed was persistent iodinated contrast media. However, if the iodinated contrast media were not taken into account, the reduction would be up to 90%. Full-scale MBR studies for hospital wastewater treatment were also investigated by Beier et al. (2011), which suggested that separation of rainwater collection and water streams with low

pharmaceutical concentrations, and maintenance of sludge age > 100 days should be considered in the design of MBR for hospital wastewater.

Table 11
Removals of some micropollutants during MBR processes.

Water type	Membrane & experimental conditions	Compounds	Removal (%)	References
Raw wastewater	Full-scale HF [a] (Koch Puron); MA[b] 235 m^2; pore size 0.1–0.2 μm; SRT: 10–15 days; HRT: 1 day; MLSS: 7.5–8.5 g/L	Ibuprofen	~ 100	Trinh et al. (2012)
		Diclofenac	43	
		Carbamazepine	24	
		Sulfamethoxazole	60	
		Trimethoprim	30	
		Estrone,	~ 100	
		Estriol	~ 100	
		BisphenolA	~ 100	
Synthetic wastewater	Lab-scale polyvinylidene fluoride HF; MA 0.2 m^2; pore size 0.4 μm; HRT: 1 day or 3 days; MLSS: 2.3–4.6 g/L	Ibuprofen	~ 100	Bo et al. (2009)
		Diclofenac	Minor	
		Carbamazepine	Minor	
Synthetic wastewater	Lab-scale polyethylene hollow fiber; MA 0.2 m^2; pore size 0.4 μm; HRT: 8, 6 and 4 h; SRT: 350 days; MLSS: 5.2–13.7 g/L	BisphenolA	> 93.7	Chen et al. (2008)
Hospital effluent	Pilot-scale submerged PES UF flat sheet; area 7 m^2; pore size 38 nm; SRT: 30–50 days; MLSS: 2 g/L	Carbamazepine	− 6	Kovalova et al. (2012)
		Trimethoprim	96	
		Sulfamethoxazole	7	
		Atenolol	99	
Synthetic wastewater	Lab-scale submerged HF UF module; MA 0.047 m^2; pore size 0.04 μm; SRT:70 days; HRT: 24 h; MLSS: 8.6–10 g/L	Ibuprofen	96.7 ± 0.7	Tadkaew et al. (2011)
		Diclofenac	17.3 ± 4.2	
		Carbamazepine	13.4 ± 4.3	
		Sulfamethoxazole	91.9 ± 0.6	
		17β-estradiol	> 99.4	
		17α-ethynylestradiol	93.5 ± 1.2	
		Bisphenol A	90.4 ± 3.1	
		Nonylphenol	99.3 ± 0.2	
		Atrazine	4.4 ± 3.7	
Hospital effluent	Full-scale 5 Kubota EK 400 flat sheet; flow rate: 130 m/d	Ibuprofen	> 80	Beier et al. (2011)
		Carbamazepine	< 20	
		Diclofenac	< 20	

a Hollow fiber.

b MA: membrane area.

Both being cost effective technologies in wastewater treatment, MBR processes and conventional activated sludge (CAS) processes have been frequently compared in terms of their performance in micropollutant removal. Radjenovic et al. (2007) compared the removal of several pharmaceutical products in a laboratory scale MBR and a CAS process. Both systems were effective in removing some compounds (e.g., naproxen, ibuprofen, acetaminophen, hydrochlorothiazide, and paroxetine). However, the results presented that pharmaceuticals showed greater and steadier elimination during MBR process (> 80% in most cases). Another comparative investigation of MBR and CAS process was performed byChen et al. (2008). Similarly, MBR was slightly more efficient in micropollutant removal. The efficiency of elimination in the MBR appeared stable regardless of changes in sludge loading and HRT.

