

Review

The Occurrence of Micropollutants in the Aquatic Environment and Technologies for Their Removal

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Abstract: The presence of micropollutants in aquatic environments is an increasing global concern due to their persistence and potential harmful effects on aquatic organisms. Among the most concerning of these micropollutants are microplastics, pharmaceutical compounds, personal care products, and industrial chemicals, posing a significant threat to human health and aquatic ecosystems. This issue is further exacerbated by the diverse sources and complex physicochemical properties of micropollutants, as well as the inability of conventional water and wastewater treatment systems to effectively remove these contaminants. The removal of micropollutants is therefore becoming increasingly important, leading to extensive research into various physicochemical, biological, and hybrid treatment methods aimed at minimizing their environmental impact. This review examines the classification, occurrence, and associated environmental and health risks of commonly detected micropollutants 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: micropollutants removal; microplastics; pharmaceutical contaminants; advanced wastewater treatment processes

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1. Introduction

In recent decades, the presence of micropollutants 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 (PCs), endocrine-disrupting chemicals (EDCs), personal care products (PCPs), industrial chemicals, microplastics (MPs), 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 micropollutants 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]. Micropollutants are often found in water at low concentrations, ranging from nanograms per liter (ng/L) to micrograms per liter ($\mu\text{g/L}$), 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 micropollutants 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 presence of micropollutants 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].

A precautionary approach is essential to managing micropollutants to protect ecosystems and human health. The EU Water Framework Directive (WFD) and Marine Strategy Framework Directive (MSFD) established legally binding regulations, prohibiting deterioration and setting environmental quality standards for 45 priority substances across EU [16], as defined by Directive 2008/105/EC [17] and later amended by Directive 2013/39/EU [18]. Among these, 13 are persistent organic pollutants, whose manufacture and use are already banned or strictly regulated under Regulation (EC) No. 805/2004 [19]. Additionally, The European Union's water policy, initiated by Directive 2000/60/EC, provided a framework to address high-risk substances [20] and the 2015/495/EU decision outlined measures for eliminating hazardous substances such as 17- α -ethinylestradiol (EE2), triallate, 17- β -estradiol (E2), oxadiazon, diclofenac, 2,6-di-*tert*-butyl-4-methylphenol, macrolide antibiotics, methiocarb, neonicotinoids, 2-ethylhexyl-4-methoxycinnamate, and estrone (E1) [21–23]. Some regulations are there for the presence in water for compounds such as pesticides, lindane, nonylphenol, and synthetic hormones [24]. Meanwhile, other harmful substances like ethoxylates and nonylphenol have already been regulated in Canada [25]. 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 [26].

Given their significance and the many ecological risks they pose, this review focuses on the removal of micropollutants, specifically pharmaceutical (PCs) and microplastic (MP) contaminants, 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 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

Micropollutants comprise a wide range of emerging contaminants, which can be categorized into several groups such as pharmaceuticals, microplastics, personal care products, steroid hormones, pesticides, and industrial chemicals. A comprehensive list of 242 chemicals is provided in the EU FP7 project [27], with approximately 70% consisting of pharmaceuticals and personal care products, while the remaining 30% includes industrial agents such as perfluoro compounds, pesticides, herbicides, and food additives. Each

group has its own distinct characteristics and applications. Table 1 summarizes the primary sources of these major categories of micropollutants in aquatic environments.

Table 1. Categories and primary sources of micropollutants in the aquatic environment.

Categories	Key Subcategories	Primary Sources	References
Pharmaceuticals	NSAIDs *, lipid regulators, antibiotics, β -blockers, contrast media, and anticonvulsants	Wastewater from domestic households, effluents from hospitals, pharmaceutical production plants, aquaculture, and concentrated animal feeding operations (CAFO) *	[2,3,27–29]
Personal care products	Fragrances, disinfectants, and insect repellents	Household sewage (from activities such as bathing, shaving, and using sprays)	[2,3,30–32]
Steroid hormones	Estrogens	Wastewater from domestic households, aquaculture, and CAFO *	[3,33–35]
Pesticides	Insecticides, herbicides and fungicides	Garden runoff and agriculture	[1–3,36–38]
Industrial chemicals	Plasticizers, fire retardants	Domestic wastewater generated from the leaching of materials and industrial discharges	[1,2,39,40]
Microplastics	Microfibers, plastic pellets, synthetic fibers	Domestic wastewater resulting from urban runoff	[41–44]

* NSAIDs: non-steroidal-anti-inflammatory drug. * CAFO: concentrated animal feeding operation.

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 [45]. 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 [46]. 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 [47–50]. Figure 1 illustrates the global distribution of pharmaceutical pollutants across five geopolitical regions.

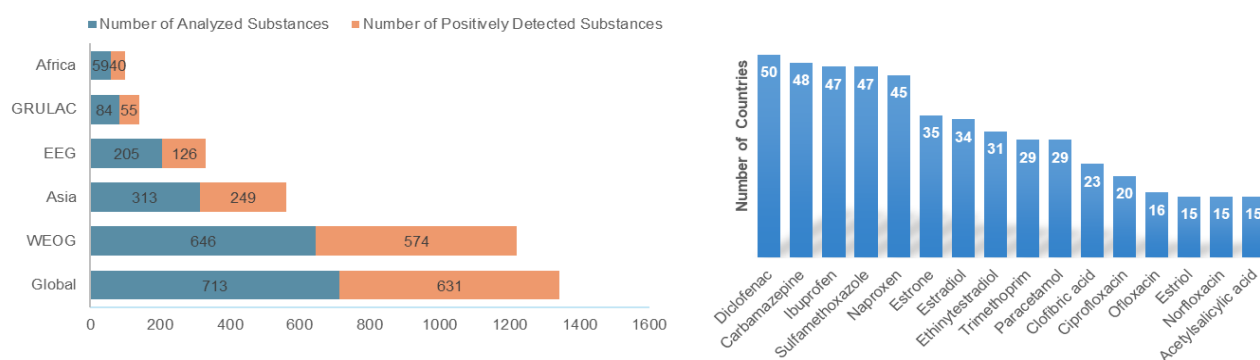


Figure 1. The global occurrence of pharmaceuticals presented on the basis of five geopolitical regional groups: Africa, Group of Latin American and Caribbean States (GRULAC), Asia-Pacific, Eastern European Group (EEG), and Western European Group (WEOG) [51,52].

