Chapter 15
Treatment Technologies for the Environmental Micro-pollutant

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Abstract Advance studies related to MPs contamination along with their metabolites are detected in the aqueous environment throughout the world. Their biological nature and continuous emission render them as "prospective pollutant" or "emerging pollutants." The major categories of MPs are divided into eight groups. For the absolute removal of MPs and their metabolites, there is no specific technique and is quite difficult and somewhat impossible because of their distinctive properties. The emission of MPs in large amounts in different aqueous bodies in different parts of the world renders a serious threat to the aquatic as well as human ecosystem. So, the most applicable methods used for MPs are activated carbon absorption, coagulation-flocculation, advanced oxidation process, and ozonation membrane bioreactor and membrane process. The typical WWTPs cannot provide the expected results for the elimination of significant MPs. However, with little efforts, upgrading and optimizing the current protocols in the WWTPs is all set to crucially decrease the loading rates of MPs. Besides all the conventional techniques and processes, advanced oxidation processes (AOPs), activated carbon adsorption (granular activated carbon and powdered carbon), coagulation-flocculation, membrane bioreactor, and membrane process are also applied for the removal of MPs. Among all these persistent treatment methods, advanced oxidation processes and membrane systems are the most efficient techniques and come to the forefront. For both removal of micropollutant and inhibiting the production byproducts and metabolites and other pollutants, a combined treatment should be preferred to achieve the desired results.

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15.1 Introduction

Among different pollutants micro-pollutants (MPs) are defined as anthropogenic chemical compounds that mainly occur in the aquatic environment quite above the natural substantial background level mainly because of urbanization and human activities in modern as well as developing worlds but with concentrations so minute that is found to be in trace levels (i.e., up to μg/L range). Therefore, MPs are specifically defined by their occurrence in low concentration level and anthropogenic origin. Billions of natural as well as anthropogenic chemicals fall into this group of pollutant. Few decades back, the occurrence and concentration level of MPs in the aquatic environment have become an alarming global issue of increasing ecological concern. MPs are also termed as emerging contaminants, engulfing a vast and expanding category of natural as well anthropogenic substances (Stamm et al. 2016). As of today, majority of the countries in the First World have successfully been able to reduce the overall level of MPs in the aquatic environment by legalizing and adopting appropriate effective measures. Consequently, the focus has been shifted to this emerging class of contaminants because of their hazardous nature to the elements of the biotic sphere. Ample of research found that it is not only the pollutants that have been introduced into the environment most recently but also the advancement in the development of analytical techniques and protocols that made it possible to detect such substances despite their minute concentration in the aquatic environment (Brack et al. 2015; Gavrilescu et al. 2015; Guibal et al. 2015).

The significance of MPs in the environment is not essentially due to persistency but because of their biological nature and continuous emission render them as "prospective pollutant" or "emerging pollutants." Advance research found that billions of MPs along with their metabolites have been detected in aqueous bodies all around the world (Escher et al. 2014). Meanwhile the existence of these MPs is very low in the aqueous environment and unlikely results into acute toxicity, but it is concluded that their long-term presence may cause chronic health conditions (Schriks et al. 2010). One study conducted in Germany detected the concentration up to several μg/L of about 55 active pharmaceuticals with nine metabolites in the wastewater of about 49 sewage treatment plants. Similarly, wastewater of several European treatment plants was analyzed, and the result detected about 27 pharmaceutical compounds and four metabolites, with the highest average concentration of about 1.0 μg/L (Larsen et al. 2004). Many of the detected MPs were active pharmaceutical components, additives, excipients, and EDCs. These concluded results are alarming, but the situation is even worse in the developing countries, where the concentration and the number of many MPs have been detected exceedingly high. This can be mainly attributed to the fact that in the developing countries, the majority of these MPs are being sold as off-exchange products, consequently resulting in increasing levels in the aqueous environment (Garcia-Galan et al. 2016). At the end, the widespread scientific viewpoint concluded that a more advanced management approach should be developed and implemented all around the globe (Brack et al. 2015). Yet precise management approach and legislation regulation for the safe permissible level of MPs in the environment needs a further understanding of their fate and distribution, and concerning their harmful effects should also be characterized. This may include the transformation mechanism of several MPs in the environment, the level of toxicity they cause in living organisms, and significant effect on the ecosystem along with the remediation strategies.

15.2 Transport and Sources of MPs in Environment

Micro-pollutants accumulated in the waterbody have a diverse origin, among which the domestic waste effluents are the main source from surface water. In the aquatic environment, pharmaceuticals which are detected frequently mainly originated from convenience stores, drug stores, and hospitals. The main drawback of such chemicals is that they are available without a prescription (i.e., ibuprofen, aspirin, naproxen, acetaminophen). However, these medicines are mainly produced for healthcare purposes for humans and animals, but they are not completely metabolized in the body (Thomas and Foster 2005). Both the residual medicine and their metabolites are excreted by animals and humans into the wastewater. Moreover, the source of waste can be from the manufacturing industries and also expired medicines. Pathways and sources of PMs in the urban water cycle are shown in Fig. 15.1.

