Review

The Occurrence of Micropollutants in the Aquatic Environment and their Removal Technologies

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Abstract: Pharmaceutical compounds in aquatic environments are recognized as emerging contaminants due to their potential risks to living organisms. The growing accessibility, widespread use, and disposal of pharmaceuticals have led to a significant increase in their presence within wastewater streams. Due to their persistence and resistance to biodegradation in aquatic environments, these water-soluble and pharmacologically active organic micropollutants often evade conventional wastewater treatment processes, leading to incomplete removal and an escalating global concern. This review examines the classification, occurrence, and associated environmental and health risks of commonly detected pharmaceutical contaminants in aquatic systems. Additionally, it provides an overview of advanced treatment methods being developed to implement a fourth purification stage in wastewater treatment plants. Biological, chemical, physical, and hybrid purification technologies are critically reviewed, with a focus on their performance characteristics and potential applications.

Keywords: wastewater treatment; micropollutants; pharmaceutical contaminants; diclofenac; ibuprofen; paracetamol; advanced treatment processes; membrane filtration; nanofiltration

1. Introduction

In recent decades, the presence of micropollutants (MPs) in aquatic environments has become a growing global concern. Often referred to as emerging contaminants (ECs), these pollutants encompass a wide range of substances, including pharmaceuticals, endocrine-disrupting chemicals (EDCs), personal care products, industrial chemicals, microplastics, and pesticides [1–3]. These contaminants originate from various sources, such as agricultural runoff, industrial emissions, and household waste, and they enter the environment through different pathways, including leaching, atmospheric deposition, and wastewater discharge [4]. Many of these pollutants persist in the environment and accumulate in the food chain, potentially posing long-term risks to both ecosystems and human health [5].

The global production of MPs has increased dramatically, rising from 1 million tons per year in 1930 to 400 million tons annually by 2000 [6]. The European Union has registered over 100,000 chemical compounds, with 30,000 to 70,000 of these being consumed worldwide on a daily basis [7]. MPs are often found in water at low concentrations, ranging from nanograms per liter (ng/L) to micrograms per liter (µg/L), making their detection and analysis challenging and complicating water and wastewater treatment processes [8–10]. These compounds vary in molecular weight, with pharmaceuticals typically ranging between 150 and 500 Da [11].

Current wastewater treatment plants (WWTPs) are not designed to specifically remove micropollutants [12,13]. Many MPs can pass through conventional treatment stages without being degraded or removed due to their persistent nature . As a result, these substances can end up in the aquatic environment, threatening wildlife and complicating the provision of safe drinking water. The

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presence of MPs in aquatic ecosystems is linked to various adverse effects, including both short- and long-term toxicity, endocrine disruption, and the development of antibiotic resistance in microorganisms [14,15].

Although some countries and regions have established regulations for the control of certain micropollutants, WWTPs do not consistently adhere to emission guidelines or micropollutant standards. Nevertheless, progress has been made. The European Union's water policy, initiated by Directive 2000/60/EC, provided a framework to address high-risk substances [16]. The 2008/105/EC Directive [17] identified 33 key substances with approved environmental quality standards [18,19]. The 2013/39/EU Directive [20]

expanded this list, recommending the monitoring and treatment of 45 priority substances, and the 2015/495/EU decision outlined measures for eliminating hazardous substances such as 17-alpha-ethinylestradiol (EE2), triallate, 17-beta-estradiol (E2), oxadiazon, diclofenac, 2,6-di-tert-butyl-4-methylphenol, macrolide antibiotics, methiocarb, neonicotinoids, 2-ethylhexyl-4-methoxycinnamate, and estrone (E1) [18,21,22]. Meanwhile, other harmful substances like ethoxylates and nonylphenol have already been regulated in Canada [23]. However, many micropollutants, particularly pharmaceuticals and steroid hormones, remain unregulated. To establish comprehensive standards, further research on the effects of these substances on human and environmental health is critical [24].

Given their significance and the many ecological risks they pose, this review focuses on the removal of pharmaceutical contaminants (PCs) from wastewater. The objective is to identify and categorize the major classes of pharmaceuticals contributing to wastewater and to provide an overview of the methods being explored to implement a fourth purification stage in WWTPs. Biological, chemical, and physical purification processes are reviewed, and their characteristics are discussed. The occurrence of the most commonly detected PCs in various water sources and regions, as well as the harmful effects of these substances on the environment, ecosystems, and human health, are also examined in the literature.

2. Micropollutants in Aquatic Ecosystems

2.1. Sources of Micropollutants

MPs comprise a wide range of emerging contaminants, which can be categorized into several groups such as pharmaceuticals, personal care products (PCPs), endocrine-disrupting chemicals (EDCs), pesticides, industrial chemicals, and microplastics. Each group has its own distinct characteristics and applications [2]. Table 1 summarizes the primary sources of these major categories of micropollutants in aquatic environments.

Table 1. Sources of micropollutants in the aquatic environment [1–3].

Categories	Key Subcategories	Primary Sources
Pharmaceuticals	NSAIDs, lipid regulators, antibiotics, β-blockers, contrast media, and anticonvulsants	Domestic sewage (from human excretion), Effluents from hospitals, Waste from animal farming and aquaculture
Personal care products	Fragrances, disinfectants, UV filters, and insect repellents	Household sewage (from bathing, shaving, and spraying)
Endocrine disrupting chemicals	Estrogens	Human excreta-derived domestic wastewater Livestock production and aquaculture activities
Pesticides	Insecticides, herbicides and fungicides	Domestic wastewater originating from inadequate cleaning practices and garden runoff

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Industrial chemicals	Plasticizers, fire retardants	Domestic wastewater generated through the leaching of materials
Microplastics	Microfibers, plastic pellets, synthetic fibers	Domestic wastewater resulting from urban runoff

The sources of micropollutants in water are highly diverse and largely depend on how the substances are used, or, in the case of transformation products/metabolites, where they are produced [21]. Often influenced by rainfall, MPs enter water bodies from both point sources and diffuse sources. These substances typically coexist in environmental compartments alongside other chemicals, forming "environmental mixtures" that can interact with one another, potentially leading to increased toxicities and risks [25].

In recent years, the rise in pharmaceutical production and consumption—largely driven by advances in medicine—has contributed to a substantial increase in pharmaceutical contaminants in waste streams. This trend is particularly pronounced during pandemics, when drug usage surges [26]. The rapid growth in pharmaceutical manufacturing and use has significantly raised the concentrations of PCs in wastewater. Additionally, the widespread availability of over-the-counter medications, often sold without prescriptions or registration, further exacerbates the environmental presence of these contaminants [27]. As a result, water-soluble and pharmacologically active organic micropollutants have become a global issue due to their persistence and resistance to degradation in aquatic environments [28–31].

Pharmaceuticals in wastewater can be categorized based on their therapeutic applications. To remove these contaminants from wastewater, various physico-chemical and biological treatment methods are employed [26,32–34]. Understanding the molecular characteristics of each compound is crucial in selecting the most suitable removal process, as these characteristics determine how the compounds interact with different treatment methods. Factors such as molecular size, charge, hydrophobicity, and polarity play a significant role in how well a compound is adsorbed by membranes, retained by filters, or degraded by biological or chemical processes [35]. Table 2 provides an overview of key PCs along with their physicochemical properties. These pharmaceutical categories include analgesics and anti-inflammatories, antidepressants, antibiotics, antivirals, anticoagulants, sedatives, cardiovascular drugs, and more [35]. These contaminants may be ionic (pKa < 2), nonionizable (pKa > 2), hydrophobic (log Kow > 2), or hydrophilic (log Kow < 2). Polar compounds (log Kow < 1) typically escape effective removal by wastewater treatment plants [36].

Table 2. Physicochemical properties of pharmaceutical pollutants [35,37–43]..

Pharmaceutical Categories	Pharmaceutical pollutants	Chemical formulas	Mass ($gmol^{-1}$)	pK_a	Log K_{ow}	Ionization State at pH 7
Analgesics and Anti-inflammatories	Aspirin	$C_9H_8O_4$	280	3.5	1.2	Negative
	Diclofenac	$C_{14}H_{11}Cl_2NO_2$	296.2	4.91	4.51	Negative
	Ibuprofen	$C_{13}H_{18}O_2$	206.3	4.15	4.51	Negative
	Paracetamol	$C_8H_9NO_2$	151.2	9.38	0.46	Neutral
	Naproxen	$C_{14}H_{14}O_3$	230.3	4.15	3.18	Negative
Antibiotics	Sulfamethoxazole	$C_{10}H_{11}N_3O_3S$	253.279	5.6-5.7	0.89	Negative
	Erythromycin	$C_{37}H_{67}NO_{13}$	733.93	8.88	2-48	Neutral
	Trimethoprim	$C_{14}H_{18}N_4O_3$	290.32	7.12	0.73	Neutral
Anticonvulsants	primidone	$C_{12}H_{14}N_2O_2$	218	-1 ;12.2	0.91	Negative
	Carbamazepine	$C_{15}H_{12}N_2O$	236.27	13	2.45	Neutral
ß-blockers	Propranolol	$C_{16}H_{21}NO_2$	259.34	9.6	3.48	Neutral
	Metoprolol	$C_{15}H_{25}NO_3$	276.37	9.49	1.88	Positive
Contrast media	Iopromide	$C_{18}H_{24}I_3N_3O_8$	790.0	2;13	-2.10	Neutral
	Iopamidol	$C_{17}H_{22}I_3N_3O_8$	777.1	10.7	-2.42	Neutral

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	Iohexol	$C_{19}H_{26}I_3N_3O_9$	821.1	11.7	-3.05	Neutral
	Clofibric acid	$C_{10}H_{11}ClO_3$	214.65	3.35	2.57	Negative
Blood lipid	Gemfibrozil	$C_{15}H_{22}O_3$	250.34	4.45	4.77	Negative
regulators	Bezafibrate	$C_{19}H_{20}ClNO_4$	361.82	3.44	4.25	Negative
	Pravastatin	$C_{23}H_{36}O_7$	24.53	4.2	3.1	Negative

2.2. Routes of Micropollutants in the Environment

Micropollutants enter the environment through a variety of sources, including industrial emissions (via air and effluents), domestic and hospital waste, livestock, and leachates from landfills, as shown in Figure 1. Among these sources, household wastewater is particularly significant in introducing micropollutants into surface waters, making aquatic environments the primary receptor of these pollutants [44].

