Chapter 1
Occurrence and Health Impacts of Emerging Contaminants in Municipal Wastewater Reuse

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Abstract Municipal wastewater reuse offers the potential to significantly increase the total available water resources. Recently, the occurrence of emerging contaminants (ECs), like pharmaceuticals and personal care products (PPCPs) and perfluorinated compounds (PFCs), in water resources is of continued concern for the public health and safety. However, the existing conventional wastewater treatment plants (WWTPs) were not originally conceived to eliminate these unidentified contaminants, which have not been monitored routinely because of the absence of stringent-specific regulation. This chapter focuses on the occurrence of these ECs and feasible opportunities for guidelines in municipal wastewater reclamation and reuse. An environmental risk assessment posed by a common means of the risk quotient shows that 27 pharmaceuticals pose high or medium risk. The concept of source control and source separation could reduce the manufacture and produce a wastewater with an optimal composition for further centralized treatment. Additional and integrated technologies for synergic treatment units are found necessary to provide high-quality recycled water and sustainable water resources.

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1.1 Significance and Importance

Water resource scarcity could be the major challenge of civilization, because of the unbalanced distribution of resources, environmental pollutants, climate change, population growth, rapid urbanization, and so on. In this sense, the concept of water resources sustainability has created increasing interest in water reuse and recycling. Water resources sustainability was defined in the report of the Brundtland Commission (World Commission on Environment and Development) as (Holdgate 1987):

> Water resources sustainability is the ability to use water in sufficient quantities and quality from the local to the global scale to meet the present needs of humans and environmental ecosystems, while not impairing the needs of future generations to do the same.

It involves the aspects of freshwater allocation, urban water use cycle, wastewater reuse, and risk management. Among various concepts, municipal wastewater reuse is quite promising in densely inhabited district, offering irrigation, landscape, recreation, humidification, cooling, and surface or groundwater replenishment, due to mechanical treatment for suspended solids and conventional organic pollutants in the mature chemical and biological process of municipal wastewater treatment plants (WWTPs) (Valipour 2014).

1.1.1 Occurrence of Emerging Contaminants

Emerging contaminants as new products or chemicals without regulatory status in sewage and aquatic environments could cause known or suspected adverse ecological and/or human health effects (high toxicity, carcinogenic and mutagenic effects) (Ratola et al. 2012). Two important chemical constituents, including pharmaceuticals and personal care products (PPCPs) and perfluorinated compounds (PFCs), have attracted global concern due to their ubiquitous occurrence in the multimedia, biota, and humans. WWTPs as "initial biological treatments" were inefficient in the removal of PPCPs and PFCs (Stamatis and Konstantinou 2013; Chen et al. 2017a). PPCPs have been entering into the environment for over 25 years from individual human and social activities (e.g., waste excretion, bathing, residues of manufacturing, agribusiness, and hospitals). About 95% of the total amount of PPCP consumption are:

Analgesics: 5-aminosalicylic acid, acetaminophen, acetylsalicylic acid, codeine, dextropropoxyphene, diclofenac, dipyrone, fenoprofen, flurbiprofen, hydrocodone, ibuprofen, indomethacin, ketoprofen, mefenamic acid, naproxen, propyphenazone, salicylic acid, tramadol

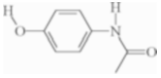
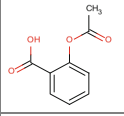
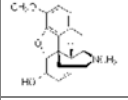
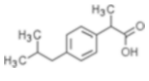
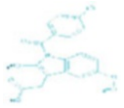
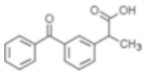
Antibiotics: amoxicillin, azithromycin, cefaclor, cefalexin, cefotaxime, chlortetracycline, ciprofloxacin, clarithromycin, doxycycline, erythromycin, lincomycin, norfloxacin, ofloxacin, roxithromycin, sulfamethoxazole, trimethoprim
Antidiabetics: glibenclamide
Antihypertensives: diltiazem, hydrochlorothiazide
Barbiturates: phenobarbital
Beta-blocker: atenolol, metoprolol, propranolol, sotalol
Lipid regulators: bezafibrate, gemfibrozil
Psychiatric drugs: carbamazepine, fluoxetine
Receptor antagonists: ranitidine, valsartan
Hormones: estradiol, estriol, estrone
Antineoplastics: ifosfamide, tamoxifen
Antiseptic: triclosan

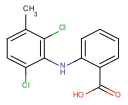
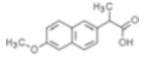
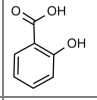
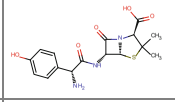
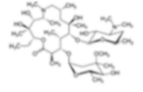
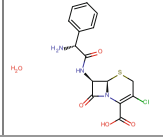
It is difficult to assess typical concentration levels of PPCPs as monitoring data of PPCPs varies with the time, the type of waste water, and geographical areas. Table 1.1 presents the molecular structure and the variability ranges for the concentrations of common compounds grouped with their therapeutic class in municipal WWTP influent based on the previous scientific literature data, which reflects common pharmaceuticals (analgesics/anti-inflammatories, antibiotics, antihypertensives, beta-blockers, lipid regulators, psychiatric drugs, receptor antagonists, and contrast agent) could be detected in wastewater that receives each municipal WWTP. The differences among these results could also be related with the types of products in different geographic markets or the possibility in some countries with limited supervision to purchase medicines without medical prescriptions.