Biological treatment combined with membrane filtration (MF or UF) are also employed for treating wastewater. Sahar et al. (2011) compared the removals of several macrolide, sulphonamide and trimethoprim antibiotics from raw sewage using a full-scale CAS system coupled with a subsequent UF filtration (CAS–UF) and a pilot scale MBR. Antibiotics removal in the MBR system was generally higher than that in the CAS–UF system. The elimination of trimethoprim, sulfamethoxazole and erythromycin was 99%, 70%, 61% in the MBR system, and 45%, 52% and 71% in the CAS–UF system, respectively. It was assumed that antibiotics removal in both systems was due either to sorption to biomass (rather than biological transformation) or to enmeshment in the membrane biofilm (as the pore size of UF is significantly larger than the antibiotic molecules).

Recently, membranes in conjunction with anaerobic reactors have been gaining popularity due to their intrinsic advantages over aerobic systems, such as low sludge production, net energy generation and a fully enclosed environment (Hu and Stuckey, 2006). The applications of anaerobic MBRs for micropollutant removal have been investigated in some recent studies (Abargues et al., 2012 and Xu et al., 2008). A pilot-scale submerged anaerobic MBR (SAnMBR), a conventional activated sludge (CAS) unit and a pilot-scale aerobic MBR were evaluated for removing some alkylphenols and hormones (Abargues et al., 2012). The observed concentrations of alkylphenols in the SAnMBR effluent were consistently at significantly higher levels than those in the permeates from other units, indicating the ineffective removal of alkylphenols by SAnMBR.

During MBR processes, several operational parameters (e.g. SRT, HRT and temperature) can influence the reduction of micropollutants. In general, MBRs have high SRTs, thus diverse microorganisms, including some slow growing bacterial, can reside in the reactors. When biomass is rich in nitrifying bacteria, higher biodegradation efficiency for certain micropollutants can be achieved (Roh et al., 2009).De Gusseme et al. (2009) reported a high elimination (99%) of 17α-ethinylestradiol (at initial concentration of 83 ng/L) when a nitrifier enrichment culture was applied in a MBR. The degradation of micropollutants by nitrifying bacteria has also been evaluated in other types of systems (e.g., activated sludge and fixed bed reactor) (Batt et al., 2006, Forrez et al., 2009 and Zhou and Oleszkiewicz, 2010). A general conclusion drawn from these studies is that nitrifying conditions have positive effects on micropollutant removal. Temperature variability has been linked to decrease in bulk water quality parameters and unreliability of system, as microbial growth and activity as well as solubility and other physicochemical properties of organics are significantly affected by temperature (Hai et al., 2011). Effects of temperature variation were explored in a lab-scale

MBR treating wastewater containing selected micropollutants (Hai et al., 2011). Both hydrophobic compounds (logD > 3.2) and less hydrophobic compounds (logD < 3.2) showed reduced elimination at 45 °C, which was ascribed to disrupted metabolic activity typically linked to such elevated temperature. The removal of hydrophobic compounds was unaffected in the temperature range of 10–35 °C, while a relatively more obvious variation was found in the removals of less hydrophobic compounds.

4.6. Attached growth treatment processes

Attached growth technology is a promising alternative to activate sludge processes for wastewater treatment which involves attached growth on inert carriers either fixed or mobilized in suspension of the reactor. The attached growth processes offer the following advantages over activated sludge processes in wastewater treatment (Guo et al., 2012): 1) better oxygen transfer, high nitrification rate and higher biomass concentrations; 2) more effective in organic removal, and can apply for high organic loading rates at relatively shorter HRT; 3) allowing the development of microorganisms with relatively low specific growth rates (e.g., methanogens); 4) less subject to variable or intermittent loadings; 5) small reactor size and lower space requirement; and 6) lower operational costs (e.g. fixed-bed biofilm processes such as trickling filters and rotating biological contactors).

The attached growth systems can be grouped into two major categories: fixed bed bioreactors and moving bed bioreactors. Table 12 presents the effectiveness of different attached growth processes in micropollutant removal.

Table 12.
Removals of some micropollutants during attached growth treatment processes.