Micropollutants (PCs) 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 [45,53–55]. 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 [56]. Table 2 provides an overview of key PCs and other micropollutants along with their physicochemical properties. The pharmaceutical categories include analgesics and anti-inflammatories, antidepressants, antibiotics, antivirals, anticoagulants, sedatives, cardiovascular drugs, and more [36].

Table 2. Physicochemical properties of pharmaceutical pollutants [2,56–69].

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 ₉ H ₈ O ₄	280	3.5	1.2	Negative
	Diclofenac	C ₁₄ H ₁₁ Cl ₂ NO ₂	296.2	4.91	4.51	Negative
	Ibuprofen	C ₁₃ H ₁₈ O ₂	206.3	4.15	4.51	Negative
	Paracetamol	C ₈ H ₉ NO 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 ₃ O ₃ S	253.279	5.6–5.7	0.89	Negative
	Erythromycin	C ₃₇ H ₆₇ NO ₁₃	733.93	8.88	3.06	Neutral
	Trimethoprim	C ₁₄ H ₁₈ N ₄ O ₃	290.32	7.12	0.73	Neutral
	Metronidazole	C ₆ H ₉ N ₃ O ₃	171.2	2.5	−0.1	Negative
	Ciprofloxacin	C ₁₇ H ₁₈ FN ₃ O ₃	331.3	6.1; 8.7	−1.70	Neutral
	Oxytetracycline	C ₂₂ H ₂₄ N ₂ O ₉	496.9	3.2; 7.46; 8.9	−1.12	Neutral/Zwitterion
Anticonvulsants	primidone	C ₁₂ H ₁₄ N ₂ O ₂	218	−1; 12.2	0.91	Negative
	Carbamazepine	C ₁₅ H ₁₂ N ₂ O	236.27	13	2.45	Neutral
β-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 ₃ N ₃ O ₈	790.0	2;13	−2.10	Neutral
	Iopamidol	C ₁₇ H ₂₂ I ₃ N ₃ O ₈	777.1	10.7	−2.42	Neutral
	Iohexol	C ₁₉ H ₂₆ I ₃ N ₃ O ₉	821.1	11.7	−3.05	Neutral
Blood lipid regulators	Clofibric acid	C ₁₀ H ₁₁ ClO ₃	214.65	3.35	2.57	Negative
	Gemfibrozil	C ₁₅ H ₂₂ O ₃	250.34	4.45	4.77	Negative
	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
Herbicide	Atrazine	C ₈ H ₁₄ ClN ₅	215.7	3.20	1.32	Neutral
	Simazine	C ₇ H ₁₂ ClN ₅	201.7	3.20	1.78	Neutral
Industrial chemicals	N-nitrosodimethylamine (NDMA)	C ₂ H ₆ N ₂ O	74.1	3.50	0.04	Neutral
	Bisphenol A	C ₁₅ H ₁₆ O ₂	228.3	9.8; 10.4	4.04	Neutral

Furthermore, modern technology has made plastics one of the most widely used materials in recent history, leading many to view them as a symbol of the Anthropocene. One of the greatest environmental concerns globally is plastic pollution [64]. Microplastics (MPs) are synthetic solid particles or polymer matrices, which can be either regular or irregular in shape, ranging from 1 µm to 5 mm in size, and are insoluble in water [65]. Global plastic production exceeded 360 million tons in 2018 and is expected to triple by 2050. A report reveals that Asia is the largest producer and consumer of plastic-based goods, with China responsible for a significant portion—32% of the “white pollution” [66]. Microplastics are created through primary processes such as cosmetics and beauty products (e.g., microbeads, microfibers) and secondary processes like weathering, friction, abrasion, and the fragmentation of larger plastic waste [67]. Nearly 80 to 90% of the microplastics found in water bodies originate from land-based sources [68,69], including plastic bottles, bags, toiletries, construction materials, and clothing [70].

The most commonly used types of plastics include polyethylene (PE), polypropylene (PP), polyvinyl chloride (PVC), polystyrene (PS), and polyethylene terephthalate (PET). The application of these polymers is largely determined by their properties (Table 3), although these properties are not exhaustive in defining their uses. In general, plastics consist of both crystalline and amorphous phases, which greatly influence their mechanical properties, such as strength and elasticity. When the amount of the amorphous phase is reduced, crystallinity and density increase. As density rises, so do properties like elastic modulus, tensile strength, stiffness, and surface hardness, while impact strength decreases [71]. PET, along with PE, is one of the primary materials used in food packaging and has become a major environmental pollution issue due to its high durability and low biodegradability.

Table 3. Physical and chemical properties of most used plastics.

Type of MPs	PS	PE	PP	PVC	PET
Chemical formula	$(C_8H_8)_n$ [70]	$(C_2H_4)_n$ [70]	$(C_3H_6)_n$ [70]	$(C_2H_3Cl)_n$ [70]	$(C_{10}H_8O_4)_n$ [70]
Specific density ($g\ cm^{-3}$)	0.07–1.05 [71]	0.85–0.94 [71]	0.905 [71]	1.35–1.39 [71]	0.96–1.45 (average 1.38–1.41) [71]
Tensile strength (MPa)	36–52 [72]	8–32 [72]	31–41 [72]	41–52 [72]	48 [72]
Melting point ($^{\circ}C$)	270 (syndiotactic) [73]	98–110 [72]	168–175 [72]	115–245 [72]	245 [72]
Crytallinity (%)	0 [74]	45–95 [75]	50–80 [75]	5–15 [74]	30–40 [76]
Lifespan (year)	50–80 [74]	10–600 [74]	10–600 [74]	50–100 [74]	450 [74]
Main use	Plastic cups, food packaging [70,71,77]	Bottles food, packaging shopping bags, cosmetic products [70,71,77]	Folders, food packaging, hinged caps, car bumper, medicine bottles [71]	Window frames, flooring and pipes, clothes [70,71]	Plastic beverage bottles, food packaging, personal care products [70,71,77]

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 2. Among these sources, household wastewater is particularly significant in introducing micropollutants into surface waters, making aquatic environments the primary receptor of these pollutants [78].