PFCs that are a class of anthropogenic compounds with repellency of water and oil have been commonly used in industrial and household products for more than 60 years (Fujii et al. 2007). There is also a long-term public concern due to their being persistent and accumulative and biotoxicity (Giesy and Kannan 2001). For the most environmental concerning and frequently detected predominant compounds, the concentration levels determined for perfluorooctane sulfonate (PFOS) and perfluorooctanoic acid (PFOA) in municipal influent and secondary treated wastewater are presented in Table 1.2. They are ubiquitously detected in influent wastewater and secondary treated wastewater due to the high bond energy of C–F long chains and their high-water solubility made by hydrophilic functional groups (such as sulfonate and carboxyl).

Besides, the presence of PPCPs and PFCs as environmental pollutants was reported in the Antarctic environment and Antarctic biota with concentrations similar to those detected in urban living areas throughout the world (Corsolini 2009). The concerning issue with PPCPs and PFCs is not their acute toxic effects but their chronic toxicity due to their ubiquitously distributed and large quantities in both consumer and industrial settings.

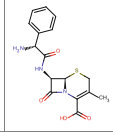
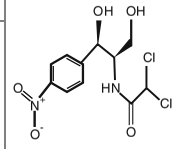
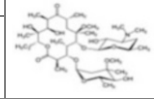
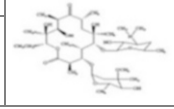
Table 1.1 Ranges of concentrations and molecular structure in the influent for the common PPCPs

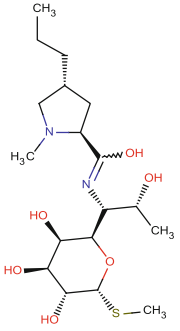
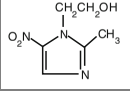
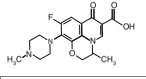
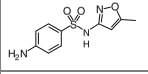
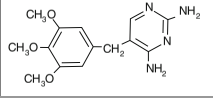
Type of PPCPs	Compound	Influent (μg/L)	Molecular structure	References
Analgesics/anti-inflammatories	Acetaminophen	18–71		Verlicchi et al. (2012)
		1.57–37.5		Rosal et al. (2010)
		7.1–11.4		Radjenovic et al. (2009)
		29–246		Gomez et al. (2007)
		104		Rosal et al. (2010)
	Acetylsalicylic acid	1.32–5.44		Kasprzyk-Hordern et al. (2009)
	Codeine	2.8–11		Gomez et al. (2007)
		2.49–12.6		Kasprzyk-Hordern et al. (2009)
		0.15–2.09		Rosal et al. (2010)
	Ibuprofen	2.6–5.7		Carballa et al. (2004)
		34–168		Gomez et al. (2007)
		9.8–19.8		Lindqvist et al. (2005)
		14.6–31.3		Radjenovic et al. (2009)
	Indomethacin	0.95		Stumpf et al. (1999)
		0.66–1		Radjenovic et al. (2009)
	Ketoprofen	1.3–3		Lindqvist et al. (2005)
		0.15–0.41		Tauxe-Wuersch et al. (2005)
		0.41–0.52		Thomas and Foster (2005)

	Mefenamic acid	0.8–1.2		Radjenovic et al. (2009)
		0.75–2.9		Tauxe-Wuersch et al. (2005)
	Naproxen	1.79–4.6		Carballa et al. (2004)
		0.62–3.5		Kasprzyk-Hordern et al. (2009)
		3.5–4.5		Rodriguez et al. (2003)
	Propyphenazone	0.04–0.09		Radjenovic et al. (2009)
	Salicylic acid	5.6–32.08		Kasprzyk-Hordern et al. (2009)
Antibiotics	Amoxicillin	0.19–0.28		Watkinson et al. (2007)
	Azithromycin	0.16–1.34		Ghosh et al. (2009)
	Cefaclor	0.5–0.98		Watkinson et al. (2007)

(continued)

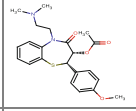
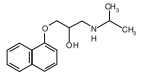
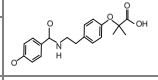
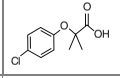
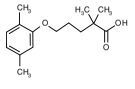
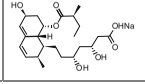
Table 1.1 (continued)

Type of PPCPs	Compound	Influent (μg/L)	Molecular structure	References
	Cefalexin	0.67–2.9		Gulkowska et al. (2008)
	Chloramphenicol	0.15–0.45		Kasprzyk-Hordern et al. (2009)
		1.73–2.43		Peng et al. (2006)
	Clarithromycin	0.33–0.6		Gobel et al. (2005)
		1.13–4.82		Ghosh et al. (2009)
	Erythromycin	0.47–0.74		Gulkowska et al. (2008)
		0.48–1.2		Karthikeyan and Meyer (2006)

	Lincomycin	0.06–0.08		Watkinson et al. (2007)
	Metronidazole	0.34–0.96		Kasprzyk-Hordern et al. (2009)
	Ofloxacin	0.52–5.56		Peng et al. (2006)
		0.89–31.7		Radjenovic et al. (2009)
	Sulfamethoxazole	0.23–0.57		Gobel et al. (2005)
		0.17–1.25		Karthikeyan and Meyer (2006)
	Trimethoprim	0.21–0.44		Gobel et al. (2005)
		0.15–0.43		Radjenovic et al. (2009)

(continued)

Table 1.1 (continued)

Type of PPCPs	Compound	Influent (μg/L)	Molecular structure	References
Antihypertensives	Diltiazem	0.41–5.26		Kasprzyk-Hordern et al. (2009)
Beta-blockers	Propranolol	0.11–1.9		Kasprzyk-Hordern et al. (2009)
Lipid regulators	Bezafibrate	1.9–29.8		Radjenovic et al. (2009)
		1.55–7.6		Clara et al. (2005)
	Clofibric acid	0.17–0.37		Tauxe-Wuersch et al. (2005)
	Gemfibrozil	0.6–1.1		Verlicchi et al. (2012)
		0.41–17.1		Rosal et al. (2010)
	Pravastatin	0.46–1.5		Radjenovic et al. (2009)