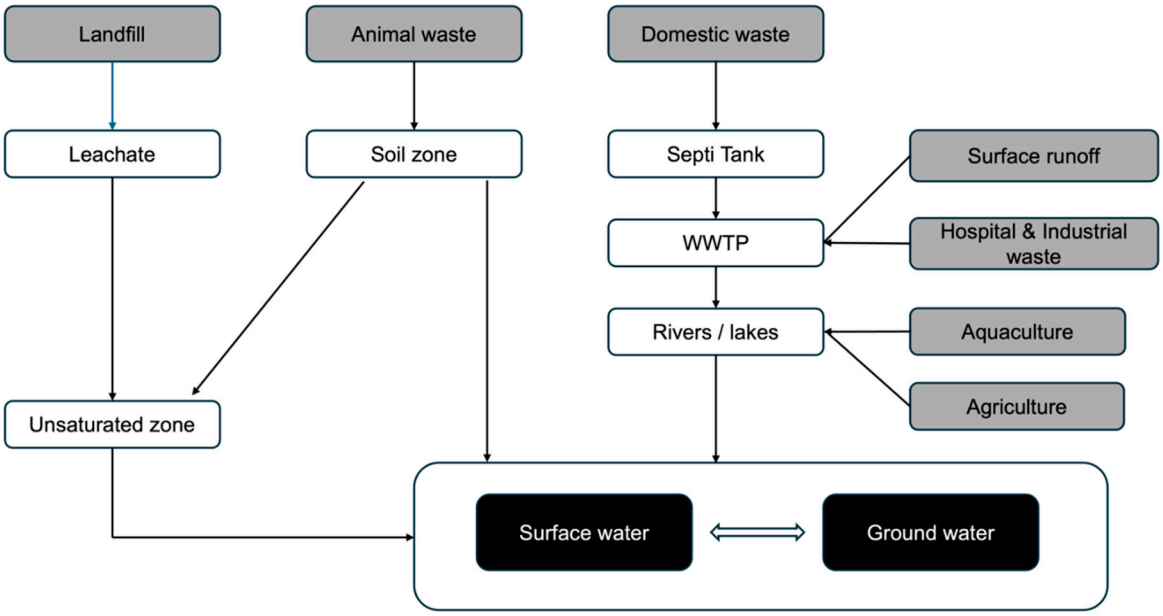

Figure 1. Sources of micropollutants in the environment [45–47].

Various pharmaceuticals are used in both human and veterinary medicine. Approximately 3,000 different compounds are employed as pharmaceuticals, with annual production quantities reaching hundreds of tons [48]. In Western Europe, the average individual consumes over 300 mg of active ingredients daily, with about 99% of this amount being concentrated in just 60 compounds [49,50]. In Germany alone, around 8,100 tons of active substances are used annually [51]. After ingestion, these pharmaceuticals are excreted in urine and feces, both as the original molecule (the portion not metabolized in the body) and as metabolites, which are typically hydroxylated, hydrolyzed, or conjugated forms of the parent compounds [52].

These substances then enter the municipal sewer system through hospitals, healthcare facilities, and private households, as shown in Figures 1 and 2 [53]. Sewage containing pharmaceutical pollutants from manufacturing or processing (depending on the industry) is treated in WWTPs. After treatment in communal or industrial WWTPs, the resulting water is generally discharged into water bodies. However, due to their chemical stability, resistance to biodegradation, and ability to pass through filtration processes based on size, charge, or solubility, pharmaceuticals and other pollutants are often not completely removed in conventional treatment plants. Consequently, these contaminants can find their way into surface and groundwater [54]. Additionally, smaller quantities enter the sewer system through manufacturing processes or improper disposal via sinks and toilets. As a result, these active substances can ultimately contaminate drinking water through bank filtration or surface water contamination [25,55].

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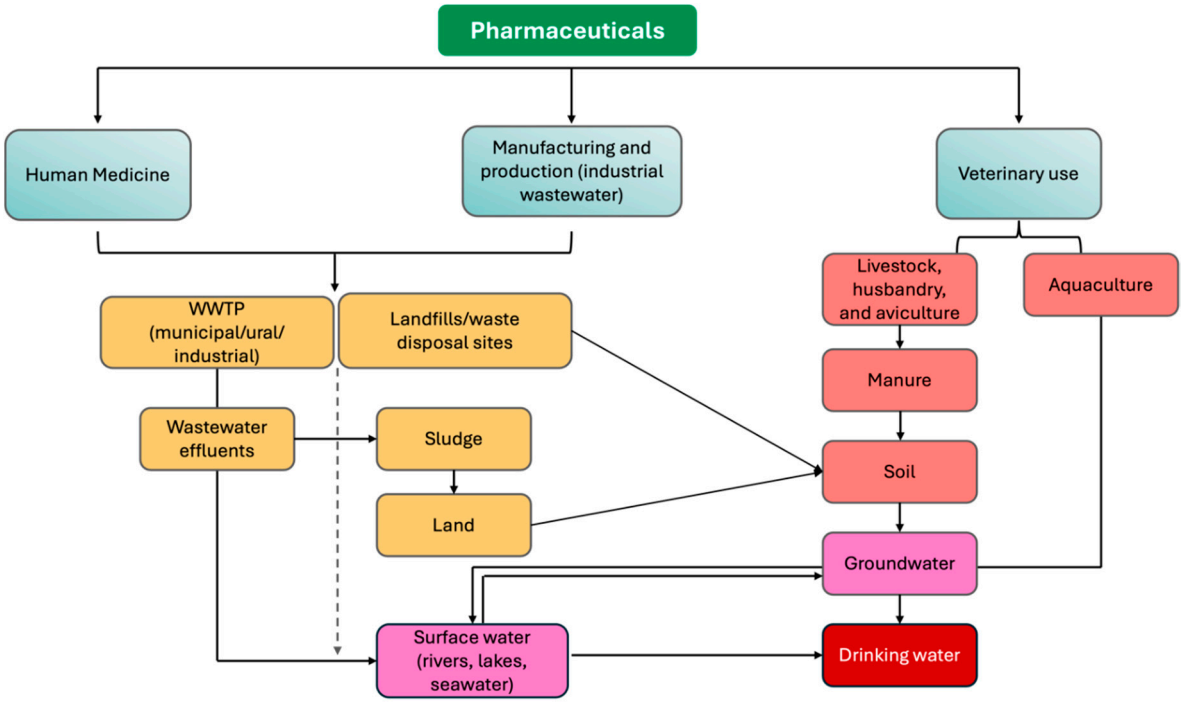

Figure 2. Routes of pharmaceutical compounds into the environment [44,56].

WWTPs are a significant source of pharmaceuticals [2,54,57]. Depending on drug consumption levels and excretion rates, the concentration of individual pharmaceuticals in untreated wastewater can range from nanograms per liter (ng/L) to micrograms per liter (μg/L) [25]. Pharmaceuticals that are commonly found in high concentrations in wastewater include non-steroidal anti-inflammatory drugs (NSAIDs), β-blockers, psychoactive compounds, analgesics, antibiotics, endocrine disruptors, antiretroviral drugs, and cancer treatments [58,59]. Among these, NSAIDs, antibiotics, and analgesics are the most frequently used worldwide [60]. For instance, approximately 35 million people use NSAIDs daily across the globe [61], and in China, domestic production increased from 41,537 tons in 2013 to 46,673 tons in 2017 [62].

In Cuernavaca, Mexico, high concentrations of naproxen (732-4,889 ng/L), acetaminophen (354-4,460 ng/L), and diclofenac (258-1,398 ng/L) have been detected in influent and effluent samples from a WWTP as well as in surface waters of the Apatalco River [63]. Similarly, diclofenac (10,221 ng/L) and acetaminophen (1,234-2,346 ng/L) have been detected in effluents from the Red Sea (Saudi Arabia) [64]. In Brazil, acetaminophen (17.4-34.6 ng/L), diclofenac (19.4 ng/L), and ibuprofen (326.1-2,094.4 ng/L) have been found in surface and bottom water samples from Santos Bay [65]. Table 2 further illustrates the global distribution of the most commonly detected pharmaceuticals in water bodies.

Among the pharmaceutical substances found in wastewater, antibiotics are of particular concern due to their persistent nature, incomplete metabolism, and their ability to spread easily through ecosystems [66]. In China, approximately 92,700 tons of antibiotics were produced, with 48% intended for human use and the remainder for livestock, 46% of which were active metabolites [67]. The most frequently detected antibiotics in wastewater include sulfonamides, quinolones, tetracyclines, fluoroquinolones, and nitroimidazoles [60]. Antibiotic concentrations in various water bodies range from 0.0013 to 0.0125 mg/mL in wastewater, 0.0005 to 0.0214 mg/mL in drinking water, and 0.0003 to 0.0039 mg/mL in river water [68–70] .

Veterinary pharmaceuticals are another direct source of MPs due to their widespread use in treating farm animals, leading to their presence in different aquatic environments [71]. Studies have identified these substances in animal production systems such as pig farming [72], poultry [73], dairy [74], sheep farming [75], and aquaculture [76–78]. In Germany, indoor livestock systems, including cattle, pigs, and poultry, are the primary sources of veterinary pharmaceutical pollution (82%),

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followed by pasture-raised animals (18%), with aquaculture contributing less than 0.5% . These substances can reach aquatic habitats through various pathways, as demonstrated by numerous studies [78–80].