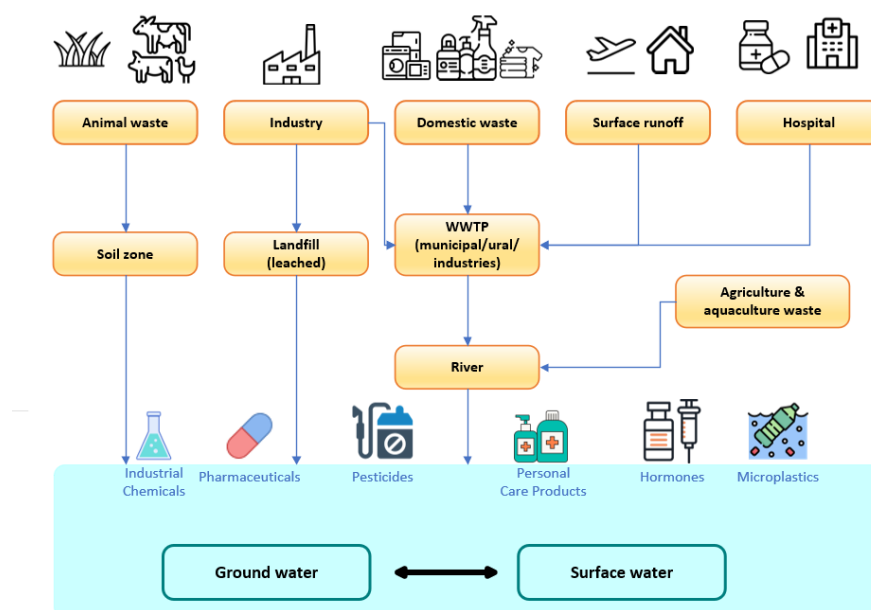


Figure 2. Sources of micropollutants in the environment [79–81].

Various pharmaceuticals are used in both human and veterinary medicine. Approximately 3000 different compounds are employed as pharmaceuticals, with annual production quantities reaching hundreds of tons [82]. 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 [83,84]. In Germany alone, around 8100 tons of active substances are used annually [85]. 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 [86].

About 70% of the pharmaceuticals in the wastewater originates from households, 20% comes from livestock farming, 5% is from hospital effluent, and the remaining 5% comes from runoff from non-point sources [87]. Sewage containing pharmaceutical pollutants 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 [88]. 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 (Figure 3) [89,90].

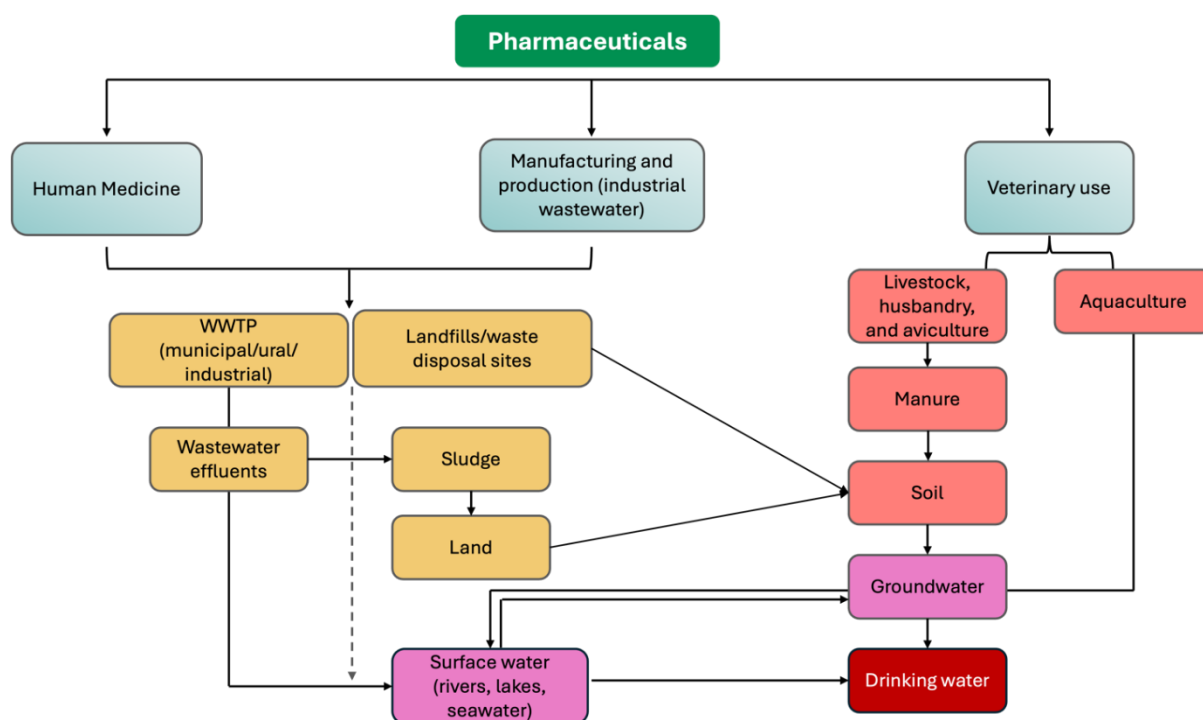


Figure 3. Routes of pharmaceutical compounds into the environment [78,91].

WWTPs are a significant source of pharmaceutical pollutants [2,88,92]. 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 ($\mu\text{g/L}$) [90]. 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 [93,94]. Among these, NSAIDs, antibiotics, and analgesics

are the most frequently used worldwide [28]. For instance, approximately 35 million people use NSAIDs daily across the globe [95], and in China, domestic production increased from 41,537 tons in 2013 to 46,673 tons in 2017 [96].

In Cuernavaca, Mexico, high concentrations of naproxen (732–4889 ng/L), acetaminophen (354–4460 ng/L), and diclofenac (258–1398 ng/L) have been detected in influent and effluent samples from a WWTP as well as in surface waters of the Apatalco River [97]. Similarly, diclofenac (10,221 ng/L) and acetaminophen (1234–2346 ng/L) have been detected in effluents from the Red Sea (Saudi Arabia) [98]. In Brazil, acetaminophen (17.4–34.6 ng/L), diclofenac (19.4 ng/L), and ibuprofen (326.1–2094.4 ng/L) have been found in surface and bottom water samples from Santos Bay [99].

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 [100]. 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 [101]. The most frequently detected antibiotics in wastewater include sulfonamides, quinolones, tetracyclines, fluoroquinolones, and nitroimidazoles [28]. 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 [102–104].