System	Media and experimental conditions	Compound	Removal (%)	References
BAC filter	Media: GAC; media height: 80 cm; diameter: 22.5 cm; EBCT: 18 min	Diclofenac	~ 91	Reungoat et al. (2011)
		Carbamazepine	~ 95	
		Sulfamethoxazole	~ 90	
		Gemfibrozil	~ 90	
SBBGR	Media: wheel shaped plastic elements	Estrone	62.2	Balest et al. (2008)
		Estradiol	68	
		Bisphenol A	91.8	
ASFBBR	Media: K1, AnoxKaldnes; volume: 1.4 L HRT: 4.3 days, 1 day, 0.3 day	Ethinylestradiol	96 (4.3 day)	Forrez et al. (2009)
		Ethinylestradiol	81 (1 day)	
		Ethinylestradiol	74 (0.3 day)	
MBBR	Media: bioplastic-based biofilm carriers; volume: 2.5 L	Bisphenol A	27	Accinelli et al. (2012)[a]
		Atrazine	~ 8	
	Media: K1; volume: 5 L; batch experiments for 24 h	Diclofenac	> 80	Falås et al. (2012)
		Ibuprofen	~ 100	
		Naproxen	~ 100	
		Ketoprofen	~ 100	
		Memfenamic acid	> 80	
		Clofibric acid	> 60	

[a] In this study, only mineralization of the selected compounds was evaluated. Total removal could be higher due to other removal pathways.

Biofiltration seems a compelling biological technique for micropollutant removal (Reungoat et al., 2011). Commonly used systems in water and wastewater treatment include trickling filter, sand filtration and biological activated carbon (BAC). A BAC filter is typically composed of a fixed bed of GAC serving as the carrier for bacterial adhesion and growth. Reungoat et al. (2011) evaluated and compared the performance of biofilters with two media, activated carbon and sand, during long-term operation. The results demonstrated that BAC had a great potential for PPCP (e.g. diclofenac, carbamazepine, sulfamethoxazole and gemfibrozil) removal (> 90%) and reduction of the potential risk of environmental and/or human health impact. On the other hand, sand filters could only achieve limited elimination for PPCPs. Dissolved oxygen was the main factor affecting the performance of BAC filters,

while empty-bed contact time (from 30 min to 120 min) did not result in considerable variation in the removals of compounds. In addition, long-term observation indicated that the main mechanism for organic matter and PPCP removal in biofiltration was biodegradation rather than adsorption. Another biofilter, namely sequencing batch biofilter granular reactor (SBBGR), was investigated by Balest et al. (2008) for removing several selected EDCs. The results showed that SBBGR achieved much higher removal efficiency for EDCs removal than the conventional activated sludge process in a municipal WWTP. The removal efficiencies for bisphenol A, estrone, estradiol and 4-tert-octylphenol were 91.8%, 62.2%, 68% and 77.9% for the demonstrative SBBGR system and 71.3%, 56.4% 36.3% and 64.6% for the conventional activated sludge process of the municipal WWTP, respectively. The excellent performance of the SBBGR was attributed to the very high sludge age (about 160 days). Due to the excellent performance, biofiltration was suggested as an efficient treatment method that could be employed in advanced treatment processes for reducing the impact of the effluent discharge into the environment and/or providing water of higher quality for reuse.

The biological removal of 17α-ethinylestradiol in an aerated submerged fixed bed bioreactor (ASFBBR) was evaluated with or without ammonium starvation (Forrez et al., 2009). Excellent removal (96%) was obtained at a volumetric loading rate of 11 μg/L of 17α-ethinylestradiol, slightly lower elimination rates (81 and 74% respectively) was reported when increasing the loading rate up to 40 and 143 μg/L of 17α-ethinylestradiol. The authors suggested that implementation of retro-fitting treatment systems, either by employing a post-treatment reactor containing separately grown ammonia-oxidizing bacteria or by continuously seeding the WWTP effluent with AOB grown in a dedicated reactor has great potential for the removal of some micropollutants (Forrez et al., 2009). In another study using a fixed film activated sludge (IFAS) to treat effluent estrogenic activities, Kim et al.