Residues from veterinary medicinal products can runoff into surface waters or seep into groundwater if not absorbed by the soil, potentially contaminating drinking water [81]. Wastewater from concentrated animal feeding operations (CAFOs) often contains a mix of pharmaceuticals, including antibiotics, anthelmintics, synthetic and natural hormones, and NSAIDs, mainly originating from animal excretion [54,82]. Antibiotics are widely used in animal farming to promote growth and improve feeding efficiency in cattle, pigs, and poultry [83]. Research by Klein et al. (2018) highlights a 65% increase in antibiotic use between 2000 and 2015, growing from 21 to over 35 billion daily doses [84].

Aquaculture, particularly fish farming, is another significant route for pharmaceutical contamination. Antibiotics are often used in fish farming to prevent or treat bacterial infections in fish populations [85]. These antibiotics can enter surrounding water bodies directly through fish excretion, uneaten medicated feed, or improper disposal of unused medications [86]. The Asian aquaculture sector has expanded rapidly in recent decades, now accounting for nearly 90% of global production [87]. Studies have shown significant antibiotic use in Chinese aquaculture, contributing to veterinary contamination [78]. However, human activities may have an even greater impact on aquatic pollution than aquaculture due to the higher density of human populations.

2.3. Impact of Pharmaceutically Active Compounds (PhACs) in Water on Human Health and Ecosystems

The older generation in modern societies is increasingly reliant on pharmaceutical drugs, and with the rising prevalence of chronic diseases, the use of therapeutic products is expected to grow [88]. The high concentration of pharmaceutical and personal care product residues in water bodies can lead to long-term (chronic) effects on both human health and ecosystems. Potential health impacts of micropollutants are illustrated in Figure 3.

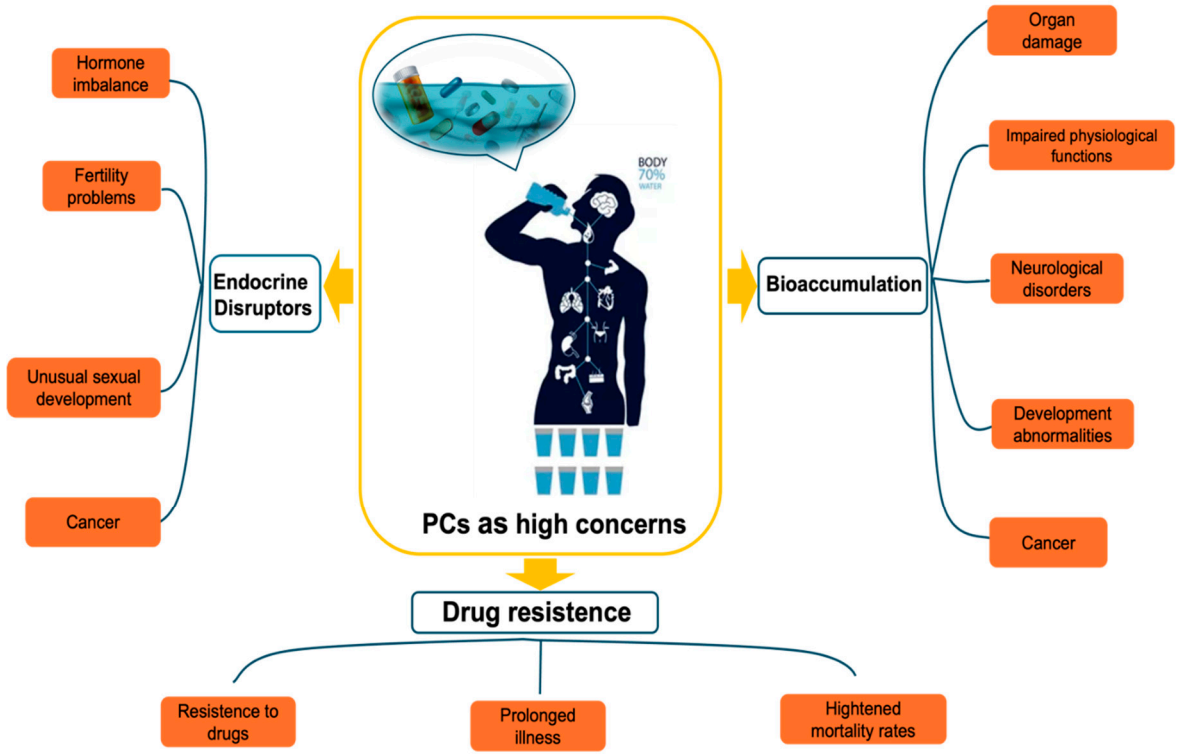

Figure 3. Key health risks linked to micropollutants, modified by [13,89,90].

Exposure to PCs can lead to changes in inherited traits and the behavior of living organisms. One example of this is the transformation of male fish into females due to the presence of estrogen in

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water bodies, which leads to the development of female characteristics [91,92]. Additionally, PCs in drinking water pose significant risks to vulnerable groups such as newborns, the elderly, and individuals with kidney or liver failure. The presence of estrogens in drinking water has also been linked to reduced male fertility and an increased risk of breast and testicular cancers [93–97].

2.3.1. Non-Steroidal Anti-Inflammatory Drugs (NSAIDs)

NSAIDs are found in trace amounts (primarily in nano- and microgram quantities) in various environmental media, including soil, wastewater, surface water, groundwater, and drinking water. Although their concentrations are low, NSAIDs can cause prolonged ecotoxicological effects on the living components of ecosystems [98]. According to Feng et al. (2013), more than 30 million doses of NSAIDs are consumed daily, and this number is rapidly increasing [38]. Research by Selderslaghs et al. (2012) has shown that the presence of diclofenac and ketoprofen in aquatic environments can result in cardiovascular defects and cardiac anomalies in freshwater fish [99].

2.3.2. Antibiotics

The presence of antibiotics in the environment can facilitate the development and spread of antibiotic resistance genes, contributing to a global public health crisis. Antibiotics can disrupt the processes at WWTPs by killing or inhibiting the growth of microorganisms essential for the microbial activity that aids in purification [37]. According to Dolliver and Gupta, antibiotics contaminate groundwater and surface water through leaching and agricultural runoff [100]. The use of antibiotics poses significant health risks, including cardiac arrhythmia, immune system disruption, liver dysfunction, bone marrow suppression, and potential impacts on the food chain [26].

2.3.3. Antidepressants

Lajeunesse and Metcalfe et al. (2010) found through their research that antidepressants and their metabolites are present in surface water, sewage, and even in the effluent from wastewater treatment plants [101,102]. Some of the significant side effects linked to antidepressants include hypoglycemia, both acute and chronic toxicity, growth inhibition in aquatic organisms, and sexual dysfunction [26]. Additionally, there is an increased risk of suicidal thoughts and behaviors among children and adolescents who take these medications. When individuals stop using antidepressants, they may experience discontinuation syndrome, which can mimic the symptoms of their previous depression [103,104].

3. Presence of Micropollutants in the Aquatic Environment

Table 3 presents a comprehensive overview of key MPs, including substances like caffeine, diclofenac, and bisphenol A, and their concentrations across various water types—such as surface water, groundwater, drinking water, and wastewater—and countries. The data illustrate variations in pollutant levels depending on the water type and geographical region, providing insights into regional pollution patterns, potential sources from industries and agriculture, and the effectiveness of wastewater treatment processes. Moreover, the table offers valuable information on the influence of regulatory measures and can help inform strategies aimed at reducing pollution.

Table 3. Micropollutant concentrations across various water types and countries.

Water Type	Micropollutants	Countries	Concentration [ng/L]	References
Surface Water	Caffeine	Germany	65 – 6,798	[105]
		Denmark	65 - 382	[106]
		Korea	268.7	[107]
		China	865	[108]

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	Diclofenac	UK	20 – 91	[109,110]
		Sweden	680	[110]
		Korea	8,8 – 127	[111]
		China	< 147	[112]
	Carbamazepine	USA	6.8	[113]
		Korea	5 - 36	[107]
	Ibuprofen	Germany	60 – 152	[114]
		Korea	11 - 38	[107]
		China	1417000	[115]
	Naproxen	Germany	70	[114]
		Sweden	90 – 250	[116]
		Korea	20 – 483	[107]
		China	< 118	[115]
	Bisphenol A	Europe	10	[117]
		USA	81	[117]
		Korea	4.5 - 61	[118]
Ground Water	Caffeine	USA	290	[119]
		Germany	102	[119]
		China	42.5	[119]
		Italy	84 - 683	[120]
	Ibuprofen	USA	3,110	[121]
		Europe	3 - 395	[115]
	Carbamazepine	Europe	12 - 390	[105]
		USA	42	[122]
	Atrazine	Europe	8 – 253	[105]
	Bisphenol A	Europe	79 – 2299	[105]
		USA	4.1 – 1990	[123]
Drinking Water	Caffeine	Spain	9.10	[124]
		Sweden	5.50	[125]
		USA	52.3	[119]
		Korea	34.3 - 95.5	[126]
		Turkey	3390	[127,128]
		Germany	611	[128]

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	Diclofenac	Japan	16	[129]
		Spain	25	[130]
		Sweden	8	[125]
		France	56	[131]
	Carbamazepine	Japan	25	[129]
		France	41,6	[131]
	Ibuprofen	Japan	6	[129]
		France	14	[131]
		Germany	244	[128]
	Naproxen	France	6	[131]
	Metoprolol	France	1	[131]
	Bisphenol A	Germany	72	[128]
	Caffeine	Europe	3002	[132]
		Korea	60	[133]
	Diclofenac	Europe	174	[132]
		Korea	49	[133]
WWTP Effluent	Carbamazepine	Europe	4609	[132]
		Korea	74	[133]
		China	55	[132]
	Ibuprofen	Europe	2129	[132]
		Korea	75	[133]
	Atrazine	Europe	36.6	[132]
	Bisphenol A	Europe	200	[134]
		China	623.6	[134]

4. Techniques for the Removal of Pharmaceutical Contaminants

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4.1. WWTPs Today

Conventional WWTPs employ a combination of advanced biological, physical, and chemical processes, which are categorized according to their operational principles, applications, and implementation methods to ensure efficient wastewater treatment (Figure 4) [135]. The mechanical stage of a WWTP consists of screening systems, sand traps, and primary clarifiers. In the biological stage, biological reactors are used to further degrade organic matter, followed by sedimentation to separate the biological flocs from the treated water. The chemical stage, which occurs after secondary treatment, involves adding a phosphate precipitant in a final clarifier to aid in the clarification process. The precipitated phosphate then settles with the sludge and is removed. Chlorine is commonly used as a chemical disinfectant in water and wastewater treatment due to its ability to inactivate microorganisms by oxidizing and damaging their cellular structures [136,137]. Some WWTPs may also include a sand filtration stage to further enhance removal efficiency. Finally, the treated and clarified wastewater is discharged into a natural water body [138].