Veterinary pharmaceuticals are another direct source of micropollutants due to their widespread use in treating farm animals, leading to their presence in different aquatic environments [105]. Studies have identified these substances in animal production systems such as pig farming [106], poultry [107], dairy [108], sheep farming [68], and aquaculture [109–111]. In Germany, indoor livestock systems, including cattle, pigs, and poultry, are the primary sources of veterinary pharmaceutical pollution (82%), 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 [111–113].

Residues from veterinary medicinal products can runoff into surface waters or seep into groundwater if not absorbed by the soil, potentially contaminating drinking water [114]. 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 [88,115]. Antibiotics are widely used in animal farming to promote growth and improve feeding efficiency in cattle, pigs, and poultry [116]. 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 [117].

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 [118]. These antibiotics can enter surrounding water bodies directly through fish excretion, uneaten medicated feed, or improper disposal of unused medications [119]. The Asian aquaculture sector has expanded rapidly in recent decades, now accounting for nearly 90% of global production [120]. Studies have shown significant antibiotic use in Chinese aquaculture, contributing to veterinary contamination [111]. However, human activities may have an even greater impact on aquatic pollution than aquaculture due to the higher density of human populations.

2.3. Environmental Impact of Micropollutants

Micropollutants, even at very low concentrations in water bodies, can have long-term (chronic) effects on both human health and ecosystems. Organic micropollutants, such as pharmaceuticals, pesticides, and microplastics, can disrupt aquatic ecosystems and pose

risks to human health when consumed through contaminated water or food. These micropollutants contribute to issues, such as antibiotic resistance, endocrine disruption, cancer risks, and environmental degradation. They can bioaccumulate in the food chain, leading to persistent damage to ecosystems, while chronic human exposure may result in serious health complications (Figure 4) [121].

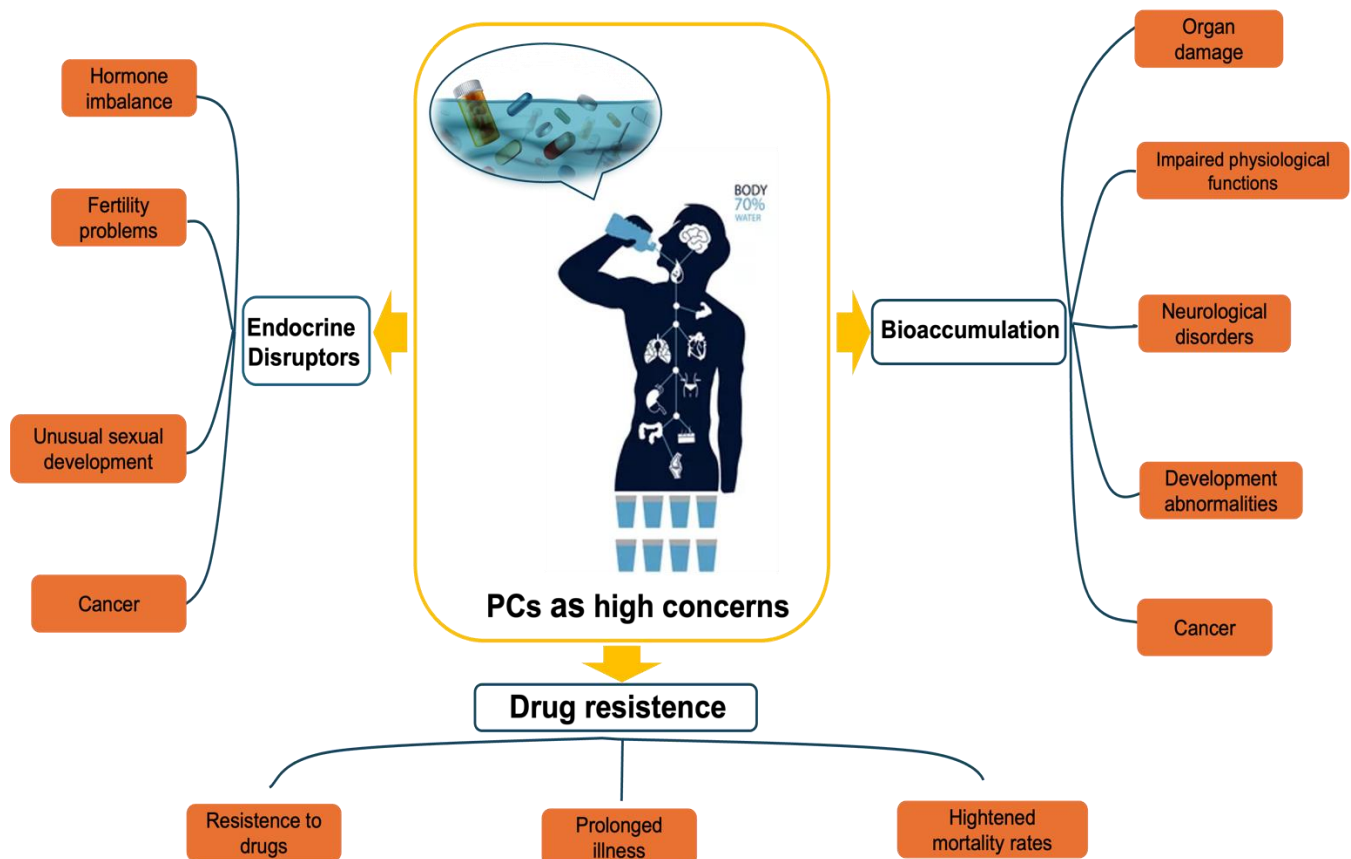


Figure 4. Key health risks linked to micropollutants, modified by [13,122,123].

2.3.1. Effect on Human Health

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 [124]. According to Feng et al. (2013), more than 30 million doses of NSAIDs are consumed daily, and this number is rapidly increasing [58].

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 [57]. According to Dölliver and Gupta, antibiotics contaminate groundwater and surface water through leaching and agricultural runoff [125]. 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 [45].

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 [126,127]. Some of the significant side effects linked to antidepressants include hypoglycemia, both acute and chronic toxicity, growth inhibition in aquatic organisms, and sexual dysfunction [45]. 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 [128,129].