(2009a) found the effluent estrogenic activities in the IFAS system were 70% lower than those in the control train (conventional activated sludge system), which suggested a high estrogen removal by IFAS.

Falås et al. (2012) conducted a set of batch experiments to evaluate the effectiveness of a hybrid moving bed biofilm-activated sludge process for the removal of various micropollutants. It was indicated that the presence of carriers could enhance the overall biological elimination of some compounds. For example, diclofenac, clofibric acid and mefenamic acid were not eliminated in the activated sludge reactors, while the carrier reactors showed more obvious and rapid removals (at least 60% after 24 h) of the three compounds. In another study, a moving bed biofilm system was investigated in terms of the removal efficiency for bisphenol A, oseltamivir and atrazine from wastewater using carriers made from existing bioplastic-based products (Accinelli et al., 2012). During the experiments with control wastewater samples, mineralization rates for bisphenol A, oseltamivir and atrazine were relatively low, accounting for only 18%, 7% and 3.5% of the initial concentrations, respectively. By contrast, the addition of incubated carriers enhanced the removals of bisphenol A, oseltamivir and atrazine by 34%, 49% and 66%, respectively. Li et al. (2011)focused their study on simultaneous PAC adsorption within a MBR. During the treatment, PAC could not only act as an adsorbent but also provided support for biomass growth. With a high PAC dosage of 1.0 g/L, enhanced elimination of sulfamethoxazole and carbamazepine was observed in the PAC-amended MBR system (82% and 92% respectively) in comparison with the MBR system alone (both 64%).

As a whole, although attached growth systems have not been applied broadly and specifically to micropollutant removal, the results from some recent bench-scale or pilot scale

studies showed that attached growth treatment processes are promising methods for reducing discharges of micropollutants. By addition of packing/moving carriers, increased microbial community can be maintained in the system, which facilitates the growth of slow-growing microorganisms for micropollutant removal (Serrano et al., 2011). Therefore, micropollutant removal by attached growth processes is a strategy showing possibility of excellence and likely to draw more attention in the future research.

5. Assessment of micropollutant removal from municipal wastewater and recommendations for future research

Micropollutants have been frequently detected in wastewater as well as important drinking water sources, such as rivers, lakes and groundwater. The evaluation of micropollutant removal from municipal sewage should cover a series of issues from sources to end uses, including selection of micropollutants with high occurrence and ecotoxicological relevance, determination of possible sources, investigation on their occurrence and fate in WWTPs and receiving waters, and estimation of their (eco)-toxicological impacts on aquatic systems and humans.

The major types of wastewater media that convey micropollutants to aquatic systems via WWTPs include domestic wastewater, hospital effluents, industrial wastewater and stormwater runoff, rural runoff and manure. Intense efforts have been taken to investigate domestic wastewater, while less focus has been put on other types of wastewaters which may also have significant micropollutant loads. For example, hospitals are a considerable source of various pharmaceuticals, including compounds generated from diagnostic, laboratory and research activities as well as pharmaceutical excretion by patients (Verlicchi et al., 2010b). Kovalova et al. (2012) elucidated that the concentrations of some pharmaceuticals in

the hospital wastewater were considerably different from those in municipal wastewater. For instance, average 32 μg/L of the antibiotic ciprofloxacin and up to 2600 μg/L of iodinated X-ray contrast media were detected in the hospital wastewater, which was around 70-time higher than those observed in the municipal wastewater. In addition, higher concentrations of antibiotics and disinfectants due to large amounts of usage in hospitals could lead to bacterial inhibition during the on-site treatment. Industrial practices (e.g. production of various commodities) can probably lead to a remarkable discharge of micropollutants, especially EDCs, due to the use or/and formation of the compounds during the production processes. The assessment of the significance for different sources can be based on the compilation of literature data (Pal et al., 2010). Scale of consumption or production (e.g. annual per capita consumption) of commodities containing micropollutants can also be used as an indicator for micropollutants emission. Zhang et al. (2008) suggested that the worldwide annual per capita consumption of drugs is 15 g and developed countries contribute three to ten times higher (50–150 g). Hence, it can be expected that the raw sewage from developed countries contains a larger amount of pharmaceutical compounds.