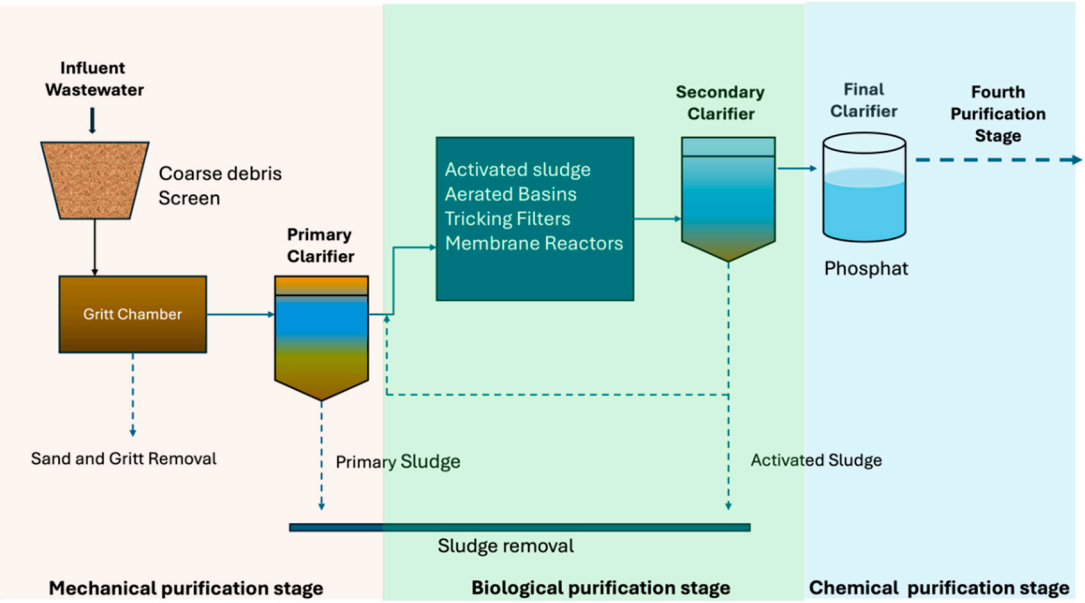

Figure 4. Schematic representation of a conventional wastewater treatment plant, modified by [138,139].

Jelic et al. (2011) conducted a study on the presence of 43 pharmaceuticals in the influent, effluent, and sludge of three WWTPs in Catalonia [140]. These WWTPs primarily utilized mechanical and biological treatment stages, except for one plant, which incorporated a flocculation basin as an additional stage. Of the 43 pharmaceuticals examined, 32 were found in the influent, 29 in the effluent, and 21 in the sludge. Compounds such as diclofenac, carbamazepine, clarithromycin, and sulfamethazine were detected across all three stages. The concentrations of these substances ranged from ng/L in the influent and effluent to ng/g in the sludge. This study, along with others, underscores the importance of introducing a fourth treatment stage in WWTPs globally [141–144]. For pharmaceuticals that are difficult to biodegrade, the traditional three-stage treatment process is often inadequate. Its efficiency is greatly influenced by the physicochemical properties of the compounds and the specific treatment conditions [145]. While the activated sludge process is commonly employed to treat wastewater containing NSAIDs, it has been shown that these drugs are not entirely removed [146].

Ongoing research and development have led to the exploration of various technologies that could serve as a fourth treatment stage in WWTPs [147–149]. These technologies are classified into biological, chemical, and physical processes, with potential combinations of these methods (Figure 5). The following sections provide an overview of these individual processes, emphasizing their functionality and effectiveness in eliminating PCs from wastewater [145,146,150].

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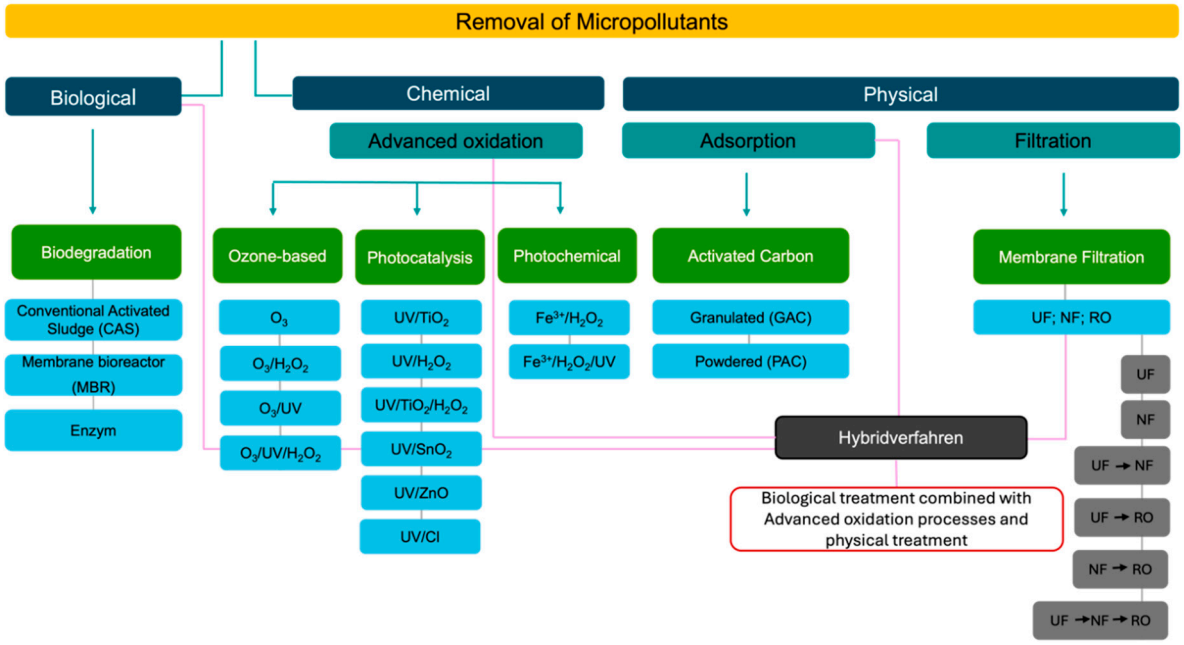

Figure 5. Processes for the removal of micropollutants, modified by [135,151].

Table 4 presents a summary of the advantages and disadvantages of the wastewater treatment processes discussed above. It highlights that achieving the desired effluent quality often requires the coupling of multiple treatment technologies to overcome the limitations inherent in relying on a single treatment method.

Table 4. Advantages and disadvantages of various treatment processes for pharmaceutical removal.

Treatment Processes	Advantages	Disadvantages	References
Conventional biological treatment	- Reduced initial investment - Versatile and straightforward technology - Environmentally sustainable	- Inefficient removal of low-biodegradable pharmaceutical contaminants - Generation of toxic metabolites - Inability to target specific pharmaceutical contaminants - High sludge production	[152–154]
Advanced biological treatment	- Focused removal of contaminants - High adaptability to diverse wastewater characteristics - Space-efficient design - Improved removal of pharmaceutical contaminants - Effective operation at elevated suspended solids concentrations	- High energy and initial investment costs - Membrane fouling issues - Challenges in degrading persistent PCs - Necessitate effective strategies for managing microbial activity	[155–157]

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Advanced Oxidation Processes (AOPs)	- Environmental compatibility - Synergy with other processes (biological or physical treatments) - Rapid processing and high efficiency - Effective in removing a broad spectrum of organic compounds	- Generation of toxic byproducts - High energy and chemical requirements - Limited scalability due to cost and technical constraints - Need for specialized equipment and expertise	[158–160]
Adsorptive treatment	- Low operational costs - Simple operation - Flexibility in using a wide range of adsorbents for specific requirements - Effluent with low dissolved solids	- Adsorbent saturation - Gradual capacity decline after several treatment cycles - Column blockage - Limited selectivity - Challenges in regeneration and production of secondary waste	[161–163]
Membrane technology	- High removal efficiency - Selective separation - Compact design, requiring less space - Versatility (able to treat a wide range of water matrices)	- High installation and material costs - High energy consumption - Membrane fouling issues - Frequent membrane cleaning required - Necessitates brine disposal and toxicity assessment	[159,164,165]

4.2. Biological Treatment Technologies

Biological treatment utilizes the metabolic activity of environmental microorganisms to oxidize and break down organic pollutants in water, transforming them into stable and harmless inorganic substances. This approach provides an effective and sustainable solution for modern water purification [166]. The biological methods used to remove micropollutants in WWTPs can be divided into conventional and advanced processes (Figure 6). Conventional technologies largely depend on microbial activity and require minimal mechanical input. These methods include the activated sludge process, biotrickling filters, biofilm reactors, and nitrification/denitrification systems [167]. On the other hand, advanced biological processes, such as two-phase partitioning bioreactors, membrane bioreactors (MBRs), immobilized cell bioreactors, and moving bed biofilm reactors, utilize enhanced technologies to improve treatment efficiency [168]. Among these, the activated sludge process and membrane bioreactors are the most used biological treatment methods, and they will be explored in more detail in the following section [169].