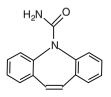
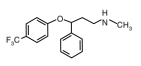
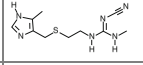
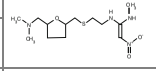
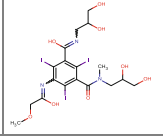
Psychiatric drugs	Carbamazepine	0.32–1.2 0.1–3.11		Clara et al. (2005) Kasprzyk-Hordern et al. (2009)
	Fluoxetine	0.12–2.3		Radjenovic et al. (2009)
Receptor antagonists	Cimetidine	0.68–6.5		Kasprzyk-Hordern et al. (2009)
	Ranitidine	0.072–0.54 2–11.15		Radjenovic et al. (2009) Kasprzyk-Hordern et al. (2009)
Contrast agent	Iopromide	0.03–3.84		Clara et al. (2005)

Table 1.2 Concentration of common PFCs in the influent and secondary effluent of WWTPs

Location	Influent wastewater (ng/L)		Secondary treated wastewater (ng/L)		References
	PFOS	PFOA	PFOS	PFOA	
USA	1.4–400.0	1.7–49.0	1.1–130.0	2.5–97.0	Schultz et al. (2006)
Kentucky, USA	7.0–16.0	22.0–184.0	8.0–28.0	122.0–183.0	Loganathan et al. (2007)
Georgia, USA	2.5–7.9	2.0–50.0	1.8–13.0	6.7–102.0	Loganathan et al. (2007)
Germany	1.0–85.0	1.8–40.0	12.0–140.0	8.7–9.3	Becker et al. (2008)
Denmark	1.5–10.1	2.0–23.5	1.5–18.1	2.0–24.4	Bossi et al. (2008)
Switzerland	18.0–449.0	4.9–35.0	16.0–303.0	8.9–35.0	Huset et al. (2008)
Greece	2.4–26.3	10.2–20.7	5.2–21.0	12.7–34.0	Arvaniti et al. (2012)
Spain	78.1	22.4	91.0	14.9	Campo et al. (2014)
Japan	14.0–336.0	14.0–41.0	42.0–635.0	10.0–68.0	Murakami et al. (2009)
Singapore	7.9–374.5	14.1–638.2	7.3–461.7	15.8–1057.1	Yu et al. (2009a)
Korea	n.d.–68.1	4.3–615.0	n.d.–5.7	6.4–591.0	Guo et al. (2010)
Taiwan	175–216.7	17.6–23.6	162.7–5663.3	19.3–480.3	Lin et al. (2010)
China	0.03–12	0.05–54.0	0.03–7.3	0.09–26.2	Pan et al. (2011)
Thailand	381.3	6.6	552.8	16.9	Kunacheva et al. (2011)
Australia	/	/	23.0–38.6	15.0–27.0	Thompson et al. (2011)

1.1.2 Municipal Wastewater Reuse

Global water shortage is placing an unprecedented pressure due to the inferior quality and insufficient quantity of water resource. Many regions, including the Middle East, Northern Africa, parts of China and India, and the southwest quarter of the USA, are experiencing high levels of water stress (with annual per capita water supplies below 1700 m^3). Projections predict that by 2025, 2/3 of the world's population will be living under the conditions of moderate to high water stress (Wu et al. 2014). Nowadays, the common municipal WWTPs consist of physical preliminary, primary treatment and secondary biochemical system with the treated sewage being discharged into natural water or reused in many aspects. The application of municipal sewage (treated to remove pathogens, organic matter, and nutrients) represents an additional opportunity in augmenting freshwater supply for irrigation, landscape, recreation, humidification, cooling, and surface or groundwater replenishment, especially in semiarid and arid regions. Unlike many organic contaminants, the existing WWTPs are ineffective at removing emerging contaminants. The potential risks of

the reuse of treated municipal wastewater associated with emerging contaminants, especially PPCPs and PFCs, are new issues garnering public attention.

Surface or groundwater replenishment: Reclamation of water after treatment in modern WWTPs will likely be an important yet currently unrealized part of sustainable water resource management. Most treated municipal wastewater discharged into the receiving aquatic environment shows a relatively reliable water source, but it can convey emerging contaminants to cities and drinking water intakes located downstream. WWTP discharges are also the main source of PPCPs and PFCs in surface water environment. The surface ocean was assumed to be the main reservoir for PFCs. According to the difference in bioconcentration in natural aquatic background or different profiles, fish and other seafood seem to be the food group in which more PFCs are detected and where the detectable concentrations of typical compounds are higher. The levels of PFCs in the analyzed foodstuffs decreased in the order seafood > animal liver > freshwater/marine fish > egg > meat > butter in the samples of five European countries (Belgium, Czech Republic, Italy, and Norway) (D'Hollander et al. 2010). This means that individuals consuming great amounts of seafood are assuming certain risks in certain countries, which are not currently quantified.

Irrigation: Up to 49–90% of consumptive water is used in agriculture irrigation; municipal wastewater reuse appears to be a valuable water resource to supplement agriculture irrigation (Wu et al. 2014). The most challenging aspect of this application is the safety attention of produce due to the residual contaminants. PPCPs and PFCs persist in both aqueous effluent and treated biosolids. Irrigation land application of secondary treated wastewater and biosolids through root uptake on crops can facilitate the PPCPs and PFCs into the terrestrial food web, especially for fresh produce that may be consumed raw (e.g., vegetables, fresh fruits) (Blaine et al. 2014; Krippner et al. 2015).