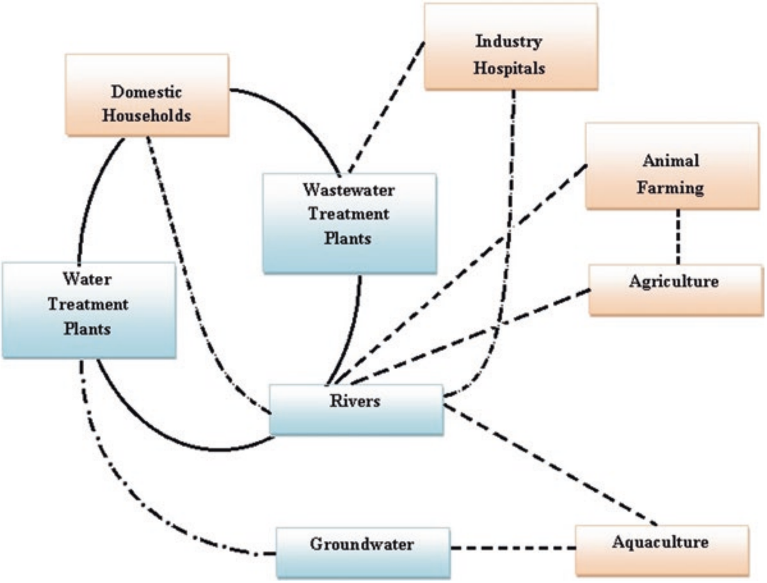

Fig. 15.1 Pathways and sources of PMs in the urban water cycle. (Ellis 2006)

EDCs consist of natural hormones, nonylphenol, insecticides, and bisphenol A, considered as significant MPs. Release of such compounds is from raw materials like flame retardants and plastics. But these compounds can also directly be generated by humans (Poulsen et al. 2005; Prevedouros et al. 2006). These compounds have hormone-like properties, and these EDCs have adversely affected the human health (Sonnenschein and Soto 1998; Ellis 2006). Excretion of these compounds from the human body into the sewage is directly discharged into the water system nearby like lakes and rivers. Therefore, waste from sewage is commonly considered a major source of MPs.

The physical and chemical properties and the bioavailability can influence the existence of MPs in the aquatic environment. A study conducted by Caliman and Gavrilescu evaluated and categorized the elimination and generation of MPs based on some significant factors, that is, environmental factors, physicochemical properties, accumulation and transformation, retention, and transport (Caliman and Gavrilescu 2009).

Moreover, it has been observed that the physical properties of MPs can influence the mobility of pollutants from one phase to another (e.g., soil-water movement). Precipitation, sorption, colloid formation, and complexation all contribute to the retention of MPs. The significant mechanisms of transport consist of dispersion, diffusion, active transport, and advection. The transformation processes, also called the decomposition of the parental compounds as a byproduct, are ineffective to prevent the complete reaching of MPs into the natural environment. In the application of adequate conversion processes to treat the wastewater, it is very difficult to control the concentration of MPs in the marine environment to be accumulated and emitted (Moon-Kyung and Kyung-Duk 2016).

15.3 Categories of MPs in Aquatic Environment

The major categories micro-pollutants in the aquatic environment are divided into eight groups, that is, personal care products (PCPs), agriculture, detergents, and perfluorinated compounds (PFCs), additives, flame retardants, new class, human pharmaceuticals and veterinary drugs, and endocrine disruptive chemicals (EDCs).

15.3.1 Personal Care Products (PCPs) (Table 15.1)

Table 15.1 Class, mode, and fate of PCPs

Micro pollutant	Class	Mode of entry	Fate	Examples	Author
Personal care products (PCPs)	Fragrances and synthetic musks	Direct disposal of industrial effluents and shower waste	Terrestrial runoff freshwater, wastewater treatment plants, estuaries, and sediments	Musk ketone, galaxolide, polycyclic and macrocyclic musks, tonalide	Ellis (2006) and Verlicchi et al. (2010)
	Antiseptics			Chlorophene Triclosan	
	Stimulants			Caffeine	
	UV filters			Methylbenzylidene camphor, Benzophenone	
	Antihypertensive			Diltiazem	
	Insect repellents			N,N-diethyltoluamide	

15.3.2 Agriculture (Table 15.2)

Table 15.2 Class, mode, and fate of agriculture

Micro pollutant	Class	Mode of entry	Fate	Examples	Author
Agriculture	Pesticides	Agricultural waste	Water and soil	DDT, chlordane, aldrin	Ellis (2006) and Verlicchi et al. (2010)
	Herbicides			Terbuthylazine, diuron, mecoprop	

15.3.3 Detergents and Perfluorinated Compounds (PFCs) (Table 15.3)

Table 15.3 Class, mode, and fate of PFCs

Micro pollutant	Class	Mode of entry	Fate	Examples	Author
Detergents and perfluorinated compounds (PFCs)	Perfluorooctanoic acid	Households, pesticides, industries, laundries, agricultural applications in dispersants and pesticides	Sewage treatment plants	Alkylphenol carboxylates, alkylphenols (octylphenol and nonylphenol)	Ellis (2006) and Verlicchi et al. (2010)
	Perfluorooctane sulfonate				