Since WWTPs are not able to provide a complete barrier for micropollutant removal, establishing optimal removal strategies for micropollutants remains a challenge to environmental engineers in order to minimize their adverse effects on the environment. Conventional treatment processes have been reported to have inadequate removals of many micropollutants. Several potential options are available for improving the elimination of micropollutants, including source controls (e.g. application of micropollutant-free products, source separation, pretreatment of hospital and industrial effluents, etc.), reassessment and optimization of current treatment processes, and end-of-pipe upgrading of WWTPs. As mentioned above, the removal of highly persistent/non-biodegradable/polar micropollutants

is commonly low and independent of operating parameters during biological treatment processes, thereby exceeding the capacity of current treatment processes. Hence, tertiary (e.g. post ozonation, sand filtration, and membrane filtration) or combined treatment processes should be taken into consideration to ensure successful treatment of the variety of micropollutants. Table 13 compares the micropollutant removal efficiency of three types of WWTPs, namely low-cost, conventional and advanced WWTPs. Low-cost treatment processes, such as trickling filter beds, lagooning and constructed wetland, are normally used for decentralized wastewater treatment for small communities and in a few cases applied in centralized WWTPs for large communities. As can be seen in Table 13, WWTPs with low-cost treatment processes exhibit inconsistent removal efficiency. Camacho-Muñoz et al. (2012) concluded that most of the pharmaceutical compounds studied were slightly better removed in conventional treatment processes as compared to low-cost treatment processes. This could be attributed to the higher amount of biosolids and better aeration condition (leading to more effective aerobic degradation) in the conventional treatment processes. Nevertheless, the differences between the mean removal efficiency in conventional (64%) and low-cost (55%) WWTPs were not significant. WWTPs with advanced treatments generally show more efficient and consistent removal of the compounds. RO as a tertiary treatment could achieve 100% removal for COD and selected EDCs (Balabanič et al., 2012). Hollender et al. (2009) found ozonation contributed 40–50% (naproxen, benzotriazole, atenolol, clarithromycin), 60–70% (metoprolol, 5-methylbenzotriazole, sulfamethoxazole), and > 80% (diclofenac, carbamazepine, trimethoprim) to the overall removal of the investigated WWTPs.Salgado et al (2012) assessed a full-scale WWTP employing UV as the post-treatment for PPCP removal. They evaluated the relevance of each removal mechanism for the overall PPCP removal and indicated that the removal fractions from biodegradation,

sorption and UV are 45%, 33% and 22% respectively. Although UV only accounted for 22% of the total removal, it was considered as an important effluent polishing process.

Table 13
Comparison of micropollutants removal effectiveness in different WWTPs.

Compounds	Removals (%) in different types of WWTPs		
	Conventional[a]	Low-cost[b]	Advanced[c]
Ibuprofen	72–100	17–> 99	> 99
Diclofenac	< 0–81	0–96	89–100
Ketoprofen	11–100	0–99	69–95
Carbamazepine	< 0–62	0–66	60–100
Estrone	75–91	68–95	84–99
Nonylphenol	22–99	< 50	82–89
Bisphenol A	60–> 99	62–79	90–99

[a] Behera et al. (2011), Céspedes et al. (2008), K. Choi et al. (2008), Gracia-Lor et al. (2012), Janex-Habibi et al. (2009), Kasprzyk-Hordern et al. (2009), Loos et al. (2013), Martin et al. (2010), Nie et al. (2012), Pothitou and Voutsa (2008), Rosal et al. (2010), Santos et al. (2009), Singer et al. (2010), Stamatis and Konstantinou (2013), Stasinakis et al. (2008), Yu and Chu (2009), Zhou et al. (2010) and Zorita et al. (2009).
[b] Cai et al. (2012), Camacho-Muñoz et al. (2012), Hijosa-Valsero et al. (2010), Janex-Habibi et al. (2009), Kasprzyk-Hordern et al. (2009), Matamoros et al. (2009), Melcer and Klecka (2011) and Song et al. (2009).
[c] Hollender et al. (2009), Nakada et al. (2007), Reungoat et al. (2010), Sahar et al. (2011), Sui et al. (2010) and Yang et al. (2011).