Microplastics

Recently, microplastics have been identified as significant pollutants causing environmental issues. These particles have been detected in food consumed by humans and in the air. As a result, exposure to microplastics primarily occurs through inhalation and ingestion, with drinking water being a major pathway [130]. The health impacts of microplastics can be classified into three categories: physical effects, chemical leaching from their matrices, and their role as carriers for microorganisms [131,132]. Prata (2018) conducted a study highlighting the health risks linked to inhaling airborne microplastics, finding a correlation between chronic exposure to low concentrations and the development of respiratory and cardiovascular disorders [133]. Inhaled microplastics can lead to oxidative stress in the airways and lungs, causing symptoms such as coughing, sneezing, shortness of breath, fatigue, and dizziness. These effects result from inflammation, tissue damage, and reduced blood oxygen levels [134]. Recent studies have also associated nano-sized plastics with mitochondrial dysfunction in respiratory cells [130]. In addition, microplastics can serve as carriers for other environmental toxins, such as polystyrene (PS). Exposure to high concentrations of PS is harmful to human lung cells, increasing the risk of chronic obstructive pulmonary disease [131].

When microplastics are ingested, they can physically irritate the gastrointestinal tract, potentially causing inflammation and symptoms such as nausea, vomiting, and abdominal pain [132]. In addition to their physical effects, microplastics present a chemical risk by adsorbing and accumulating harmful environmental pollutants, such as heavy metals and polycyclic aromatic hydrocarbons. When microplastics are ingested, these toxic substances can enter the body through the gastrointestinal tract, exacerbating gastrointestinal distress and contributing to other health complications [135]. Recent studies have linked microplastics to an increased risk of cancer, nervous system disruption, and tissue damage, depending on exposure levels and individual sensitivity [135]. Plastics also leach chemicals like carcinogenic and endocrine-disrupting additives, such as bisphenol A (BPA) and phthalates, which can contribute to obesity, diabetes, and other health issues [136]. Campanale et al. (2020) highlighted that BPA-contaminated food was associated with nearly 12,404 cases of childhood obesity in 2008 [137]. Indirect effects, such as the transmission of microorganisms on microplastic surfaces, have also been documented [131,132].

Pesticides

Humans are exposed to pesticides both directly and indirectly, often through the consumption of fruits and vegetables grown in contaminated soil and water. Prolonged exposure to pesticides increases the likelihood of developing long-term diseases such as neurotoxicity, cancer, asthma, reproductive disorders, heart disease, and diabetes, as toxins accumulate in organs [136]. While the exact mechanisms are not fully understood, several pesticides, including paraquat, have been linked to neurodegenerative diseases like Parkinson's [137]. Pesticides can also interact with DNA, causing mutations that elevate

the risk of cancer. Organophosphorus insecticides, such as parathion and malathion, are particularly associated with breast cancer due to their impact on cellular growth and proliferation [138]. The potential genetic damage caused by occupational pesticide exposure is far greater than that from smoking and alcohol consumption, with DNA damage in exposed individuals, such as farmers, being 4.63 times higher than in non-exposed groups, as shown in a meta-analysis [139]. This underscores the severe risks associated with pesticide exposure.

2.3.2. Effects of Microplastics on Aquatic Ecosystems

Microplastics have extensive and diverse negative impacts on marine life [138]. In Marcharla, E.; Vinayagaming harmful chemicals and causing physical disruptions. These particles can leach toxic additives, limiting the growth and survival of phytoplankton [139]. Due to their small size, microplastics can be absorbed by phytoplankton, impairing their ability to capture sunlight for photosynthesis. Additionally, microplastics may accumulate on the water's surface, blocking sunlight and hindering photosynthesis not only in the affected phytoplankton but also in nearby organisms [140]. Phytoplankton, as primary producers, form the foundation of the marine food web, supporting zooplankton, small fish, and other marine organisms. Disruptions to the abundance and health of phytoplankton can trigger a chain reaction, negatively affecting the organisms that rely on them for food [141]. These ecological imbalances can extend to larger marine species, including commercially important fish, threatening fisheries and food security [142]. Furthermore, when larger marine fauna consume zooplankton or prey that contain microplastics, the concentration of contaminants can increase through biomagnification, posing serious risks to the health of larger marine animals [143]. Microplastic pollution has implications that extend beyond the aquatic environment, potentially endangering human health and well-being. Microplastics found in seafood are ingested by humans, accumulating in our bodies [144]. While the long-term effects remain uncertain, studies have established a link between microplastics and issues such as endocrine disruption, genotoxicity, and inflammation [145,146].

The accumulation of pharmaceuticals and their residues in aquatic ecosystems poses significant risks to aquatic organisms, including amphibians, fish, invertebrates, and phytoplankton [147]. Synthetic estrogens from contraceptives, for instance, can lead to reproductive dysfunctions and the feminization of male fish, resulting in altered sex ratios. Annually, human activities release around 30,000 kg of natural estrogens and 700 kg of synthetic estrogens, primarily from contraceptive pills [148,149]. Furthermore, substances such as antibiotics, beta-blockers, endocrine disruptors, and anticancer medications have been shown to impair physiological functions, increase mortality rates, and hinder reproductive capabilities in aquatic organisms [149].

Surface runoff transports pesticide residues into aquatic environments, posing a significant threat to the plants and animals living there. This polluted water, in turn, inhibits the growth rates and decreases the abundance of aquatic species [150]. In cases of extreme exposure to endocrine-disrupting pesticides, aquatic species may develop both male and female sexual characteristics, experience premature sexual development, and have affected regulatory genes [151,152]. Pesticides primarily target organs, such as the liver, kidneys, gills, and the nervous system in fish. Since fish are a food source for humans, exposure to pesticides can result in various health problems for consumers [150]. Pesticides used in agriculture and aquaculture also threaten bottom-feeding fish by disrupting their natural food sources, altering aquatic ecosystems, and affecting the fish food chain [153]. As pesticide concentrations increase, the availability of aquatic plants and insects, which are essential food sources for fish and other aquatic organisms, declines. This disruption also impacts insect-eating birds and broader food chains [154].

3. Presence of Micropollutants in the Aquatic Environment

Table 4 presents a comprehensive overview of key micropollutants, including substances such as caffeine, diclofenac, bisphenol A, estrogen, microplastic, etc., 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 4. Micropollutant concentrations across various water types and countries.