15.3.4 Additives (Table 15.4)

Table 15.4 Class, mode, and fate of additives

Micro pollutant	Class	Mode of entry	Fate	Examples	Author
Additives	Gasoline	Disposal of exhausted engine oil and mobile exhaust	Water, soil, and air	Methyl t-butyl ether, dialkyl ethers	Ellis (2006) and Verlicchi et al. (2010)
	Industrial	Municipal waste and food resources		Aromatic sulfonates, chelating agents (EDTA)	

15.3.5 Flame Retardants (Table 15.5)

Table 15.5 Class, mode and fate of flame retardants

Micro pollutant	Class	Mode of entry	Fate	Examples	Author
Flame retardants		Industries and household stuff (electronics, baby products, furniture, appliances)	Dry and wet disposition on sediment and soil that leads to bioaccumulation in the food chain	Hexabromocyclododecane, diphenyl ethers, tetrabromobisphenol A tris(2-chloroethhyl) phosphate, C10–C13 chloroalkanes, polybrominated	Ellis (2006) and Verlicchi et al. (2010)

15.3.6 New Class (Table 15.6)

Table 15.6 Class, mode, and fate of new class pollutants

Micro pollutant	Class	Mode of entry	Fate	Examples	Author
New class	Antibiotic resistance genes	Genetic adaptation and mutations	Transfer of horizontal gene in microorganisms	tet (O), tet (W), sul (I), sul (II)	Ellis (2006) and Verlicchi et al. (2010)
	Nanomaterials	Research institutes	Water		

15.3.7 Human Pharmaceuticals and Veterinary Drugs (Table 15.7)

Table 15.7 Class, mode, and fate of pharmaceuticals and veterinary drugs

Micro pollutant	Class	Mode of entry	Fate	Examples	Author
Human pharmaceuticals and veterinary drugs	Antibiotics	Hospital disposal/ discharges, farmland waste, accidental spills	Groundwater, streams, river and wastewater treatment plants	Cefazolin, ciprofloxacin, erythromycin, lincomycin, amoxicillin, chlortetracycline, norfloxacin, doxycycline, penicillin	Ellis (2006) and Verlicchi et al. (2010)
	Antidiabetics			Glibenclamide	
	Analgesics			Acetylsalicylic acid, diclofenac, ibuprofen, indomethacin, acetaminophen, codeine, dipyrone, ketoprofen, paracetamol, mefenamic acid naproxen	
	Blood lipid regulators			Bezafibrate, etofibrate, fenofibric acid, atorvastatin, clofibric acid, pravastatin, gemfibrozil	
	Cardiovascular drugs (β-blocker)			Metoprolol, sotalol, atenolol, propranolol, timolol	
	Psychiatric drugs			Gabapentin, salbutamol, phenytoin, primidone, carbamazepine, diazepam	
	Veterinary drugs			Flunixin	
	X-ray contrast agent			Diatrizoate, iopromide, iopamidol	
	Drugs of abuse			Cocaine, tetrahydrocannabinol, amphetamine	

15.3.8 Endocrine Disruptive Chemicals (EDCs) (Table 15.8)

MPs are commonly found in water bodies at very low concentrations, ranging from a few mg/L to more than a few μg/L. The "minimum concentration" and the vast variety of MPs are not only difficult to detect by the associated analysis procedure but also generate challenges for wastewater and drinking water treatment processes. Wastewater treatment plants (WWTPs) nowadays are not designed specifically to eliminate MPs. Consequently, the majority of these MPs are able to pass through the treatment methods used for wastewater because of their continuous introduction and significance of persistency. Furthermore, monitoring actions and precautions for MPs have been fully implemented in majority of the wastewater treatment plants (WWTPs) (Bolong et al. 2009). So, as a result, the fate of most of these MPs lies within the aquatic environment, where they pose great threats to the biological ecosystem and also create mass trouble for the drinking water plants. The occurrence of MPs in the marine environment has been most commonly associated with a large number of harmful effects which consist of long-term and short-term toxicity and microorganisms resistant to antibiotic and disrupting effects of endocrine (Fent et al. 2006; Pruden et al. 2006).

Currently, for most of the micro-pollutants, standard protocols and discharge guidelines do not exist. In order to set proper guidelines and standard permissible limits for significant MPs, further advanced research on the biotic responses to these pollutants (including long-term and short-term toxic effects) is of utmost importance. Furthermore, the regulatory and scientific communities should provide insight into the impact of each of the MP and also their antagonistic and synergistic effects. Throughout the globe many research articles have been published regarding the occurrence of emerging MPs in different aquatic environments and water bodies such as groundwater (Deblonde et al. 2011) and wastewater (Lapworth et al. 2012) as well as effective treatment methods for the removal of MPs (Bolong et al. 2009). Additionally, researchers reviewed the removal of pharmaceutical and its efficiency by the conventional activated sludge systems by analyzing the municipal wastewater (Verlicchi et al. 2010). Similarly, Ze-Hua et al. (2009) studied the significant biological, chemical, and physical removal of endocrine-disrupting compounds. Moreover, no data have been recorded yet concerning the comprehensive summary of the occurrence of such miscellaneous MPs in the aquatic environment and also the removal of significant MPs in advanced treatment processes.