Table 14 summarizes the advantages and disadvantages of different treatment techniques reviewed. The provided information is based on the recent literature and may be helpful to select suitable techniques for micropollutants treatment. However, the table only gives the qualitative assessment of these techniques. Comprehensive quantitative assessment is needed in future research to better compare different techniques from both economic and technical points of view.

Table 14
Assessment of different treatment processes for micropollutants removal.

Technique	Common removal efficiency[a]				Major factors		Disadvantage/problems	Residues
	P	PCP	SH	IC	Process-specific	MP-related		
Coagulation	L–M	M–H	L	L–H	● Dosage ● pH ● Wastewater composition	● Hydrophobicity ● Molecular size	● Ineffective MP removal ● Large amount of sludge ● Introduction of coagulant salts in the aqueous phase	Sludge
AC	M–H	M–H	H	M–H	● Adsorbent properties ● Dosage ● Contact time ● pH	● Hydrophobicity ● Molecular size ● Structure ● Functional group	● Relatively high financial costs ● Lower efficiency in the presence of NOMs ● Need for regeneration ● Disposal of used carbon	Used material
Ozonation and AOPs	M–H	M–H	H	M–H	● Dosage ● pH ● Interfering ions (e.g., Br^-) ● Wastewater composition	● Compound structure	● High energy consumption ● Formation of byproducts ● Interference of radical scavengers	Residual oxidants
NF	M–H	H	M–H	M–H	● Membrane properties ● pH ● Transmembrane pressure ● Feed quality	● Hydrophobicity ● Molecular size	● High energy demand ● Membrane fouling ● Disposal of concentrate ● Desorption of sorbed chemicals from membrane	Concentrate
RO	M–H	H	H	H	● Membrane properties ● pH ● Transmembrane pressure ● Feed quality	● Hydrophobicity ● Molecular size	● High energy consumption ● Disposal of concentrate ● Corrosive nature of the finished water	Concentrate
Activated sludge	L–H	M–H	M–H	L–H	● SRT ● HRT ● Organic loading ● Redox conditions	● Hydrophobicity ● Biodegradability	● Inconsistent removal of polar and resistant compounds ● Increase of environmental risk due to the disposal of sludge containing micropollutants	Wasted sludge

Technique	Common removal efficiency[a]				Major factors		Disadvantage/problems	Residues
	P	PCP	SH	IC	Process-specific	MP-related		
MBR	L–H	M–H	H	M–H	● SRT ● HRT ● Organic load ● Redox conditions	● Hydrophobicity ● Biodegradability	● Moderately high energy consumption ● Inconsistent removal of polar and resistant compounds ● Membrane fouling ● Less sorption of micropollutants on the aged MBR sludge	Wasted sludge
Attached growth	L–H	M–H	M–H	M–H	● HRT ● Organic loading ● Redox conditions	● Hydrophobicity ● Biodegradability	● Long start-up time ● Difficulty in control of biofilm thickness	Wasted sludge

[a] P: pharmaceutical; SH: steroid hormone; IC: industrial chemical; L: low; M: medium; H: high.