Humidification: Reclaimed municipal wastewater began to be used for sprinkling roads to mitigate heat island and reduce road dust in urban areas. The results of measured temperatures of air and road with/without sprinkling show that sprinkling reclaimed wastewater lowered the road surface temperature by eight degrees during the daytime in Japan (Ogoshi et al. 2001). Inhalation is one of the most important routes of transmission of inflammatory agents, and it is noteworthy that the evaporation process can affect human health directly through inhalation during this application (Xue et al. 2016).

1.1.3 Exposure Routes of PPCPs and PFCs

As emerging contaminants, there are multiple potential input pathways for PPCPs and PFCs into the environment as shown schematically in Fig. 1.1.

Industrial wastewater has been regarded as a main point source for PPCPs and PFCs as well as their precursors entering into the receiving water bodies (Arvaniti and Stasinakis 2015). Besides, other sources of PPCPs and PFCs intake by humans

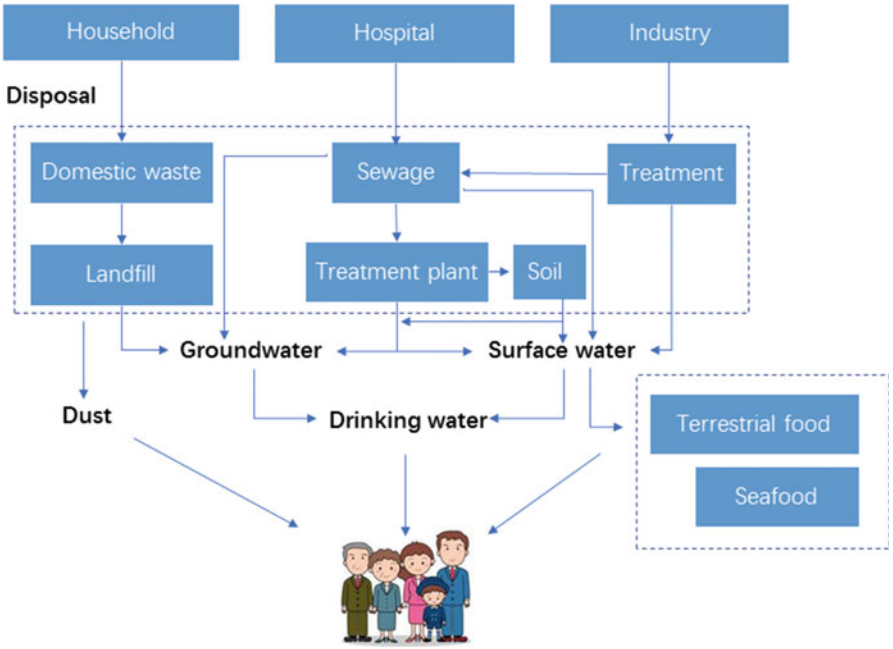


Fig. 1.1 Exposure routes of emerging PPCPs and PFCs for humans

could be through consumption of contaminated food and inhalation of air or fine particles contaminated with volatile PPCPs and PFCs precursor that would be degraded to become PFOS, PFOA, or other intermediates. Unlike the PFCs, parts of the PPCPs are usually disposed as solid waste or in wastewater effluent by improper disposal from private households and hospitals. Most hospital sewers are directly connected to the municipal drainage system, and no additional treatment is performed. Sorption of lipophilic PPCPs and hydrophobic PFCs onto sewage sludge may be a relevant removal pathway from the wastewater stream. Partial PPCPs can be assumed to be destroyed by chemical oxidation in sludge incineration process, while PFCs could be retained due to their highly polar and strong carbon–fluorine bonds. When sewage sludge is used as a fertilizer, PPCPs and PFCs may return to the surrounding aquatic environment through the surface runoff (De Sanctis et al. 2017).

WWTPs are generally not equipped to deal with complex PPCPs and PFCs, and irrigation of treated wastewater on arable land can also lead to a pollution of groundwater. As a result, effective environmental risk assessment and tertiary treatment technologies of upgrading WWTPs should be implemented in order to ensure the safe usage of municipal wastewater.

1.2 Environmental Risk Assessment

Risk assessment is a key component of many environmental regulatory decisions, which provides an evaluation of the probability and impact of detrimental effects attributable to the potential toxicity of contaminants (Lammerding and Fazil 2000). Figure 1.2 shows the general principles of the environmental risk assessment, which are accepted extensively, including five steps (hazard identification, exposure assessment, effects assessment, risk characterization, and risk management).

It is noted in the effect assessment that the risk for humans from environmental contaminants is not discussed in this step but rather the risk for organisms in the environment caused by anthropogenic use of these compounds. Besides, the possible long-term, low-level effects of PPCPs and PFCs should not be overlooked, which can be helpful to test the standard scheme of environmental risk assessment with "familiar contaminants" and to supplement potentially hazardous characterization.