Table 15.8 Class, mode, and fate of EDCs

Micro pollutant	Class	Mode of entry	Fate	Examples	Author
Endocrine disruptive chemicals (EDCs)	Steroids and hormones		Groundwater and soil	Estriol, diethylstilbestrol, estradiol, androstenedione, ethinylestradiol, estrone, testosterone, progesterone	Ellis (2006) and Verlicchi et al. (2010)

15.4 Environmental Effects

Environmental risks and effects posed by MPs mainly depend on their chemical and physical speciation and affinity for water and solid matter, which can cause a significant change and impact on their bioavailability. Moreover, the danger of such MPs for the biotic entities is also dependent on the mobility and their ability to accumulate and end up in the food chain. In recent studies, it has been revealed that contaminants get accumulated in the tissues of marine organisms mainly by suspended matter or ingesting water. Results however concluded that the concentration level of MPs in the tissue of marine organisms may be recorded at a level comparable with the concentration level found in the marine environment or even more. The vast variation in the ecological conditions in different aqueous regions can also influence the bioavailability. Conditions such as temperature, pH changes, salinity, and turbidity can be illustrated. Additionally, the physicochemical parameters along with the species sensitivity can change the ability to bioaccumulate the malicious pollutant. The majority of different MPs have different potential levels to bioaccumulate, even when they are being exposed to the same concentration level of a specific pollutant. Similarly, individuals of one species belonging to a specific group when exposed to an equal concentration of contaminants during the same period of time cannot possibly accumulate the contaminant at the same rate. It is associated with many factors such as individual size, age, sex, and other physiological condition of the organism (Garnaga 2012).

The current data recorded for the concentration of MPs in the treated effluents are quite low in order to assess the risk posed to the marine ecosystem. Target and nontarget compounds after being chemically analyzed provided only a little information about the significant danger associated with MPs to the human life and the surrounding environment. Furthermore, the analysis and detection of nontarget elements posed difficulties for a research analyst. Despite the fact that in treated sewage effluents a complex mixture of MPs is present along with transformation and degradation of MPs is also occurring, therefore it is difficult to foresee the hazard associated with this type of approach, which is entirely based on the criteria for each of the chemical substance (Fang et al. 2017). Most of the MPs in the treated wastewater that are present exhibit toxic properties. Therefore, the major detrimental consequence of MPs is basically attributed to the potential sublethal and acute toxicity effects on the marine biota. Several studies on ecotoxological provided the desirable results and seem to be an effective and suitable tool for assessing the negative impacts arising from the treated wastewater flooded with MPs. In certain marine ecosystems, the occurring results reflecting from the ecotest are posing the actual threat to the organisms. They are performed in less time, and the need of specialized analytical tools and analyst is excluded. Ecotoxicity experiments are carried out on a biological sample, that is, a population of a specific species of organism, exposed to certain modifications, that is, a particular contaminant for a period of time. Advance studies associated with the ecotoxicological studies are based on marine organisms, that is, bacteria, macrophytes, mollusks, crustacean, algae, and fish.

Furthermore, it is highly recommended to perform experiments incorporating different species that represent several trophic levels (Tran et al. 2018).

Research studies made in decade back revealed that many of the MPs identified have a great potential to interrupt the endocrine processes in many organisms. These chemicals are termed as endocrine-disrupting chemicals (EDCs). Basically, EDCs are occurring naturally as well as anthropogenically in the environment. The definition adopted by the World Health Organization (WHO) is that EDCs are exogenous and a mixture of EDCs compounds have the ability to disrupt the function of the entire endocrine system which will consequently show negative responses in an individual organism or affect their offsprings or in the entire subpopulation. The EDCs belong to different families and are being able to disturb the natural hormonal system by counteracting or mimicking as a natural hormone in the organisms that are exposed to such chemicals (Huerta et al. 2016). At present estimation, there are about a hundred thousand of emerging compounds among which thousand are EDCs (Gore et al. 2014). Those chemicals may include bisphenol, phthalates, brominated flame retardants, polychlorinated biphenyls (PCBs), organic tin compounds, and some pesticides (Kima et al. 2015). Standard protocols for the biological treatment of waste effluents incorporated in a typical WWTPs result only to remove a certain fraction of compounds from the entire group of EDCs, comprising mainly of polar nature (Välitalo et al. 2016). The presence of EDCs is detected in samples taken from the surface water as well as in the groundwater. This observable fact is alarming due to EDCs, when released into the water bodies are more likely to affect the biotic entities, even when they are present at very low concentration (Kima et al. 2015). Ample of literature reported that even at very low concentrations EDCs can cause a very adverse effect on the marine environment. One study reported that zebrafish were susceptible to estradiol at a concentration very low, that is, 0.2 ng/L (Westerlund et al. 2000).