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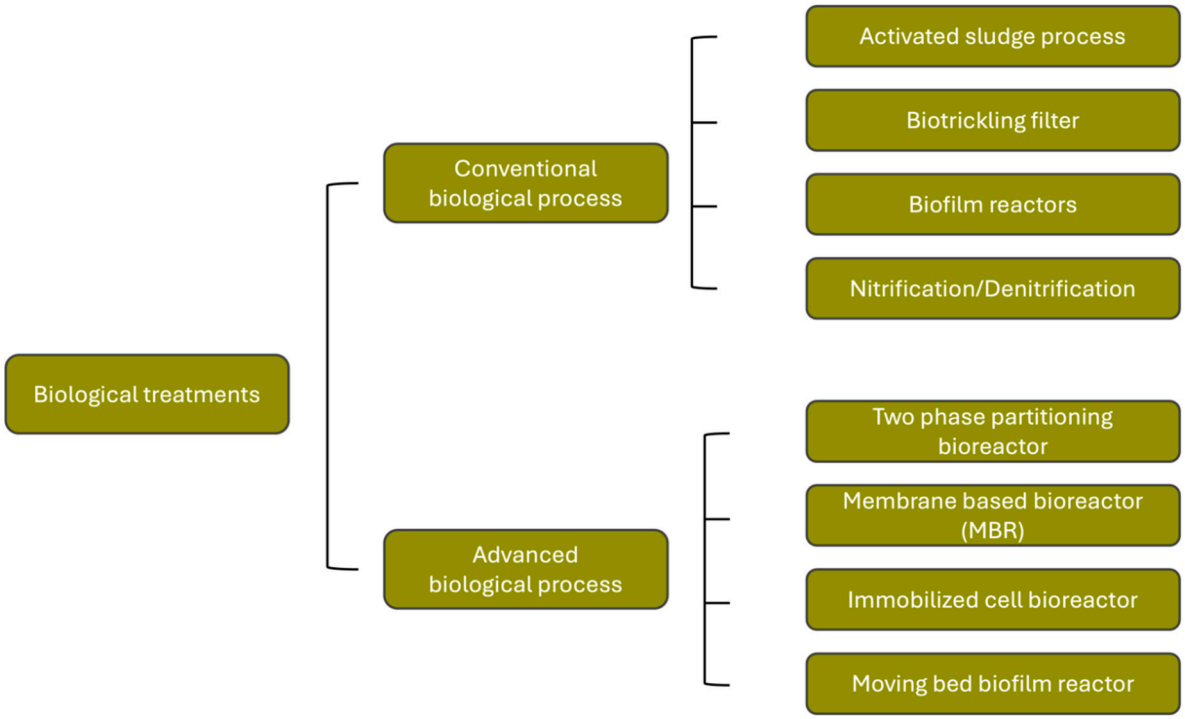

Figure 6. Traditional and advanced biological treatment methods for wastewater treatment plants contaminated with pharmaceutical compounds, modified by [167,168].

4.2.1. Conventional Biological Processes

4.2.1.1. Activated Sludge

The removal of pharmaceuticals entering a WWTP primarily takes place during the biological treatment stage [133,170,171]. It is evident that the removal rates of these compounds can differ significantly between WWTPs, as the effectiveness of removal depends on a variety of factors, including the type of treatment process, the characteristics of the mixed liquor suspended solids (e.g., type of sludge), operational parameters of the WWTP (such as sludge retention time, hydraulic retention time, pH, and temperature), as well as the physicochemical properties of the pharmaceuticals themselves [37]. However, the type of treatment process used in the WWTP and the characteristics of the pharmaceuticals are likely the most significant factors influencing the removal efficiency of these compounds [172,173].

Treatment efficiencies for pharmaceuticals in the biological process of WWTPs can range from 20% to 99% [133,174], with some compounds, like carbamazepine, showing removal rates as low as below 20% [133,172,174,175]. This low removal rate for carbamazepine can be attributed to its persistent nature and water-soluble properties [133]. In Germany, about 68-69% of diclofenac was removed through secondary treatment in WWTPs, and similar results were found in five WWTPs in Ulsan, South Korea [133,176]. Typically, diclofenac removal rates range around 40%, but biodegradation can occur with varying efficiencies depending on the conditions [177]. A study by Sim et al. (2010) found the removal rates of naproxen and gemfibrozil to be 82 ± 20% [178], while Clara et al. (2005) reported a 99% removal of ibuprofen during biological treatment in a WWTP [179].

Behera et al. (2011) studied pharmaceutical removal in five WWTPs in Ulsan, South Korea, and found that WWTPs D and E achieved higher lincomycin removal rates (58% and 74%, respectively), likely due to the presence of anoxic-oxic conditions [133]. WWTP E, which employed the Symbio process, also showed enhanced removal of various pharmaceuticals, including acetaminophen, ibuprofen, ketoprofen, clofibric acid, gemfibrozil, caffeine, atenolol, estriol, and estradiol. The Symbio process creates dual oxic-anoxic zones within sludge flocs by controlling dissolved oxygen, enabling simultaneous nitrification and denitrification. This mechanism likely improves biodegradation,

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consistent with findings by Zwiener and Frimmel, who observed that anoxic-oxic processes enhance diclofenac degradation [180].

Pharmaceutical removal in biological treatment relies on two primary mechanisms: biodegradation and sorption (including absorption and adsorption) [37,173,181]. Biodegradation can occur via co-metabolism, where other substances serve as the primary carbon or energy source, or through microbial metabolism [182]. Sorption efficiency depends on electrostatic interactions [173,181,183] and compound hydrophobicity, with higher hydrophobicity often resulting in increased removal efficiency [174]. For example, Chen et al. (2022) observed strong adsorption of antibiotics like erythromycin and azithromycin during biological treatments, primarily driven by these interaction mechanisms [184]. Conversely, compounds with high water solubility (low hydrophobicity), such as sulfamethoxazole, are expected to exhibit lower sorption and removal rates [173].

The biological treatment of pharmaceuticals can lead to mineralization (conversion into CO_2, water, and inorganic ions), degradation into smaller molecules, or minor structural modifications [185]. The formation of metabolites or biotransformation products depends on the pharmaceutical's nature and the type of microorganisms involved [186]. For example, during the treatment of the X-ray contrast agent iopromide, conventional activated sludge oxidized primary alcohols into carboxylates, while nitrifying activated sludge caused dehydroxylation of side chains, indicating a co-metabolism pathway [187].

4.2.2. Advanced Biological Processes

4.2.2.1. Membrane Bioreactor

A membrane bioreactor (MBR) combines biological treatment through activated sludge with a membrane process for separating liquids from solids. Compared to the conventional activated sludge process (CAS), the integration of microfiltration (MF) or ultrafiltration (UF) membranes offers notable advantages [188]. MBRs occupy less space, produce higher-quality wastewater, and can operate at higher suspended solids concentrations than traditional CAS plants. This single-step process maintains suspended solids concentrations between 8,000 and 12,000 mg/L, while CAS typically operates at around 5,000 mg/L, as higher mixed liquor suspended solids (MLSS) concentrations can lead to settling issues in the sedimentation basin [189].

MBRs use membranes that effectively retain sludge and remove pathogens, leading to cleaner water. Additionally, MBRs support long sludge retention times (SRT), which promote the growth of bacteria that degrade specific pollutants and reduce excess sludge production [188]. CAS is often insufficient for removing organic pollutants due to the quantity and variability of micropollutants [190]. Studies have shown that MBRs can efficiently remove various micropollutants, including pharmaceuticals, with varying degradation rates for compounds like ibuprofen, naproxen, and diclofenac. The microbial community in MBRs plays a crucial role in this process, with specific bacterial groups being more effective in degrading certain micropollutants [188,191,192].

As Mert et al. (2018) [192] and Nghiem et al. (2020) [188] have pointed out, MBRs are a promising alternative for micropollutant removal in water treatment. They offer efficient removal of these pollutants, making them particularly valuable for water reuse and environmental protection. However, MBRs show low efficiency in removing persistent, high molecular weight, hydrophilic organic pollutants [193]. Compounds like Enalapril (a pharmaceutical) and Atrazine (a pesticide) are highly resistant to biological degradation. Membrane bioreactors (MBRs) only achieve about 20% removal efficiency for these compounds due to their inherent stability [193]. To address this, high retention membrane bioreactors (HRMBRs), such as osmotic membrane bioreactors (OMBRs) [194], membrane distillation bioreactors [195], and bio-electrochemical membrane reactors (BEMRs) [196], can be used to enhance the removal of persistent, high molecular weight micropollutants [197].

Research by Song, Luo et al. (2018) has shown that combining membrane distillation (MD) with anaerobic membrane bioreactors (AnMBRs) can achieve almost complete removal of large organics and phosphates, including 26 trace organic contaminants (TrOCs) categorized as hydrophobic (Log

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D > 3.2) and hydrophilic (Log D < 3.2). These TrOCs represent emerging contaminants commonly found in municipal wastewater, such as pharmaceuticals, personal care products, endocrine disruptors, industrial chemicals, and pesticides. While the effectiveness of the AnMBR varied by compound, the addition of the MD process resulted in the complete removal of these contaminants [198].

Cornelissen et al. (2011) investigated the OMBRs, which integrates activated sludge treatment with forward osmosis (FO) membrane separation and reverse osmosis (RO) post-treatment for wastewater reclamation. Their research focused on FO membrane fouling and performance, using different activated sludge in both laboratory and pilot-scale systems. They concluded that the OMBR holds promise as a new development for industrial and municipal wastewater reuse, providing a double barrier against pathogens, organic micropollutants, and particulate matter [199]. Nguyen Hai et al. (2013) also explored the integration of UV oxidation and NF/RO membrane filtration in MBRs for removing several TrOCs. Their study showed that UV oxidation and NF/RO membrane filtration significantly improved MBR performance, achieving high overall removal of hydrophilic and biologically persistent TrOCs [200].