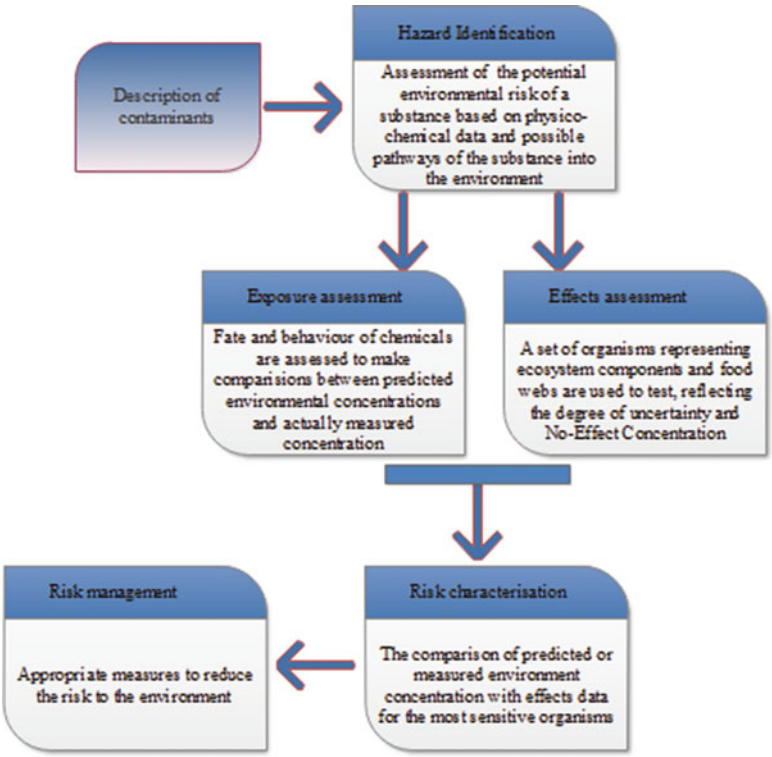


Fig. 1.2 Steps of emerging contaminants' environmental risk assessment

1.2.1 Predicted Environmental Concentrations (PEC) of PPCPs

A program for the risk assessment of PPCPs has been outlined by the European Medicines Agency (EMA). There are several models for PEC prediction in different draft versions of the EMA, which can provide options for prioritizing species on which to focus the concentrated research efforts (Liebig et al. 2005). The EMA model for initial exposure assessment presented in 2001 is based on the predicted, prescribed, or used volumes of pharmaceuticals in Eq. (1.1).

$$\mathrm{PEC_{SW}[g/L]} = \frac{A \times (100 - R)}{365 \times P \times V \times D \times 100} \tag{1.1}$$

where A (kg) is the predicted use/prescription amount per year, R (%) is the removal rate, P is the number of inhabitants of the specific geographic area, V (m^3) is the volume of wastewater per capita and day, and D is the factor for dilution of wastewater by surface water flow.

The second EMA model for initial PEC assessment is based on the maximum daily dose of the active ingredient of a pharmaceutical ($DOSE_{ai}$) in Eq. (1.2). A factor for the market penetration F_{pen} represents the proportion of the population being treated daily with a specific drug.

$$\mathrm{PEC_{SW}[mg/L]} = \frac{\mathrm{DOSE_{ai}} \times F_{pen}}{V \times D \times 100} \tag{1.2}$$

where the defined daily dose (DDD) referred to the World Health Organization Collaborating Centre for Drug Statistics Methodology was defined as $DOSE_{ai}$ (mg) and the recommended default value of 1% was defined as F_{pen}. For V and D, the same default values as in Eq. (1.1) were used; hence the $DOSE_{ai}$ (DDD) was the only variable in this formula which results in a direct correlation of the PEC_{sw} to the DDD.

1.2.2 Environmental Risk Assessment of PPCPs in Secondary Biological Effluent

The environmental risk assessment with the presence of PPCPs in natural water should be considered after predicting the environmental concentration. Although the safety threshold values have been defined for a limited number of PPCPs, more research need to be further strengthened for many compounds themselves and their mixture toxicity, because of the previous studies only in single compound-single organism toxicity. Table 1.3 presents the predicted no-effect concentrations (PNECs) and the average concentrations of predominant PPCPs for fish in the

Table 1.3 PNECs and average concentrations of predominant PPCPs for fish in the secondary effluent (Sanderson et al. 2003; Stuerlauridsen et al. 2000; Jones et al. 2002; Lee et al. 2008; Ferrari et al. 2004; Hallingsorensen 2000; Kim et al. 2007)

Compounds	Toxicity (mg/004C)	Average secondary effluent concentration (μg/L)	PNEC (μg/L)
Acetaminophen	1	0.89	1
Acetylsalicylic acid	796	0.36	61
Codeine	238	1.7	16
Dextropropoxyphene	13	0.10	1
Ibuprofen	5	3.6	1.65
Indomethacin	3.9	0.47	3.9
Ketoprofen	32	0.21	15.6
Mefenamic acid	0.43	0.63	0.43
Naproxen	34	1.0	2.62
Phenazone	3	0.16	1.1
Propyphenazone	0.8	0.04	0.8
Salicylic acid	1.28	0.17	1.28
Amoxicillin	0.1	0.01	0.0037
Azithromycin	0.15	0.16	0.15
Cefaclor	11,524	0.01	687.42
Cefalexin	2.5	0.13	2.5
Cefotaxime	0.04	0.02	0.04
Chloramphenicol	1.6	0.05	1.6
Ciprofloxacin	246,000	0.86	938
Clarithromycin	20	0.29	0.07
Clindamycin	0.5	0.01	0.5
Doxycycline	0.3	0.04	0.3
Erythromycin	61	0.73	0.02
Lincomycin	1391	0.06	82
Metronidazole	2.5	0.25	2.5
Ofloxacin	10	0.45	0.016
Oxytetracycline	16,600	0.01	0.207
Roxithromycin	50	0.50	4
Sulfadimethoxine	3.5	0.09	3.5
Sulfamethoxazole	890	0.28	0.027
Trimethoprim	795	0.36	2.6
Diltiazem	23	0.12	1.9
Metoprolol	116	0.32	8
Propranolol	29.5	0.17	0.244
Bezafibrate	5.3	0.90	5.3
Clofibric acid	53	0.21	40.2
Fenofibric acid	7.6	11	7.6
Gemfibrozil	0.9	0.93	0.9
Pravastatin	1.8	0.02	1.8
Carbamazepine	101	1.04	13.8