15.5 Treatment Technologies for Micro-pollutant Removal

For the complete elimination of MP groups, there is no specific technique and is quite difficult and somewhat impossible because of their distinctive characteristics. The treatment methods cannot remove both MPs and bulk compounds with a maximum efficiency rate. The most applicable treatment technique used for MPs is activated carbon absorption (GAC and PAC), coagulation-flocculation, advance oxidation process (AOPs) and ozonation, membrane bioreactor (MBR), and membrane processes.

15.5.1 Coagulation-Flocculation

Coagulation-flocculation treatment process is generally used to eliminate most of the dissolved particulate matter and colloids. Table 15.9 represents the removal efficiencies of some of the significant MPs processed by the coagulation-flocculation process.

Commonly, the coagulation-flocculation treatment procedures are ineffective for the removal of most of the MPs. A study is conducted by Matamoros and Salvadó (2013) to evaluate the elimination efficiency of MPs in a coagulation-flocculation. Similarly, a maximum elimination efficiency recorded was 50% in treated hospital wastewater by the process of coagulation-flocculation, and a significant reduction was recorded up to 80% of compounds such as musk, that is, tonalide and galaxolide. Similarly, other elimination efficiencies were 23%, 42%, and 46% for ibuprofen, naproxen, and diclofenac, respectively. Another similar study was done by Asakura and Matsuto (2009) which concluded that by the treatment technique of coagulation, the removal of bisphenol A was not very effective for the treated landfill effluents, but comparatively results for MPs such as nonylphenol (90%) and DEHP (70%) were quite impressive.

Taking into account, all the techniques and process of coagulation-flocculation provided a minimum elimination efficiency for majority of MPs except for some significant pharmaceuticals and musk, that is, nonylphenol and diclofenac. This procedure also showed poor results for the pesticides. Moreover, neither the temperature factor nor the dose of coagulant effect of the removal of pesticide substantially was recorded in various studies (Thuy et al. 2008). The chemical composition of wastewater, when treated by the coagulation-flocculation processes, influences the elimination rates of MPs either positively or negatively. However, the waste effluents have a huge content of fats that enable to remove large amounts of

Table 15.9 Eliminations of some MPs during coagulation-flocculation progression

Coagulant	pH and dosage	Compounds	Removal efficiency (%)	Author
$Al_2(SO_4)_3$/ $FeCl_3$	7 and 25, 50 ppm	Diclofenac	21.6 ± 19.4	Surez et al. (2009)
$Al_2(SO_4)_3$/ $FeCl_3$	7 and 25, 50 ppm	Carbamazepine	6.3 ± 15.9	
$Al_2(SO_4)_3$/ $FeCl_3$	7 and 25, 50 ppm	Tonalide	83.4 ± 14.3	
$Al_2(SO_4)_3$/ $FeCl_3$	7 and 25, 50 ppm	Ibuprofen	12.0 ± 4.8	
$Al_2(SO_4)_3$/ $FeCl_3$	7 and 25, 50 ppm	Naproxen	31.8 ± 10.2	
$Al_2(SO_4)_3$/ $FeCl_3$	7 and 25, 50 ppm	Sulfamethoxazole	6.0 ± 9.5	
$Al_2(SO_4)_3$/ $FeCl_3$	7 and 25, 50 ppm	Galaxolide	79.2 ± 9.9	

hydrophobic compounds (Surez et al. 2009). However, due to the fact that the dissolved humic acid maximizes the elimination rates of common pharmaceutical compounds such as ibuprofen, diclofenac and bezafibrate (Vieno et al. 2006). On the other hand, the suspended organic matter in the waste effluents may block the elimination of MPs (Choi et al. 2008). Factors like pH, temperature, alkalinity, and mixing conditions also affect the efficiency of coagulation-flocculation (Alexander et al. 2012).

15.5.2 Activated Carbon Adsorption

Basically, the treatment technique of activated carbon adsorption (ACA) is used to control odor and taste in treated water, especially in drinking water. ACA techniques provide better removal of more specifically the secondary waste effluents for treatment. ACA procedure when compared to coagulation-flocculation is more efficient in the elimination of MPs from the treated wastewater (Choi et al. 2008). Furthermore, granular activated carbon (GAC) and powdered activated carbon (PAC) have been widely applied for the adsorption purposes. The efficient elimination of MPs is dependent on the properties and type of adsorbate and also the adsorbent used (Kovalova et al. 2013).