In conclusion, Pathak Van Tran et al. (2020) emphasized the advantages of using HRMBR in water treatment. These combined processes have demonstrated promising results, particularly in enhancing the efficiency of pollutant removal. However, a complete understanding of the nature of pollutants, their interactions, and the practical integration of various technologies is still lacking, and further research is necessary to address these gaps [197].

4.3. Adsorptive Treatment Technologies

4.3.1. Activated Carbon Adsorption

The adsorption process using solid adsorbents has shown significant potential as one of the most efficient methods for treating waters and wastewaters containing pharmaceutical products [201]. Adsorbents are generally classified into two categories: natural and synthetic. Natural adsorbents include materials like charcoal, clays, clay minerals, zeolites, and ores, while synthetic adsorbents are derived from agricultural products, waste, household waste, industrial waste, sewage sludge, and polymeric substances. Each type of adsorbent possesses unique characteristics, such as porosity, pore structure, and the nature of the adsorbent surface [202].

Activated carbon (AC) is the most widely used adsorbent for removing pharmaceuticals from wastewater due to its high adsorption capacity. This is attributed to its extensive porosity, large surface area (often exceeding 1000 m^2/g), and strong surface interactions between pharmaceuticals and the adsorbent surface [203]. Additionally, newer materials like molecular imprinted polymers (MIPs) and magnetic nanoparticles have been explored, though these still lack substantial development and supporting case studies compared to AC [204]. Activated carbon is typically categorized into two types based on particle size: powdered activated carbon (PAC), with particles smaller than 0.2 mm, and granular activated carbon (GAC), which has particles ranging from 0.2 to 5 mm. Its pore structure is further divided into macropores (≥50 nm), mesopores (2–50 nm), and micropores (0.8–2 nm) [205].

The efficiency of adsorption is influenced by various factors, including both the properties of the wastewater and the adsorbent, as well as operational conditions. Key properties of adsorbents that affect efficiency include surface morphology, functional groups, pore size, and the content of ash and minerals [206]. For pollutants, factors such as solubility, molecular size, charge, and structural composition play a crucial role in determining the adsorption effectiveness. Operational conditions that impact the process include the initial concentration of pollutants, the pH of the wastewater, temperature, and the quantity of adsorbent used [207–209]. Table 5 demonstrates the high removal efficiency of certain pharmaceutical compounds from water and wastewater.

Table 5. Removal efficiency of selected pharmaceuticals by activated carbon from various precursors.

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Pharmaceuticals	Water type	Concentration (mg/L)	AC	Removal efficiencies (%)	References
Diclofenac	Various	10-30	AC from cocoa pod husks	76.0-93.6	[210]
Carbamazepine	Wastewater	2	PAC	93	[211]
Naproxen	Wastewater	1-30	PAC	67.2 – 89.2	[212]
Sulfamethoxazole	Distilled	50-500	AC	90	[213]
Penicillin G	Distilled	50-1000	AC	12.0 -78.3	[214]
Atenolol	Various	5-900	GAC	88	[215]

The AC/GAC process is limited by the specific properties of AC material and the contact time with the water, making it less adaptable [216]. Another drawback of AC is its declining removal performance as the volume of treated wastewater increases, requiring the AC to be replaced or regenerated to restore its effectiveness [217]. Additionally, the regeneration and activation of AC come with high costs, and an extra washing stage is necessary to eliminate chemical agents used in the process [218]. The frequent need for regeneration, replacement, and disposal of AC raises environmental concerns. In industrial settings, the presence of background organic matter can reduce the efficiency of the adsorption process and increase material consumption [219]. High levels of dissolved organic matter in wastewater (e.g., >20 mg/L) compete for adsorption sites on the AC, thus reducing the removal efficiency of pharmaceutical pollutants. Despite these limitations, AC remains a viable option for removing pharmaceutical compounds from effluents if the wastewater is pre-treated to reduce organic matter and suspended solids substantially [205].

The removal of PCs via the AC process is influenced by both the dose of AC and the contact time. Higher initial concentrations of pollutants typically require higher carbon dosages to achieve effective removal [162]. However, the required dosage varies between pharmaceuticals. For example, strongly adsorbing compounds like diclofenac can be removed by over 90% with low doses of PAC (5–10 mg/L) [220]. In contrast, compounds that adsorb weakly, such as sulfamethoxazole, require significantly higher PAC dosages, typically ranging from 10–50 mg/L or even up to 100 mg/L [221].

4.4. Physical Treatments Technologies

4.4.1. Membrane Technology

Membrane filtration is increasingly gaining attention in both research and industrial applications for drinking water and wastewater treatment, particularly in water reuse scenarios [222–224]. Recent studies demonstrate promising and feasible results when integrated systems are applied, showing improvements over conventional treatment technologies. Membranes act as physical barriers that either reject or reduce the flux of substances, effectively separating them from the rest of the stream. Membranes are classified into four types based on particle size: microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), and reverse osmosis (RO) [225,226].

4.4.1.1. Removal Mechanism of PCs by Membrane Separation Processes

The removal mechanisms of PCs during membrane filtration are influenced by a combination of their properties, solute parameters, and the characteristics of the membranes used. Key properties of PCs, such as molecular weight, size, hydrophobicity or hydrophilicity, charge, and chemical structure, significantly affect their rejection by membranes. Similarly, membrane properties such as molecular weight cut-off (MWCO), pore size, surface charge (zeta potential), hydrophobicity or hydrophilicity (measured via contact angle), and surface morphology (measured via roughness) play a crucial role in facilitating the rejection of contaminants [3,227].

Membrane selectivity is governed by several mechanisms, including size exclusion, electrostatic interactions, hydrophobic interactions, adsorption, diffusion, solute-solute interactions, and fouling.

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Simon et al. [228] studied the adsorption of ibuprofen by NF and RO membranes and found that this phenomenon is closely related to electrostatic repulsion between the pollutant and the membrane material, as well as the pH of the solution. When the pH drops below the ibuprofen's pKa (acid dissociation constant), electrostatic repulsion decreases, as the membrane acquires a positive charge, thus facilitating the drug's adsorption onto the membrane, which has a negative surface charge. For instance, ibuprofen rejection by the loose NF membrane (NF 270) decreases from approximately 60% at pH levels above its pKa (4.4) to below 40% at pH levels below its pKa. This finding aligns with Shanmuganathan et al. (2017) [229], who observed that NF and RO membranes achieve higher removal rates for ionic substances (97%) compared to nonionic compounds (82%).

In further studies, Licona et al. (2018) found a strong correlation between PCs' molecular weight (MW) and hydrophobicity in their rejection by NF and RO membranes. They identified size exclusion and adsorption as the main removal mechanisms, with negatively charged PCs, such as ibuprofen, dipyrone, and diclofenac, achieving higher removal efficiencies (>95%) at 20 bar and pH 7 due to electrostatic repulsion. On the other hand, neutrally charged PCs, such as caffeine and acetaminophen, showed lower rejection rates, with caffeine removed at below 95% and acetaminophen below 90% [230]. Similarly, Albergamo et al. (2019) found a complex relationship between the size, hydrophobicity, and chemical structure of PCs and their removal efficiency in RO membranes. While size exclusion predominantly influences the removal of neutral-hydrophilic and anionic PCs, hydrophobic interactions play a significant role in the passage of moderately hydrophobic PCs, resulting in lower removal efficiencies. The authors attributed this lower efficiency to the affinity between hydrophobic structures, such as aromatic rings and hydrocarbon chains, and the active layer of RO membranes [231]. Both studies emphasize the significant role of size and hydrophobicity in PCs removal, with electrostatic repulsion enhancing the rejection of negatively charged PCs.

4.4.1.2. MF and UF

The pore size of membrane filtration (MF and UF) significantly influences their efficiency in removing PCs. MF and UF membranes are considered less restrictive compared to other membrane separation techniques. Their pore diameters range from 0.1 to 0.005 μm for UF and 5 to 0.1 μm for MF. Since most PCs have molecular sizes smaller than these pore sizes, MF membranes are not effective in removing these substances from water. To enhance removal, a pretreatment process, such as coagulation or adsorption, is often used to increase the size of the pollutants, making them easier to filter. For even better performance, MF and UF membranes are often used in conjunction with reverse osmosis (RO), as pretreatment can improve removal efficiency and reduce membrane fouling [3].

In a study by Comerton et al. (2007), the removal of 22 endocrine-disrupting compounds (e.g., bisphenol A, estriol, estrone, 17α-estradiol, 17β-estradiol) and pharmaceuticals (e.g., acetaminophen, caffeine, carbamazepine, gemfibrozil, sulfamethoxazole) was tested using Polysulfone UF, polyamide NF, and polyamide RO membranes. They found that NF and RO membranes were more effective than UF membranes in removing the substances. However, adsorption played a more significant role in removing these compounds using UF membranes. Adsorption was strongly correlated with the compound's log Ko/w (octanol-water partition coefficient) and membrane water permeability, and moderately correlated with water solubility. The study showed that adsorption increases with compound hydrophobicity and decreases with compound water solubility. The UF membrane exhibited the highest level of adsorption, followed by NF and RO membranes, as membranes with larger pores allow more access to the membrane's internal adsorption sites [232].

Similar findings have been reported in other studies. While the removal efficiency of MPa using UF may be relatively low, hydrophobic adsorption remains the primary mechanism for removal, and it is influenced mainly by the hydrophobicity of the pollutants [233,234]. Jermann et al. (2009) pointed out that while adsorption onto some UF membranes can help retain pollutants during the initial filtration period, it cannot be considered a long-term removal mechanism [234]. Yoon et al. (2006) confirmed this by stating that once steady-state operation is reached, size exclusion becomes the

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dominant mechanism in retaining EDCs and pharmaceutical and personal care products (PPCPs) by UF membranes [235,236]. These observations highlight the importance of considering both size exclusion and adsorption mechanisms in membrane filtration for effective removal of pollutants. The summarized results are presented in Table 6.

Table 6. An overview of membrane processes using organic ultrafiltration membranes for micropollutant removal.