(continued)

Table 1.3 (continued)

Compounds	Toxicity (mg/004C)	Average secondary effluent concentration (μg/L)	PNEC (μg/L)
Diazepam	28	9.1	2
Fluoxetine	11	0.24	0.05
Cimetidine	571	3.5	35
Ranitidine	1076	0.51	63
Ifosfamide	140	0.97	11
Iopromide	86,500	2.5	370,000

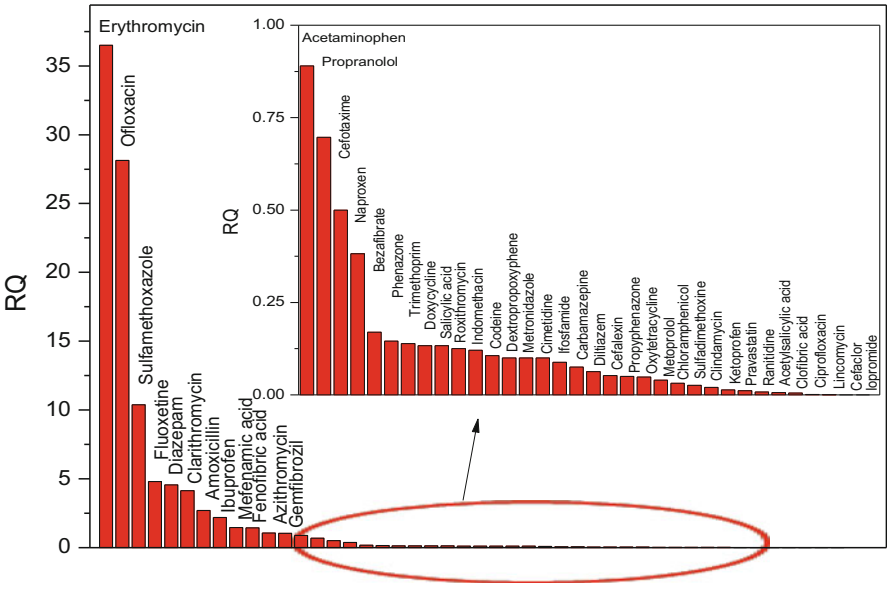


Fig. 1.3 Risk quotient (RQ) of the predominant PPCPs

secondary effluent. A common means of the risk quotient (RQ), the ratio between the average PPCPs concentration measured and its corresponding PNECs, can be used to evaluate the environmental risk by PPCPs: RQ < 0.1, low risk to aquatic organisms; $0.1 \leq RQ \leq 1$, medium risk; and RQ > 1, high risk (Caldwell et al. 2012).

The RQ of the predominant PPCPs in descending order is reported in Fig. 1.3. Several PPCPs have been found in high levels (27 compounds pose high or medium risk), thereby evidencing the risk that environmental concentrations of PPCPs can be higher than their PNECs, especially in effluent-dominant rivers whose dilution capacity and self-purifying processes are insufficient to temper the risk to aquatic life.

1.3 PPCPs and PFCs Removal from Wastewater

The initial purpose of treatment plants of municipal wastewater was conceived to reduce the organic load into aquatic environment. WWTPs are ineffective to removal PPCPs and PFCs due to their recalcitrant characteristic. Centralized WWTPs are considered as important micropollutant point sources of PPCPs and PFCs for the receiving aquatic environment, which provide the potential of implementing centralized removal processes within urban drainage systems for facilitating the municipal wastewater reuse (Wang and Wang 2016; Arvaniti and Stasinakis 2015).

1.3.1 Removal of PPCPs

Removal technologies of PPCPs can be mainly composed of physical, biological, and chemical methods. It is noteworthy that the different categories may be involved in the same removal process, such as sorption, biodegradation, stripping, and photodegradation, which would be included in the aerobiotic/anaerobic activated sludge processes. Besides, advanced oxidation processes (AOPs) (or combination with other technologies) have also been widely tested against recalcitrant contaminants.

1.3.1.1 Removal of PPCPs by Physical Sorption

Carbonaceous materials (e.g., activated carbon, graphene and graphene oxide, and carbon nanotubes) as suitable adsorbents have been widely applied for removing trace (pseudo)-persistent PPCPs. These carbonaceous adsorbents have high sorption capacity for the typical PPCPs, which mainly depend on the hydrophobicity and charge of adsorbates. Many factors such as organic matter existing, contact time, solution pH, and structure of carbonaceous adsorbents have remarkable effects on the removal efficiency (Mailler et al. 2015; Meinel et al. 2014; Kyzas et al. 2015; Liu et al. 2014; Jung et al. 2015; Wan and Wang 2016). Notwithstanding, many problems should be solved for the large-scale applications.

Due to the competing sorption and steric effect, the deterioration of these porous adsorbents may occur with the increase of operation time in the complex wastewater systems.

More efforts should be focused on producing the high-surface-area graphene and nanotubes with relative low cost.

The toxic effect (nanotoxicity) of the coexistence of graphene/nanotubes and PPCPs should be investigated and prevented.