15.5.3 Powdered Activated Carbon (PAC)

Removal of biodegradable organic compounds and resistant compounds when treated with powdered activated carbon (PAC) is considered an exclusive and effective adsorbent. One of the main advantages of using PAC is that they supply continuously fresh carbon, and that in turn can be utilized in certain prevailing circumstances, that is, when the level of contaminant rise in water (Snyder et al. 2007). One research was conducted by Kovalova et al. (2013) in which PAC procedure was applied to evaluate the elimination efficiency of MPs in the treated effluents taken from the MBR hospital wastewater. In the conducted study, PAC dosages were chosen as 8 mg/L, 23 mg/L, and 43 mg/L, and the time selected for retention was about 2 days. Results for the study revealed that PAC adsorbent provided substantial elimination efficiency, especially for metabolites, industrial chemicals, and pharmaceuticals. The removal rate of total load was recorded as 86%. In another study batch tests were conducted and concluded that the high removal rate was >94 % for bisphenol, personal care products, and nonylphenol (Hernandez-Leal et al. 2011).

The removal efficiency of PAC reactors for many MPs also depends on many factors like contact time, physical properties of targeted contaminants, PAC concentration/dosage, and water composition (Snyder et al. 2006; Boehler et al. 2012). Similarly, a research study is conducted by Westerhoff et al. (2005), and they observed in their experiments that at the higher dosage of PAC (i.e., 20 mg/L), the elimination efficiencies of MPs were quite impressive regardless of their initial MP concentration. So it was concluded from the study that the addition of PAC in the wastewater treatment plants seems to be an efficient way for the elimination of majority of micro-pollutants in unit time (Bolong et al. 2009).

15.5.4 Granular Activated Carbon (GAC)

Rossner et al. (2009) assessed that the dose of about <10 mg/L of granular active carbon (GAC) was used in order to control the taste and odor of drinking water. The dose used was sufficient enough for the treatment of lake water and the majority of compounds were removed, providing an elimination efficiency of about 99%. Elimination efficiencies of pharmaceuticals and steroidal estrogen were evaluated and found to be in a full-scale GAC plant for the treatment of wastewater. Maximum removal rates were recorded for steroidal estrogens, but the removal rates recorded for pharmaceuticals were found to be very low. More specifically, the elimination efficiencies of indomethacin, diclofenac, and mebeverine ranged from 84% to 99%. However, the elimination efficiencies of propranolol and carbamazepine ranged from 17% to 23% (Grover et al. 2011). Evaluating PAC the contact time of GAC also influenced the efficiency rates. The minimum contact time of GAC reactor decreased its adsorption performance. More specifically, the removal of contaminants depends upon the association between contaminant and particle and pore blocking (Bolong et al. 2009). So treating high contaminated waste effluent with GAC provides very poor results. Overall study results showed that PAC and GAC processes can be considered as efficient techniques for the removal of MPs from the treated wastewater. Moreover, maximum elimination rates of MPs can be achieved by some significant factors such as shape of contaminant, its high compliance of pore size, and its nonpolar characteristics (Rossner et al. 2009; Verlicchi et al. 2010). However, the blocking of pores is basically due to the existing organic matter (OM) that minimizes the efficiency of active carbon (Table 15.10).

15.5.5 Ozonation and Advanced Oxidation Processes

Conventional biochemical and physicochemical actions are not effective for the elimination of major MPs due to their determined structure. In such type of cases, advanced oxidation process and ozonation are the solution considered. Having more degradation rates, the stated technology is not selective to remove contaminants. Besides this, these procedures have an effect of disinfection for water to reuse (Hernandez-Leal et al. 2011). Ozone destroys the pollutants directly or indirectly, but most of the time by producing the hydroxyl (OH), which is strong enough and less choosy for the emerging compounds. The nature of most of the MPs are very

Table 15.10 Elimination of MPs during the process of adsorption

Adsorbent	Dosage mg/L	Contaminants	Removal efficiency (%)	Author
PAC	8, 23, and 43	Sulfamethoxazole	2, 33, 62	Grover et al. (2011) and Kovalova et al. (2013)
PAC	8, 23, and 43	Diclofenac	96, 98, 99	
PAC	8, 23, and 43	Carbamazepine	98, 99, 100	
PAC	8, 23, and 43	Propranolol	>91, >94	
GAC	Full scale	Carbamazepine	23	
GAC	Full scale	Diclofenac	>98	
GAC	Full scale	Estrone	64	
GAC	Full scale	Proponolol	17	
GAC	Full scale	17α-Ethinylestradiol	>43	
GAC	Full scale	17β-Estradiol	>43	

sensitive towards advanced oxidation processes (AOPs) and ozone such as naproxen but some of the MPs are only sensitive to (OH) radicals like atrazine. However, some MPs like TCEP and TCPP have resistance to both forms of oxidation and ozonation (Gerrity et al. 2011). The presence of ultraviolet, Fenton reagent, and H_2O_2 are responsible for the production of hydroxyl radicals (OH).