Water Type	Micropollutants	Country	Conc. [ng/L]	Ref.
Surface Water	Caffeine	Germany	65–6798	[140]
		Denmark	65–382	[141]
		Korea	268.7	[142]
		China	865	[143]
	Butylbenzylphthalate (BBP)	Mexico	5–201	[144]
	Diclofenac	UK	20–91	[145]
		Sweden	680	[146]
		Korea	3	[147]
		China	<147	[148]
	Carbamazepine	USA	6.8	[149]
		Korea	5–36	[142]
	Estrone	Germany	71	[140]
		Korea	3.6	[147]
	Ibuprofen	Germany	60–152	[150]
		Denmark	9–22	[141]
		China	1,417,000	[151]
	Naproxen	Germany	70	[150]
		Sweden	90–250	[152]
		Denmark	17–36	[141]
		China	<118	[148]
	Bisphenol A	Europe	10	[153]
		USA	80	[153]
		Korea	59	[154]
	Perfluorooctane sulfonic acid (PFOS)	Germany	5–101	[140]
	Atrazine	Korea	0.61	[154]
	Triclosan	USA	10–600	[12]
		Mexico	16–19	[144]
	Nonylphenol	Denmark	10–60	[141]
		China	36–33,231	[2]
Ground Water	Microplastics	India	$2.75 \pm 0.92 \text{ p}\cdot\text{L}^{-1}$	[155]
		China	$266 \pm 56 \text{ p}\cdot\text{L}^{-1}$	[156]
		Hongkong	$2.18 \pm 0.165 \text{ p}\cdot\text{L}^{-1}$	[157]
	Caffeine	USA	290	[158]
		Germany	102	[159]
		China	42.5	[159]
		Italy	84–683	[160]
	Estrone	Europe	4	[161]
	BBP	Mexico	1–82	[144]
	Ibuprofen	USA	3110	[162]
		Europe	3–395	[151]

	Carbamazepine	Europe	390	[161]
		USA	42	[158]
	Atrazine	Europe	253	[161]
		Europe	79–2299	[161]
	Bisphenol A	Mexico	1–10	[144]
		Europe	135	[161]
	PFOS	Mexico	1–345	[144]
		Mexico	18.7 p·L ⁻¹	[163]
Drinking Water	Caffeine	Spain	9.10	[164]
		Sweden	5.5	[165]
		USA	52.3	[159]
		Korea	34.3–95.5	[166]
		Turkey	3390	[167]
		Germany	611	[168]
		Japan	16	[169]
	Diclofenac	Spain	25	[170]
		Sweden	8	[165]
		France	56	[171]
		Japan	25	[169]
	Carbamazepine	France	41.6	[171]
		Japan	6	[169]
	Ibuprofen	France	14	[171]
		Germany	244	[168]
		Spain	1.19	[164]
	Naproxen	France	6	[171]
	Metoprolol	France	1	[171]
	Bisphenol A	Germany	72	[168]
	PFOS	Spain	0.55	[164]
	4-methyl-1,2,3-benzotriazole	Germany	45	[168]
	Terbutylazine	Spain	4.56	[164]
	Sucralose	Sweden	12	[165]
	Microplastics	China	2–23 p/bottle	[172]
		Australia	13 ± 19	[173]
		Iran	8.5 ± 10.2	[174]
WWTP Effluent	Caffeine	Europe	3002	[175]
		Korea	60	[176]
		China	376.5	[177]
	Diclofenac	Europe	174	[175]
		Korea	49	[176]
	Carbamazepine	Europe	4609	[175]
		Korea	74	[176]
		China	55	[177]
	PFOS	Europe	2101	[175]
	Ibuprofen	Europe	2129	[175]
		Korea	75	[176]
	Acesulfame	Europe	30.000	[90]
	Atrazine	Europe	36.6	[175]
	Bisphenol A	Europe	200	[90]
		China	623.6	[177]
	Dimethylphthalate (DMP)	Europe	340	[90]
	Estrone	Europe	217	[90]
		Asia	<51.2	[178]
		North America	56	[178]
	Sucralose	Europe	10.000	[90]

	North America	18.700–48.900	[178]
	Asia	1300–5490	[178]
1H-benzotriazole	Europe	2,210,000	[179]
Diethyl Phthalate (DEP)	Europe	800	[90]
	India	148 ± 51	[180]
Microplastics	China	30.6 ± 7.8	[181]
	Africa	86 ± 7.49	[182]

4. Techniques for the Removal of Micropollutants

Due to the hazard of microplastics (MPs) and pharmaceuticals (PCs), it has become a very important topic to understand the current status of the physical, chemical, and biological methods currently used to remove both. This section outlines various technologies for removing microplastics and pharmaceuticals, detailing their respective advantages and limitations.

4.1. Removal Technologies of Microplastics

Industrial wastewater, domestic wastewater, agricultural runoff, and livestock wastewater often contain significant amounts of microplastics, and WWTPs are not capable of removing all these contaminants [183]. Recently, numerous technologies have been developed to address the removal of microplastics (MPs) from aquatic environments, which can be classified into physical, chemical, and biological technologies based on their mechanisms of capture and removal (see Figure 5). WWTPs are the primary facilities for the abatement of MPs, where these methods are typically implemented. The methods, removal efficiencies, working media, and operational conditions for each technology are summarized in Tables 5–7.