1.3.1.2 Removal of PPCPs by Biological Degradation and Chemical Advanced Oxidation

Microbial degradation is the most prevalent process to remove organic contaminants in the environment. Activated sludge treatment has been regarded as the effective technology to degrade PPCPs easily, which contains combined effects of volatilization, sorption, and biodegradation. Compared with the contribution of biodegradation, the volatilization with aeration and sorption by activated sludge play only a small part in the whole process. However, the concentrations of PPCPs are low; biodegradation would not be always effective for degrading them, which is caused by the low abundance (or lack) of degraders and the contaminant load (Gobel et al. 2005; Suarez et al. 2010; Li et al. 2015). Some measures, such as biological acclimation and bioaugmentation, can conquer the above limitations.

Based on the previous studies on the PPCPs concentration and contamination profiles, different compounds of PPCPs have been frequently tested in the secondary effluent of WWTPs, indicating that the PPCPs would not be removed completely in the conventional WWTPs. Chemical advanced oxidation technologies, such as ozonation (estrone (>95%), sulfamethoxazole (>99%), bezafibrate (>89%), trimethoprim (100%)) (Lin et al. 2009; Garoma et al. 2010; Tootchi et al. 2013; Kuang et al. 2013), UV treatment (estrone (>60%), sulfamethoxazole (100%), bezafibrate (>90%), trimethoprim (>99%)) (Kim et al. 2009; Ma et al. 2015; Wols et al. 2015), Fenton or Fenton-like oxidation (estrone (>98.4%), sulfamethoxazole (100%), bezafibrate (100%), trimethoprim (100%)) (Dirany et al. 2010; Feng et al. 2005; Ternes et al. 2002), and ionizing irradiation (Chu et al. 2015), were effective for the PPCPs. However, the toxic and resistant intermediates appeared during the operating process are another issue in order to complete mineralization. Therefore, the combination of advanced chemical processes and biological technologies as tertiary treatment can provide satisfactory performance.

1.3.2 Removal of PFCs

As two most typical PFCs, both PFOS and PFOA have been included in the forbidden/limited list or the relevant regulations. Some regions and organizations still have expressly restricted the use of PFCs due to the lag in regulation and lack of effective substitute. Unlike PPCPs, biological degradation is effective in removing PFCs attributed to the high-energy carbon–fluorine (C–F) bonds (110 kcal/mol) (Rahman et al. 2014). Therefore, effective remediation technologies should be developed continually.

1.3.2.1 Sorption and Sorption Mechanisms

Sorption is considered as an effective and economical method to remove PFCs from wastewater. Besides conventional adsorbents, many synthetic adsorbents have been applied to remove PFCs. Table 1.4 summarizes the sorption of PFOS and PFOA onto different adsorbents reported in the literature.

Nonpolar hydrophobic interaction between the adsorbents and PFCs existed during the sorption processes is the main affinity, because the C–F chains in PFCs molecule exhibit hydrophobic properties. Self-assembly of hemi-micelle and micelle has been proved to be present in the sorption process, which can block the inner pores of porous adsorbents. In addition, there is a negatively charged shell around a positively charged core of the PFCs molecules due to the high electronegativity of fluorine atoms, and weak interactions may be generated between negative dipole and charged adsorbent surface. Since most PFCs are anionic species by their low pK_a, the sorption processes, such as electrostatic attraction and ion exchange column, can be involved in the treatment. The tails of PFCs molecule as paired groups can replace the hydroxyl groups on aluminum oxides by ligand exchange, as described in Eq. (1.3) (Wang et al. 2012):

$$\mathrm{Al-OH} + \mathrm{CF_3(CF_2)_nCOO^-} \rightarrow \mathrm{Al-OOC(CF_2)_nCF_3} + \mathrm{OH^-} \tag{1.3}$$

1.3.2.2 Filtration Processes

Membrane filtration is a widely used technology in wastewater and drinking water treatment due to its high efficiency in the reduction of wide range of contaminants.

Commercially available reverse osmosis membranes are effective to remove PFOS in a wide range of inlet concentrations from 0.5 to 1600 ppm. Rejection is not affected by membrane zeta potential, but membranes flux would decrease with the increase of PFOS concentration. Nanofiltration membrane such as NF270 has a higher rejection (>95%) to PFCs than microfiltration and ultrafiltration membranes, as well as the higher water flux than reverse osmosis (Zhao et al. 2016). Increased calcium chloride concentrations also can improve PFCs rejection due to increasing permeation of the smaller anions, as well as pore blockage and calcium-bridging between PFCs and calcium ions (Zhao et al. 2013).Sorption, coagulation, and other technologies as a pretreatment can be chosen to enhance the PFCs permeate flux rejection and prevent severe membrane fouling. Multistage filtration membrane arrays can be conceived to further increase the removal efficiency.

1.3.2.3 Advanced Treatment Technologies

Advanced oxidation technologies such as Fenton and Fenton-like oxidation, UV/H_2O_2, sonolysis, as well as photolysis have been widely applied in recalcitrant