Ozonation is an effective method of removing tiny pollutants in a full-scale WWTPs (Hollender et al. 2009). Hernandez-Leal et al. (2011) examined the rate of elimination of MPs in the biological way of treatment gray water by ozonation by ozone dose of 5 mg/L. In a wide range, all MPs are selected and treated under substantial levels. Under the same environment with the only change in ozone dose of 5 mg/L, it showed higher removal percentage for most of MPs (Sui et al. 2010). The elimination rates of most significant MPs such as diclofenac, carbamazepine, sulpiride, trimethoprim, and indomethacin exceeds more than 95%. However, the rate for bezafibrate removal was evaluated, which resulted in 14% only because of the stable molecular structure of bezafibrate (Kim et al. 2009) compared the elimination efficiencies of compounds like pharmaceutical compound using UV. The results show us that the UV process alone acquires high rates of removal (>90%) for diclofenac, antipyrine, and ketoprofen, but the rate of elimination for macrolides ranged from 24% to 34%. Another study confirmed that H_2O_2 and UV together achieved much higher rate of efficiencies for most micro-pollutants. However, under the same situation when UV is applied to the Fenton process, the total rate of removal is increased. In addition, the presence of such dissolved organic material in wastewater enhances the removal rate of MPs. The oxidation process is not able to provide the complete mineralization of such emerging compounds and produce byproducts. Also, metabolite arises from such reactions (Hollender et al. 2009; Reungoat et al. 2011). Sand filtration or activated carbon filtration may be applied to eliminate these unwanted compounds (Table 15.11).

Table 15.11 Removals of some MPs during ozonation and AOPs

Treatment (Dose)	Compounds	Removal efficiency (%)	Author
O_3 (5 mg/L)	Metoprolol	80–90	Luo et al. (2014)
O_3 (5 mg/L)	Trimethoprim	>90	
O_3 (5 mg/L)	Bezafibrate	0–50	
O_3 (5 mg/L)	Carbamazepine	>90	
O_3 (5 mg/L)	Ibuprofen	83	
O_3 (5 mg/L)	DEET	50–80	
O_3 (5 mg/L)	Diclofenac	>90	

15.5.6 Membrane Processes

Usually, the removal of micro-pollutants by the process of membrane is acquired by adsorption process onto charge repulsion, membrane and size of pores. The removal percentage of membrane processes mostly depends upon the membrane process type, blocking of membrane pores, operating condition, properties of selected tiny pollutants, and characteristics of membrane (Schäfer et al. 2011). Ultrafiltration (UF) and microfiltration (MF) are more effective in eliminating process for turbidity, and such type of processes are inadequate for eliminating micro-pollutants because of the molecular sizes of significant MPs. Contaminants, however, can be eliminated via contact with the natural organic matter (NOM), or it can be eliminated through adsorption onto the polymers of membrane. Jermann et al. (2009) examined the efficiency removal of estradiol and ibuprofen by ultrafiltration without the existing natural organic matter. In hydrophilic ultrafiltration membrane, removal rates of estradiol and ibuprofen were found nearly 8% and negligible, respectively. In hydrophobic membrane, eliminating efficiencies of estradiol and ibuprofen are generally increased to 80% and 25%, respectively. However, UF and MF processes worked alone in removing of MPs due to their poor performance. So these processes have to combine with other methods of treatment, like reverse osmosis (RO) or nanofiltration (NF). Garcia et al. (2013) combined the RO and MF processes for the reuse of domestic wastewater and for the removal of micro-pollutants. For example, up 50% DEHP was removed with the microfiltration treatment technique only. However, the combined system of RO and MF improved the rate of elimination of micro-pollutants. Removal efficiencies of such MPs lied between 65% and 90% excluding nonylphenol and ibuprofen. A study presented that the combined system of RO and MF has significant removal efficiencies greater than 95% for most MPs, except caffeine and mefenamic acid (Sui et al. 2010) (Table 15.12).

Reverse osmosis (RO) has such a great effect for the complete removal of almost all the persistent micro-pollutants (Yangali-Quintanilla et al. 2011). Comparatively, the performance rate of reverse osmosis treatment is more effective than nanofiltration for pesticides, endocrine disruptors and pharmaceuticals. The removal rate of micro-pollutants obtained by RO was very similar to NF's result. Removal efficiencies for ionic contaminants and neutral contaminants treated by the NF were estimated as 97% and 82%, respectively. Removal efficiencies of same pollutants treated by reverse osmosis were found as 99% and 85%, respectively.

Table 15.12 Elimination of some MPs by membrane processes

Membrane	Water type	Membrane type	Compounds	Removal efficiency (%)	Author
UF	Synthetic water	RC4 flat-sheet	Estradiol	Up to 80	Yangali-Quintanilla et al. (2011)
UF	Synthetic water	PES flat-sheet	Ibuprofen	Negligible	
UF	Synthetic water	PES flat-sheet	Ibuprofen	7	
UF	Synthetic water	RC4 flat-sheet	Estradiol	Up to 25	
RO	Secondary effluent	Filmtec TW30	Sulfonamides	>93	
RO	Secondary effluent	Filmtec TW30	Ibuprofen	>99	
RO	Secondary effluent	Filmtec TW30	Bisphenol A	>99	
RO	Secondary effluent	Filmtec TW30	Macrolides	>99	

15.5.7 Membrane Bioreactor (MBR)

Membrane bioreactor is a process that combines the treatment of membrane filtration and stimulated sludge biological treatment. There are so many benefits of this technology (MBR) associated with conventional WWTPs. Such benefits involve the higher effluent quality, precise control of the SRTs, higher biomass concentration, less requirement of space, minimum increasing of the sludge problem, and converting the flexibility of current WWTPs to MBR system. Membrane bioreactor has a great ability to eliminate a very wide range of MPs that include the emerging compounds resistant to stimulated sludge process (Radjenovic et al. 2009). The removal of MPs through the MBR process most of the time depends upon the SRT, content of water, concentration, conductivity, operating temperature, and pH (Kovalova et al. 2012).