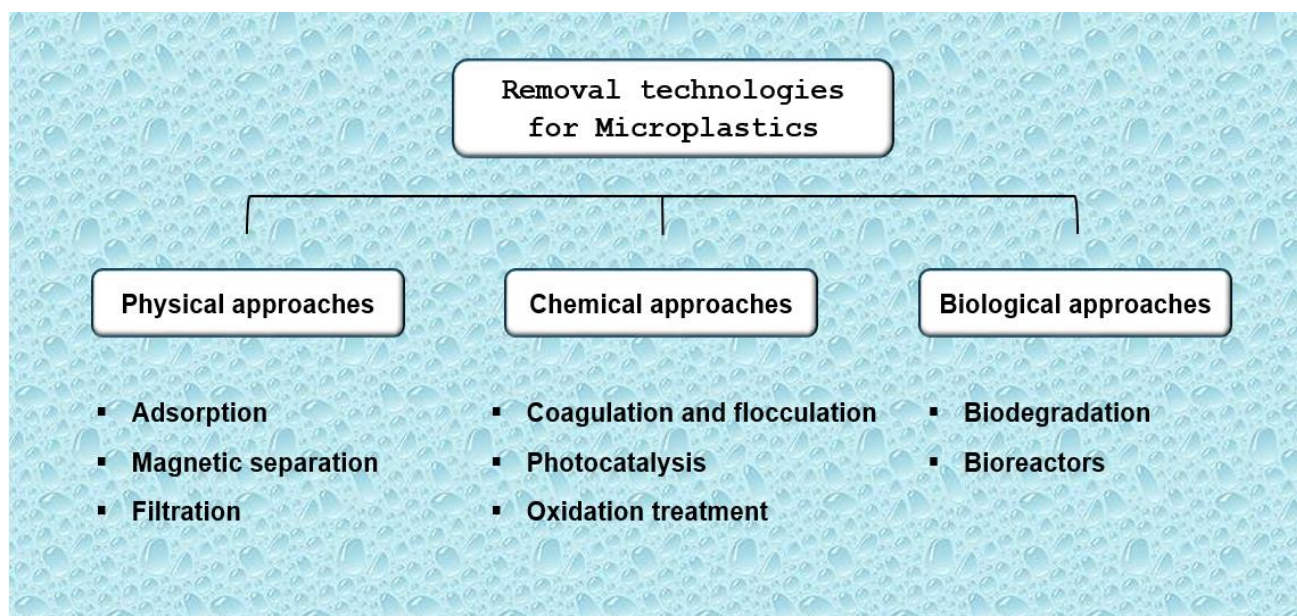


Figure 5. Overview of technologies for removal of MPs in water [184–186].

4.1.1. Physical Techniques for Removal of Microplastics

Adsorption is a key method for removing MPs and nanoplastics (NPs), particularly those smaller than 10 µm. This approach involves incorporating various adsorbents, such as graphene oxide, chitin, biochar, activated carbon, and metal-organic framework (MOF) materials. These adsorbents share common characteristics, including biocompatibility and

biodegradability [187]. Biochar, activated carbon, and MOF materials are three-dimensional, porous materials that provide advantages due to their high surface area, excellent mechanical strength, and robustness. The adsorption process relies on several mechanisms, including electrostatic interactions, hydrophobic interactions, hydrogen bonding, and π - π interactions. These interactions are driven by the surface properties of both the microplastics/nanoplastics and the selected adsorbent materials [188–190]. For instance, Sun et al. (2020) [188] developed an adsorption-based separation process using a biodegradable chitin and graphene oxide sponge. This system could remove up to 89% of polystyrene from water after being regenerated through ethanol washing and freezing treatment, which allowed it to be reused three times. An alternative and efficient method involves the rapid separation and removal of MPs by magnetizing them and applying an external magnetic field. The magnetization of MPs relies on the same adsorption principles [185]. For optimal removal efficiency, combining adsorption with magnetization is crucial, especially when magnetic adsorbent materials are used, as this enhances the overall removal performance. Additional literature on the removal of MPs through adsorption and magnetization is summarized in Table 5.

Table 5. Physical techniques for the removal of microplastics.

Microplastic Types	Physical Methods	Experimental Details	Removal Efficiency (%)	Ref.
PS, PS-NH ₂ , and PS-COOH (1 μ m)	Adsorption	- Adsorbents: Chitin and graphene oxide - MPs/NPs: 1 mg/L - pH: 4, 6, 8, 10 - Temp.: 25, 35, 45 °C	72.4–89.8	[188]
MPs in sewage treatment plant	Adsorption	- Adsorbents: Granular activated carbon (800 kg in tower) - Flow rate: 10 m ³ /day	92.9	[191]
PE, PET, and PA (48 μ m)	Adsorption and magnetization	- Adsorbents: Magnetic carbon nanotubes (M-CNTs); 2–7 g/L - MPs/NPs: 5 g/L	100	[192]
PS (1 μ m)	Adsorption and magnetization	- Adsorbents: Magnetic biochar (MBC); 10 mg - MPs/NPs: 100 mg/L, 10 mL - pH: 3, 5, 7, 9	95.02 (MBC) 95.79 (Zn-MBC) 94.60 (Mg-MBC)	[193]
MPs	Filtration (disc filter)	- MWCO: 18 μ m - Media: Treated wastewater	89.7 (particle); 75.6 (mass)	[194]
MPs	Filtration (disc Filter)		80–98	[195]
MPs	Filtration (UF)	- Media: Landfill leachate - Pretreated landfill leachate samples entered the membrane bioreactor and ultrafiltration, nanofiltration, and reverse osmosis	75	[196]
MPs	Filtration (sand filter systems with biochar)		>90	[197]

Additionally, filtration is another physical method commonly used for removing MPs and NPs from polluted water. Various membrane filtration technologies, including MF, UF, dynamic filtration, and membrane bioreactors, have been employed to effectively remove MPs/NPs. Furthermore, media filtration techniques such as sand filtration and the use of activated carbon particles have also been utilized. Malankowska et al. (2021) provide a detailed review of recent advancements in MF, UF, and nanofiltration (NF) for microplastic removal [198].

According to studies by Komorowska-Kaufman and Marcinak (2024) and Simon et al. (2019), approximately 80–98% and 89.7% of microplastic particles were retained by a

disc filter, respectively [194,195]. Disc filtration, when applied in the tertiary treatment process, can significantly reduce the number of microplastics in effluent wastewater [199]. Michielssen et al. (2016) identified granular sand filtration and membrane filtration as the most effective methods for microplastic removal in WWTPs, based on a meta-analysis of available technologies [200]. Large-scale studies also support these findings, with rapid sand filtration able to remove up to 97% of microplastics [201]. However, high-pressure membrane filtration faces challenges such as pore blockage and flux reduction. Ultrafiltration, for instance, experiences a 38% reduction in flux within just 48 hours due to the accumulation of microplastics and their interaction with organic matter in the water [202]. Additionally, the size of microplastics in raw water has been found to be a major factor influencing fouling during ultrafiltration, with the most severe effects occurring when microplastic particles are around 1 µm in size [203].