Table 1.4 Sorption capacities of PFOS and PFOA on different adsorbents

Adsorbents	Adsorbates	C_0 (mg/g)	pH	q_m (mg/g)	References
Powdered activated carbon	PFOS	20–300	5–7	374–550	Yu et al. (2009b), Liang et al. (2011), Punyapalakul et al. (2012) and Rattanaoudom and Visvanathan (2012)
	PFOA	20–300	5–7	175–524	
Granular activated carbon	PFOS	15–250	4.4–7.2	160–229	Yu et al. (2009b), Ochoaherrera and Sierraalvarez (2008) and Carter and Farrell (2010)
	PFOA	15–250	5–7.2	112–161	
Zeolites	PFOS	15–300	3–5	8–126	Punyapalakul et al. (2012) and Ochoaherrera and Sierraalvarez (2008)
	PFOA	15–300	3	34–37	
Anion-exchange	PFOS	20–400	3–5	210–2575	Yu et al. (2009b); Deng et al. (2010)
	PFOA	20–250	5	1206	
Hydrotalcite	PFOS	1–1000	/	998	Rattanaoudom et al. (2012)
	PFOA	1–1000	/	1033	
Quaternized cotton	PFOS	95–459	5–9	1647	Deng et al. (2012)
	PFOA	78–380	5–9	1280	
Aminated rice husk	PFOS	0–250	5	1322	Deng et al. (2013)
	PFOA	0–207	5	1028	
Quaternized Polyacrylonitrile fiber	PFOS	20–300	2–10	1496	Chen et al. (2017b)
	PFOA	20–300	2–10	1086	
Chitosan bead	PFOS	46–371	3	2745	Zhang et al. (2011)
Mesoporous carbon nitride	PFOS	280	3.25	388	Yan et al. (2013)
Maize straw-origin ash	PFOS	1–500	7	811	Chen et al. (2011)
Molecular imprinted polymer	PFOS	50	5	550	Yu et al. (2008)

contaminants. The ideal treatment for the recalcitrant PFCs should be the cleavage of long C–F chains to F^- ions, which can easily combine with Ca^{2+} to form environmental harmless CaF_2. Hydroxyl radicals (HO·) are generated in most advanced oxidation processes in situ, which can attack the PFCs to form short C–F chain or carbon center radicals through direct electron transfer. Table 1.5 shows the conditions and removal efficiencies under different advanced treatment processes.

Based on the high removal efficiency, it seems that some of the aforementioned processes can achieve better removal efficiency for PFCs. It should be noted that most of these researches have been performed under high temperature, high pressure, or high radiation source. These harsh laboratory reaction conditions indicate

Table 1.5 PFCs removal using different advanced treatment processes

	Conditions	Removal (%)	Reference
UV/H_2O_2(PFOA)	[PFOA] = 1.35 mM [H_2O_2] = 1.0 M λ = 240–460 nm (24 h)	35.6%	Hori et al. (2004)
UV-($S_2O_8^{2-}$) (PFOA)	[PFOA] = 1.35 mM[$S_2O_8^{2-}$] = 50.0 mM λ = 240–460 nm (4 h)	100%	Hori et al. (2005)
UV-photocatalyst ($H_3PW_{12}O_{40}$•$6H_2O$)(PFOA)	[PFOA] = 1.35 mM [$H_3PW_{12}O_{40}$•$6H_2O$] = 6.68 mM λ = 240–460 nm (24 h)	100%	Hori et al. (2004)
Zerovalent iron (PFOS)	[PFOS] = 372 mM[Fe (0)] = 9.60 mM T = 350°°C, 20 MPa (6 h)	100%	Hori et al. (2006)
UV-alkaline isopropanol (IPA) (PFOS)	[PFOS] = 40.0 μM [IPA] = 68.0 mM λ = 254 nm	76% (1 d) 92% (10 d)	Yamamoto et al. (2007)
Sonolysis (PFOS)	[PFOS] = 0.19 mM Ultrasonic generator (200 kHz) (60 min)	60%	Cheng et al. (2008)

that the degradation of PFCs would be unrealistically costly and be incomplete under conditions commonly found in the field or practical PFCs-containing wastewater in large-scale application.

1.4 Final Considerations and Future Perspectives

The aforementioned advanced treatment technologies are mainly directed toward the removal of priority PPCPs or PFCs, as well as partial other types of contaminants. Since both inorganic and organic contaminants would occur in wastewaters, combined treatment processes or advanced technologies should be used for municipal wastewater. Therefore, future efforts should concentrate on the integrated technologies for synergic treatments as tertiary treatment processes. Centralized treatment approach for emerging contaminants can separate promptly and has high efficiency by controlling the removal process and treatment scale. However, the end control treatment requires vast occupancy, high cost of construction, and operation. Emergency situations, such as storm overflow, leakage of the treatment system, and influent loads exceeding the treatment capacity may lead to more serious environmental risks. Therefore, more strategies and legislation for management of PPCPs/PFCs need to be further developed and strengthened for source control and source separation.

The concept of source control is to reduce the manufacture and use through strict legislations and regulations, optimizing PPCPs/PFCs use. PPCPs are usually disposed as solid waste or in wastewater effluent by improper disposal from private households and hospitals; many measures such as supporting studies on quantifying the effectivity, reducing the over-the-counter sale, and limiting the authorization of new pharmaceuticals to products with improved efficacy or application range may be conducive to reduce probability of disposing of rest drug. For PFCs, PFOS with its salts and perfluorooctane sulfonyl fluoride (PFOSF) have been added to the list of "persistent organic pollutants" in the Stockholm Convention. They are still allowed to be limitedly used in some areas including electroplating, polytetrafluoroethylene manufacturing, and optoelectronic industries. More environmentally friendly substitutes should be developed in the next step.

From the pollution sources point of view, the ideal of source separation is to produce a wastewater with an optimal composition for further centralized treatment, preventing PPCPs/PFCs-containing wastewater from being discharged into WWTPs. Nowadays, "cleaner production" programs which contribute to an ecologically, economically, and societal sustainable future are adapted in many industries (Lora et al. 2000). These preventive applications can realize recycling through "green chemistry." Specific treatment technologies toward the priority PPCPs or PFCs can be easily selected to apply in centralized treatment process after source separation. With the development of the advanced analytical instrument and standard analytical procedures, more emerging micro-contaminants would be detected. The environmental legislations, standards, and related removal technologies need to be updated, preventing emerging contaminants transferring into the environment.

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