Trinh et al. (2012) investigated that the MBR process eliminates the micro-pollutants on a full scale. Higher rates of elimination were found for most of the micro-pollutants. However, the removal efficiencies of carbamazepine, diclofenac, amitriptyline, diazepam, sulfamethoxazole, fluoxetine, omeprazole, trimethoprim, and gemfibrozil ranged in between 24a% and 68%, and such compounds are said to be the indicators due to their less rate of removal in MBR treatment. The main source of drugs is waste effluents that arise from hospitals (Verlicchi et al. 2010). Kovalova et al. (2012) examined the fate of such MPs in the membrane bioreactor process treating hospital waste. Hence, the wastewater is mainly composed of iodinated contrast mean, and total eliminating rates of metabolites and pharmaceuticals were found at only 22%. Total reduction would be around 90% in case if such content were ignored. Beier et al. (2011) suggested that the waste of hospitals could be efficiently treated if we maintain the age of sludge very high (>100 days) in a membrane bioreactor system designed especially for treating the hospital effluent.

MBR technology and conventional activated sludge process usually linked with each other in the sense of removing the MPs. Radjenovic et al. (2007) compared the performance of treatment of laboratory-scale conventional activated sludge and MBR process in terms of removing the pharmaceuticals. Both systems are treated with ibuprofen, naproxen, hydrochlorothiazide, paroxetine, and acetaminophen in high level. However, results showed that membrane bioreactor system was comparatively stable for removing several contaminants, and some MPs were treated somewhat more than the process of conventional activated sludge.

Like other technologies of treatment, MBR processes were also influenced by numerous factors such as HRT, operating temperature, and SRT. MBR systems functioned at greater sludge age offer greater eliminating efficiency for such pollutants due to diverse MPs present in wastewater (Roh et al. 2009) (Table 15.13).

15.6 Conclusion

In the present time, MPs are frequently detected in significant drinking water reservoirs and sources like rivers, lakes, and groundwater. Presences of the MPs in high amount in different aqueous bodies in various parts of the world pose a threat to the aquatic as well as human ecosystem severely. However, the typical WWTPs cannot provide the expected results for the elimination of the majority of MPs. In order to achieve the desired results, it is important to apply appropriate treatment technologies to minimize the ecotoxicological effects of MPs in the surrounding environment. Many of the existing conventional WWTP elimination performances of MPs are futile because of the presence of low amount of MPs in the waste effluents and also because of the vast MP physicochemical properties. MPs especially having the biodegradable nature and polar molecular structure pass during the WWTPs to the water bodies receiving such treated water without being sufficiently treated. However, with little effort, upgrading and optimizing the current process in the WWTPs is all set to crucially decrease the loading rates of MPs. Besides all the conventional procedures and processes, coagulation-flocculation, advance oxidation processes (AOPs), activated carbon adsorption (granular activated carbon and powdered activated carbon), membrane bioreactor, and membrane processes are also applied for the removal of MPs. Within the persistent treatment procedures, membrane system and advanced oxidation processes come to forefront. However, these treatment techniques are very effective in eliminating the MPs, but they also have some disadvantages such as causing to produce new byproducts and metabolites at a very high operating cost. In the removal of micro-pollutant and inhibiting the production byproducts and metabolites and other pollutants, a combined treatment should be preferred to achieve the better results.

Table 15.13 Elimination of some MPs by MBR

Water type	Membrane type	Contaminants	Removal efficiency (%)	Author
Raw wastewater	Full-scale hollow fiber	Carbamazepine	24	Radjenovic et al. (2007)
Raw wastewater	Full-scale hollow fiber	Estriol	~100	
Raw wastewater	Full-scale hollow fiber	Ibuprofen	~100	
Raw wastewater	Full-scale hollow fiber	Estrone	~100	
Raw wastewater	Full-scale hollow fiber	Bisphenol A	~100	
Raw wastewater	Full-scale hollow fiber	Diclofenac	43	
Raw wastewater	Full-scale hollow fiber	Trimethoprim	30	
Raw wastewater	Full-scale hollow fiber	Sulfamethoxazole	60	
Hospital effluent	Full-scale flat sheet	Ibuprofen	>80	
Hospital effluent	Full-scale flat sheet	Diclofenac	<20	
Hospital effluent	Full-scale flat sheet	Carbamazepine	<20	

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