Membrane Processes	Matrix	Target MP (PPCPs)/ initial concentration	Removal efficiencies (%)	References
UF (10,000 Da)	Two natural water sources (drinking water source and MBR effluent) and lab-scale water (control sample)	22 EDCs and PCs		[232]
UF (2000-20,000 Da)	Secondary effluent spiked with various compounds	11 contaminants: Acetaminophen, metoprolol, caffeine, antipyrine, sulfamethoxazole, flumequine, ketorolac, atrazine, isoproturon, hydroxybiphenyl, diclofenac	<50%, except for hydroxybiphenyl	[233]
UF (8000 Da)	Synthetic (model) water and natural freshwater sources	52 EDCs/PPCPs	Up to 80% removal efficiency for hydrophobic compounds	[236]
UF (100 kDa)	Synthetic (model) water	Estradiol and ibuprofen	25% ibuprofen, 80% estradiol	[234]

4.4.1.3. NF and RO

NF and RO membrane processes, particularly those using polymeric membranes, have been extensively studied for the removal of micropollutants from water and wastewater. These processes are highly effective at retaining dissolved salts and solutes, and are suitable for most micropollutants with molecular weights ranging from 200 to 400 Da [223,237]. As the utilization of NF and RO for water treatment, wastewater treatment, and desalination continues to grow, these processes are becoming more integral despite their high-pressure requirements [238].

The primary removal mechanisms for micropollutants in RO and NF membranes include size exclusion, electrostatic repulsion, and sometimes, hydrophobic adsorption. These mechanisms differ significantly from those in ultrafiltration (UF) membranes, where adsorption to the membrane surface is the predominant removal mechanism. In contrast, NF membranes mainly operate by size exclusion, allowing the rejection of larger molecules while permitting smaller ones to pass through [3].

Several factors influence the removal efficiency of micropollutants in NF and RO membranes. Ionic strength, hydrophobicity, and the conditions of the feed solution, such as pH, are all known to

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affect the rejection rates of contaminants. The ionic rejection is largely due to electrostatic repulsion between charged compounds and the surface charge of the membrane. Uncharged compounds are generally rejected via size exclusion. The surface charge of polymeric NF membranes is typically negative, which repels negatively charged species (anions) while attracting positively charged species (cations) to maintain electroneutrality. As a result, NF membranes tend to reject divalent anions more effectively than monovalent ions, allowing the latter to pass through more easily into the permeate.

A review of available studies (as shown in Table 7) indicates that NF membranes generally exhibit very low MWCO, which makes them highly efficient for rejecting larger and more complex micropollutants. However, more research is needed to further optimize and understand the removal mechanisms and the effects of varying feed conditions on the performance of these membranes for micropollutant removal.

Table 7. Rejection of selected pharmaceuticals by polymeric NF membranes.

PCs	pH	Membrane Name	Materials	MWCO (Da)	Removal efficiencies (%)	References
PARA*	6.5	NF270	Polyamide	200-300	44	[239]
PARA	7	NF 200	Aromatic polyamide	~ 300	22	[237]
IBU*					89	
PARA	6 - 7	NF 90		~ 200	75	
IBU					96	
PARA	7.4 - 7.6	NF 270	Thin aromatic or semi-aromatic polyamide	200-300	0	[240,241]
IBU					99	
DIC*					95	
PARA		NF 90	Polyamide	~200	99	
IBU					99	
DIC					90	
PARA	7	NF 270	Polypiperazine with polymeric active layer	220	31	[241]
		NF 90	polyamide supported by a Polysulfone	102	102	
DIC	8	FM	Hydrophilic Polyethersulfone	1000	61	[242]
IBU	8	NP010			55	
PARA	12	NF 50	Sulfonated Polyethersulfone	1000	36.16	[224]
DIC	3				99.74	
IBU	6 - 7				80.54	

*PARA: Paracetamol (151.16 g/mol); *DIC: Diclofenac (206.29 g/mol); *IBU: Ibuprofen (294,18 g/mol).

The use of loose NF membranes with larger pore sizes for micropollutant removal, including PCs, has been studied with promising results. These membranes can reject micropollutants via mechanisms such as absorption and Donnan exclusion, with minimal fouling. Optimization of parameters like pH, charge effects, and feed polarization has been shown to enhance rejection efficiency. For example, sulfonated polyethersulfone NF membranes have demonstrated improved rejection of micropollutants through dissociation of surface groups such as sulfonated or carboxyl acids. However, the efficiency of these membranes can decrease when exposed to a mixture of organic micropollutants, highlighting the complexity of membrane performance in real-world conditions. Hydrophilic functional groups like sulfone, carboxyl, hydroxyl, and amine groups, present on polyethersulfone membranes, assist in rejection by forming bonds with the pollutants, thereby enhancing retention [224,243,244].

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Regarding inorganic membranes, research on their ability to remove pharmaceutical micropollutants from water remains limited, although a few studies have explored their potential. One such study, conducted by the Institute of Bioprocess Engineering and Pharmaceutical Technology (IBPT) at the University of Applied Sciences Mittelhessen, investigated ceramic UF and NF membranes made from materials such as Al_2O_3 and TiO_2. The study focused on the removal of diclofenac and ibuprofen, with removal rates for diclofenac reaching up to 40%, and ibuprofen reductions ranging from 32% to 47%. Since the molecular weights of these pharmaceuticals are smaller than the membrane's cutoff, electrostatic interactions are believed to play a significant role in the retention of these compounds. This study suggests that ceramic membranes, particularly those composed of Al_2O_3 and TiO_2, hold promise for the removal of pharmaceutical micropollutants from water [135,245–247].

Additionally, research by Radeva et al. (2021) demonstrated that coating ceramic membranes with polyelectrolytes could increase the retention of diclofenac by up to 84%. However, factors such as the long-term stability of the coated membranes and the influence of different pH levels were not explored in detail, indicating areas for future research. The studies on ceramic membranes for pharmaceutical separation are summarized in Table 8. These studies contribute to the understanding of inorganic membranes as a viable alternative for the removal of pharmaceutical micropollutants from water, though more work is needed to optimize and stabilize these systems in real-world applications [247].

Table 8. Rejection of selected pharmaceuticals by inorganic membranes.

Membranes	MWCO (Da)	PCs	Removal efficiencies (%)	References
TiO_2	200	41 organic compounds (PCs e.g. IBU, DIC and CARBA*)	95-100	[245]
LC1/LC2*	630/440	PCs e.g. SUL* and CBZ	50-80	[246]
TiO_2 (UF-Membrane)	3,0 nm	DIC and IBU	32 - 47	[135]
Al_2O_3/ LBL Coating with Polyelectrolytes (polystyrene sulfonic acid)	~ 200	IBU, DIC, SUL, Clofibric Acid	56 % für SUL, up to 84 % für DIC	[247]

*CARBA: Carbamazepine; *SUL: Sulfamethoxazole; *LC1 and LC2 manufactured with active layers of TiO_2/ZrO_2.

NF is a highly effective method for separating organic substances, including pharmaceutical residues, from WWTPs. NF membranes can retain molecules with a molar mass starting from approximately 200 g/mol, which aligns well with the size of most pharmaceutical pollutants. This makes NF an ideal technique for removing a wide range of pharmaceutical micropollutants from water.

Inorganic or ceramic membranes are particularly promising for NF applications due to their superior durability compared to polymeric membranes. These membranes offer excellent chemical and thermal stability, making them more resilient in harsh conditions. Furthermore, ceramic membranes exhibit better fouling resistance, which reduces the frequency of cleaning and extends the service life of the system. This robustness makes ceramic membranes an excellent choice for long-term water treatment processes, particularly when dealing with complex and challenging wastewater streams, such as those from WWTPs [248].

Incorporating inorganic membranes into NF systems could improve the efficiency and sustainability of water treatment, especially in the removal of persistent pharmaceutical pollutants.

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Their longevity and reduced maintenance needs can lead to more cost-effective and reliable water treatment solutions, particularly in industrial and municipal applications

4.5. Chemical Treatment Technologies

4.5.1. Oxidative Treatment Technologies for Pharmaceutical Contaminants

4.5.1.1. Ozonation

Ozonation is a process in which ozone is introduced into water, typically by bubbling it through a sparger at the bottom of a tank. Ozone functions as a potent oxidizing agent, engaging in direct reactions or through a series of oxidative radical processes. When acting alone, ozone is effective in oxidizing specific organic compounds [249]. However, in the presence of water, it can react with hydroxide ions to produce hydroxyl radicals (HO•), which are less selective but significantly more powerful oxidants [250].

Studies have demonstrated that ozone can effectively target electron-rich pharmaceutical aromatic compounds such as sulfamethoxazole, ciprofloxacin, carbamazepine, azithromycin, clarithromycin, diclofenac, erythromycin, and metoprolol [221,251–253]. On the other hand, hydroxyl radicals (HO•) operate more rapidly and non-selectively, enabling the oxidation of a wide array of pharmaceuticals, including those that are resistant to ozone under alkaline conditions, such as primidone, loperamide, cephalexin, and penicillin [251,253]. Table 9 illustrates a list of pharmaceuticals successfully degraded by ozonation.

Due to its strong oxidation potential, which exceeds that of many traditional oxidants, ozone has become a favored method for tertiary treatment of wastewater containing pharmaceuticals. Nonetheless, a significant operational challenge when employing ozonation in wastewater treatment is the abundant presence of organic carbon and other oxidizable substances. This necessitates the use of higher ozone quantities to achieve complete treatment of typical sewage [254]. A notable drawback of ozonation is the formation of potentially harmful by-products, which require additional filtration for removal. While increasing ozone doses can improve the removal of the targeted compounds, it also leads to higher operational costs and a greater likelihood of producing toxic by-products, such as bromate [219,255]. According to Östman et al., ozone was found to be less effective at removing pharmaceuticals compared to GAC [256].

Table 9. Ozonation-based removal of selected pharmaceuticals.