Understanding and predicting the fate of micropollutants in WWTPs is helpful in identifying the potential for improvement of current treatment configurations. To date, enormous efforts by many researchers have been put into developing accurate and succinct models for micropollutant prediction. Precise models for micropollutant fate are not easy to establish. Modelers should take into account numerous aspects, including possible removal pathways and factors that affect the removal. Pomiès et al. (2013) reviewed different models from the perspective of removal pathways. Sorption and volatilization can be characterized by partition coefficient K_d and Henry's law constant, both of which can be determined experimentally. Biodegradation modeling is a more complicated process due to the involvement of microorganisms. Two issues have been addressed for the biodegradation of micropollutants. First issue is the lack of conformity in determining biodegradation sites (only in aqueous phase, only in solid phase or in both phases). The other is the incorporation of parent compounds and by-products as well as co-metabolism in the models.

The discharge of micropollutants can contribute to water pollution due to their potentially ecotoxicological impacts on aquatic organisms. Furthermore, human exposure to micropollutants is also harmful and can occur via various routes. Micropollutants can return to humans via drinking water. Other exposure pathways to humans include food chain and wastewater reuse for household purposes. Given their adverse effects, effective monitoring strategies and risk assessment should be considered as important components for micropollutants control. Nevertheless, monitoring programs for micropollutants are far from universal and have only been carried out in sizable rivers, such as Rhine (Sacher et al., 2008) and Han River (K. Choi et al., 2008), as those programs are time consuming and costly (Alder et al., 2010). Therefore, the establishment of estimation tools for the concentrations and mass flows of micropollutants in surface waters is of vital importance. Generally, the estimation should be based on the various sources, use/consumption of compounds and their fate in WWTPs as well as receiving waters. Coetsier et al. (2009)indicated that predicted environmental concentrations (PECs) offer the possibility to predict pharmaceutical occurrence in surface water. Although the PEC values seemed to be able to properly estimate WWTP wastewater effluents, they are subjected to uncertainties because the differences between predicted and measured values can become significant when applied to local areas with consumption levels being considerably different from assumed average levels.

After having been discharged into surface waters, micropollutants experience various processes, including dilution and attenuation (biodegradation, sorption, volatilization and photolysis). A comprehensive understanding and modeling of micropollutant fate in surface waters are essential for effectively predicting micropollutants' impacts on the receiving environment. Although integrated urban water system (IUWS) modeling is usually used as a tool for evaluating the quality of the surface water receiving the municipal WWTP discharge

combining sewer overflows and stormwater drainage systems, many micropollutants tend to distribute to more than one environmental compartment (air, water, sediment, soil, groundwater, etc.). Hence, a multimedia fate and transport model (MFTM) was proposed by De Keyser et al. (2010) to meaningfully characterize the attenuation and distribution of micropollutants.

6. Conclusion

Enormous research effort has been directed toward the assessment of occurrence of micropollutants in the aquatic environment. In particular cases, the occurrence levels of some micropollutants in surface waters were much higher than their PNECs, which revealed an environmental concern. WWTP effluent has been considered as the primary source of many micropollutants in aquatic systems. Given their diverse properties (e.g., hydrophobicity and biodegradability) and low concentrations, micropollutant removal in current WWTPs is commonly incomplete and variable, ranging from 12.5% to 100% for some frequently reported compounds. Hence, optimization of wastewater treatment, in order to create an absolute barrier to micropollutants emission, remains a task of high priority. Biological treatment is commonly unable to remove polar persistent micropollutants. However, its efficacy can be improved under favorable conditions (e.g., extended SRT and HRT, warm temperature, and fine tuning redox conditions). Although advanced treatment technologies, such as adsorption processes, AOPs and membrane processes, have been demonstrated to be promising alternatives for micropollutant removal, there are two issues associated with the applications of advanced treatments: high operation costs and formation of by-products and concentrated residues. Moreover, to effectively predict the impact of micropollutants on the receiving environment, a comprehensive understanding and modeling of micropollutants fate is needed.

Conflict of interest

On behalf of all the co-authors, I declare that: (1) we have no financial and personal relationships with other people or organizations that can inappropriately influence our work; and (2) there is no professional or other personal interest of any nature or kind in any product, service and/or company that could be construed as influencing the position presented in, or the review of, the manuscript entitled “A review on the occurrence of micropollutants and their fate and removal during wastewater treatment”.

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