Applied treatment (concentration and duration)	Pharmaceuticals	Elimination efficiencies (%)	References
O_3 (5 mg/L, 15 min)	Carbamazepine	>90	[8]
	Diclofenac	>90	
	Metoprolol	80-90	
	Trimethoprim	>90	
O_3 (n/a, n/a) *	Ibuprofen	83	[257]
	Diclofenac	99	
	Carbamazepine	80	
O_3 (33 mg/L, 20 min)	Tetracycline	95	[258]

*n/a: not available.

4.5.1.2. Advanced Oxidation Processes (AOPs)

Advanced oxidation processes (AOPs) generate hydroxyl radicals, which oxidize organic compounds, including pharmaceuticals, transforming them into more stable and less toxic by-products. AOPs are increasingly employed for wastewater treatment, where they oxidize organic pollutants into CO_2 and H_2O. Various methods are used to produce hydroxyl radicals (-OH),

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including ozone (O_3)-based AOPs [259], hydrogen peroxide (H_2O_2)-based approaches [260], heterogeneous photocatalysis [261–264] , sonochemical and electrochemical processes (EAOPs) [265], and combinations of these techniques. These processes aim to effectively eliminate pollutants. Table 10 highlights recent studies on the development and application of AOPs for wastewater treatment and the removal of micropollutants, mostly pharmaceuticals.

Table 10. The most widely used types of advanced oxidation processes for micropollutant removal.

AOPs-Type	Micropollutants	Removal efficiencies of PCs (%)	References
Peroxone	Pharmaceuticals, pesticides and beta-blockers	97-100	[266]
UV/H_2O_2	anticancer drug fluorouracil (5-FU)	>99	[260]
Photo-Fenton	pharmaceuticals, corrosion inhibitors and biocides/pesticides	97- 98	[264]
Electrochemical Oxidation	antibiotics e.g. Ofloxacin	~ 90	[267]
TiO_2 -solar photocatalysis	Pharmaceuticals, Diclofenac	~90	[263]

Gahrouei et al. (2024) present significant findings on the effectiveness of various AOPs for removing antibiotics from water. They highlight that AOPs, including ozonation, photo-Fenton processes, UV/H_2O_2, TiO_2 photocatalysis, and sonolysis, have proven successful in degrading antibiotics like ciprofloxacin, metronidazole, and sulfamethoxazole, achieving removal rates of up to 90% under optimal conditions. The specific removal efficiency varies based on the antibiotic type and operational parameters used in the AOP. While these processes effectively degrade antibiotics, the potential formation of toxic byproducts during treatment raises significant concerns. The authors stress the importance of considering the nature and toxicity of these byproducts when evaluating the overall efficacy of AOPs. They also call for further research, including cost analyses and pilot-scale studies, to better understand antibiotic removal dynamics in complex water matrices and bridge the gap between laboratory results and practical wastewater treatment applications [268].

Sturm et al. (2022) investigated the removal of 10 PCs, including ibuprofen, diclofenac, carbamazepine, metoprolol, and sulfamethoxazole, at a tertiary WWTP in Landau. They compared two advanced treatment methods: (1) AOP using UV and H_2O_2, and (2) GAC. The average removal efficiencies for micropollutants were 76.4 ± 6.2% for AOP and 90.0 ± 4.6% for GAC. However, GAC performance declined over time as the material became saturated, dropping from 97.6% in the first week to 80.7% by week 13 after processing 2,184 bed volumes. For AOP, optimizing UV and H_2O_2 doses significantly improved performance, achieving a removal efficiency of 97.1% with 40 ppm H_2O_2 and 10 kJ/m² UV. The adaptability of the AOP process to real-time water quality changes, its modular design, and the potential for reusing hydrogen peroxide in secondary treatment stages make it a promising option for enhancing the sustainability of wastewater treatment systems [217] .

4.5. Hybrid Technologies

Hybrid technologies combine two or more conventional or advanced treatment methods to achieve maximum, or even complete, removal of micropollutants. The need for hybrid systems arises because no single treatment technology appears capable of ensuring high removal efficiency for all parent compounds and their transformation products. Therefore, the degradation of persistent pollutants, such as pharmaceuticals, can be enhanced by combining processes to take advantage of synergistic effects and the strengths of individual methods.

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Recent applications of hybrid membrane/adsorption processes have increased, particularly the combination of AC adsorption with MF or UF [269]. This approach effectively removes pollutants by combining AC's adsorption and biodegradation properties with the membrane's particle filtration capabilities. The hybrid system can be designed in three main ways: membrane filtration followed by AC adsorption, AC adsorption followed by membrane filtration, or both processes operating together in a single tank.

When membrane filtration is used before AC adsorption, it removes particles larger than the membrane pores, reducing clogging and minimizing backwashing frequency in the AC filter, which in turn enhances its performance. Baresel et al. (2019) reported that this hybrid system effectively removed micropollutants to below detection limits, achieving removal rates of 90-98% [270]. However, a challenge is that some AC fines may be carried over with the treated water, necessitating an additional post-treatment separation process [271]. In contrast, more studies have focused on AC adsorption followed by membrane filtration, which helps reduce membrane fouling by removing fouling agents and extending membrane lifespan [272,273]. This approach also improves permeate flux [274], reduces transmembrane pressure (TMP), and lowers energy consumption [275]. Both GAC [276–278] and PAC [279,280] have been used for this pretreatment. However, GAC requires regular backwashing to prevent blockages, and its continuous use can promote microbial growth, potentially extending its contaminant removal effectiveness [281] .

MBRs combined with nanofiltration NF or RO systems significantly enhance the removal of microorganic contaminants. Dolar et al. (2012) found that MBR + RO systems can nearly eliminate these contaminants [282]. MBRs are particularly effective for removing degradable hydrophilic contaminants through biological processes [193], while NF and RO primarily target hydrophobic contaminants via electrostatic interactions and resistance effects [283]. The integration of MBRs with NF/RO technologies effectively removes both hydrophilic and hydrophobic contaminants from wastewater [193,283] . In a long-term study, Melo-Guimarães et al. demonstrated that a system combining UF, AS, and flocculation (FC) was more effective at removing micro-organic contaminants, such as drugs and pesticides, than single-method approaches. AS was particularly effective for removing acidic drug compounds, while UF targeted phenolic chemicals. FC assisted AS in removing acidic drugs but did not enhance UF's efficiency [284].

Moreover, Asheghmoalla et al. (2024) reviewed integrated and hybrid processes for microplastic (MP) removal from actual wastewater. They highlighted enhanced MBR systems with PAC and hybrid Moving Bed Biofilm Reactor (MBBR-MBR) systems as promising advancements. They also emphasized the need for more research on the performance of these integrated and hybrid technologies in real-world wastewater, which would provide better insights into their feasibility and effectiveness at a large scale [285].

5. Conclusions

The widespread presence of pharmaceuticals, personal care products, industrial chemicals, microplastics, and EDCs in water, at concentrations ranging from ngL^{-1} to μgL^{-1}, poses a significant risk to both human health and aquatic ecosystems. This issue is exacerbated by the diverse sources and complex physicochemical properties of organic micropollutants, alongside the inability of conventional water and wastewater treatment systems to effectively eliminate these contaminants. As a result, water management authorities are increasingly turning to alternative and advanced treatment technologies. Of particular concern are pharmaceutical contaminants, which are commonly found in hospital, household, and industrial wastewaters, as well as in natural water bodies. The detection of pharmaceutically active compounds in aquatic environments has led to a surge in research into effective removal methods, including MBRs, ozonation and other AOPs, physical separation techniques, adsorption using activated carbon, and membrane filtration processes. These methods have shown promise in mitigating pharmaceutical pollution in water treatment.

Given the persistence and low biodegradability of pharmaceuticals, a single treatment approach is insufficient for complete removal. This highlights the need for advanced and hybrid treatment systems. Membrane technologies, including RO, NF, and UF, are recognized as highly effective for

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removing PCs from water. The efficiency of these processes is influenced by factors such as the physicochemical properties of the PCs, operating conditions, and membrane characteristics. Studies have shown that optimal removal typically occurs at pH values above the pKa of the compound, likely due to electrostatic repulsion. Additionally, the exclusion phenomenon, which is based on molecule size, plays a crucial role in removing pharmaceuticals. Membrane technologies also offer the advantage of adaptability, allowing them to be integrated with other treatment processes such as biological treatment and/or adsorption, enhancing their overall efficiency.

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Abbreviations

The following abbreviations are used in this manuscript:

AC	Activated Carbon
AnMBRs	Anaerobic Membrane Bioreactors
AOP	Advanced Oxidation Products
BEMRs	Bio-electrochemical Membrane Bioreactors
CAFOs	Concentrated Animal Feeding Operations
CAS	Conventional Activated Sludge
ECs	Emerging Contaminants
EDCs	Endocrine Disrupting Chemicals
FO	Forward Osmosis
GAC	Granular Activated Carbon
HRMBRs	High Retention Membrane Bioreactors
HRMBRs	High Retention Membrane Bioreactors
KOW	Octanol-Water Partition Coefficient
MBR	Membrane Bioreactor
MD	Membrane Distillation
MF	Microfiltration
MIPs	Molecular Imprinted Polymers
MLSS	Mixed Liquor Suspended Solids
MW	Molecular Weight
MWCO	Molecular Weight Cut-Off
NF	Nanofiltration
NSAIDs	Non-Steroidal Anti-Inflammatory Drugs
OMBRs	Osmotic Membrane Bioreactors
PCs	Pharmaceuticals Contaminants
PAC	Powdered Activated Carbon
PhACs	Pharmaceutically Active Compounds
PPCPs	Pharmaceutical and Personal Care Products
PCPs	Personal Care Products
PKa	The Acid Dissociation Constant
RO	Reverse Osmosis
SRT	Sludge Retention Time
TMP	Transmembrane Pressure
TrOCs	Trace Organic Contaminants
UF	Ultrafiltration
WWTPs	Wastewater Treatment Plants

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