4.1.2. Chemical Techniques for Removal of Microplastics

Coagulation and flocculation are common processes used in WWTPs, and the combination of these methods is often considered effective for microplastic removal. The primary goal of coagulation and flocculation is to separate pre-existing colloidal particles in the solution by neutralizing their charges, forming flocs, and subsequently removing them through sedimentation or filtration [204]. Charge neutralization occurs when the negatively charged microplastic particles are neutralized by the hydrolyzed products of the coagulant, causing the coagulant to adsorb onto the microplastic surfaces. This interaction leads to flocculation, allowing the particles to settle. The most commonly used coagulants are aluminum sulfate ($\text{Al}_2(\text{SO}_4)_3$), ferric sulfate ($\text{Fe}_2(\text{SO}_4)_3$), and ferric chloride (FeCl_3) (see Table 6) [205].

Table 6. Chemical techniques for the removal of microplastics.

Microplastics	Chemical Methods	Experimental Details	Removal Efficiency (%)	Ref.
PS (1, 6.3 µm)	Coagulation/flocculation	- Coagulant: FeCl_3 , polyaluminium chloride (PACl), polyamine - Media: Wastewater	99.4 (FeCl_3) 98.2 (PACl) 65 (polyamine)	[206]
PE (10, 140 µm), PS (10, 140 µm), and polyester fibers	Coagulation/flocculation	- Coagulant: Alum, aluminum chlorohydrate (0–10 mg/L) - pH: 7.2 ± 0.1 - Media: Surface water	82–99	[207]
PE (5–15 µm) and polyester (17.5–50.6 µm)	Coagulation/flocculation	- Coagulant: Alum [$\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$]	86–99	[208]
High-density PE	Photocatalytic degradation	- Semiconductor catalyst: C, N-TiO ₂ (200 mg) - MPs: 200 mg - pH: 3–11	71.77	[209]
PS and PE	Photocatalytic degradation	- Semiconductor catalyst: TiO ₂ nanoparticle film	98.40 (400 nm PS) 95.30 (700 nm PS)	[210]
MPs	Ozonation	- Ozone 12.6 mg/L for 1 min during tertiary treatment - Media: Real wastewater (after tertiary treatment)	99.2	[211]

Photodegradation has been recognized as a highly effective and promising technology for treating hazardous organic pollutants, including microplastics in wastewater [212]. In this process, semiconductor materials absorb visible or ultraviolet light, generating free radicals such as superoxide and hydrogen radicals, which then oxidize and break

down the microplastics [213]. As shown in Table 6, commonly used semiconductor materials in photocatalysis include TiO₂ and ZnO nanocomposites, due to their excellent optical properties, high redox potential, good electron mobility, and non-toxicity [214–216]. In a study by Ariza-Tarazona et al. (2020) [209], TiO₂ doped with carbon (C) and nitrogen (N) led to a significant 72% mass loss in high-density polyethylene (PE) beads after 50 h of treatment. The researchers enhanced the degradation performance by lowering both the water temperature and pH, which improved the formation of radicals under optimized conditions and caused alterations in the plastic's properties at lower temperatures. Ozonation is another commonly used tertiary treatment in WWTPs, designed to remove residual pollutants from the coagulation process and purify the effluent. Hidayatullah et al. (2019) [211] reported that combining ozonation with primary, secondary, and coagulation treatments removed 99.2% of microplastics in a full-scale treatment plant. However, a major drawback of ozonation is that insufficient ozonation can generate intermediate compounds, which may pose risks to human health and the environment due to the production of reactive oxygen species.

4.1.3. Biological Techniques for Removal of Microplastics

Biological methods for MP removal primarily involve the hydrolysis and digestion of MPs by microorganisms, such as fungi, bacteria, and extracellular enzymes, through both aerobic and anaerobic processes [185]. Traditional water treatment systems often incorporate biodegradation to remove organic matter. However, these systems are not effective in removing MPs, leading researchers to explore alternative solutions. In a comprehensive review on MP removal, Iyare et al. (2020) [217] found that 19 out of 21 traditional wastewater treatment systems employed activated sludge treatment as a secondary step. On average, activated sludge was reported to remove about 16% of MPs from the water, with removal rates varying widely from 0.2% to 52%. Similarly, Liu et al. (2019) [218] observed a reduction of only 16.6% in microplastics through biological treatment with activated sludge (see Table 7).

Microbial digestion presents a highly promising solution for addressing the plastic waste problem in a potentially sustainable manner. However, one of the main challenges is the long treatment times required for large-scale effectiveness. Additionally, biological systems face difficulties in managing the diverse nature of plastic waste, as enzymatic digestion typically targets specific polymer chains [219]. To address the wide variety of polymers found in waste, it is crucial to employ a diverse range of plastic-degrading microorganisms [220]. Most of the bacteria involved in degradation are Gram-negative bacilli, with *Pseudomonas* species being particularly effective at breaking down various plastics, including both natural and synthetic polyethylene [221].

Table 7. Biological techniques for the removal of microplastics.

Microplastic Types	Biological Methods	Experimental Details	Removal Efficiencies (%)	Ref.
MPs	Activated sludge	Media: Wastewater		
		Influent MPs: 47.4 ± 7.0 n/L Effluent MPs: 34.1 ± 9.4 n/L	16.6	[218]
PE, PET, PS, and PP (75 µm)	Biodegradation	Media: Aqueous phase	6.2 (PE)	
		Microorganism: <i>Bacillus gottheilii</i>	3.0 (PET)	[222]
		Room temperature	5.8 (PS)	
		Time: 40 days	3.6 (PP)	
PP (250 µm–4 mm)	Biodegradation	Media: Aqueous phase		
		Microorganism: <i>Pseudomonas</i> sp.	17.3	[223]
		Temperature: 10 °C		
		Time: 40 days		
MPs	MBR	Media: Activated sludge	99.4	[224]

The membrane bioreactor (MBR) is a highly effective hybrid treatment method used to treat contaminated water [225]. MBRs have proven effective in removing high-strength contaminants, including microplastics and polymeric debris [226]. In a study by Talvitie et al. (2017), several advanced final-stage treatments were evaluated, including MBR, disk filters, rapid sand filtration, and dissolved air flotation. They found that MBRs achieved a remarkable 99.9% reduction, decreasing microplastic particles from 6.9 to 0.005 per liter [201]. While this technique effectively eliminates microplastics, preventing membrane fouling remains a critical factor for maintaining its efficiency [227].

4.2. Removal Technologies of Pharmaceuticals

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 6) [228]. 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 [229,230]. 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 [231].

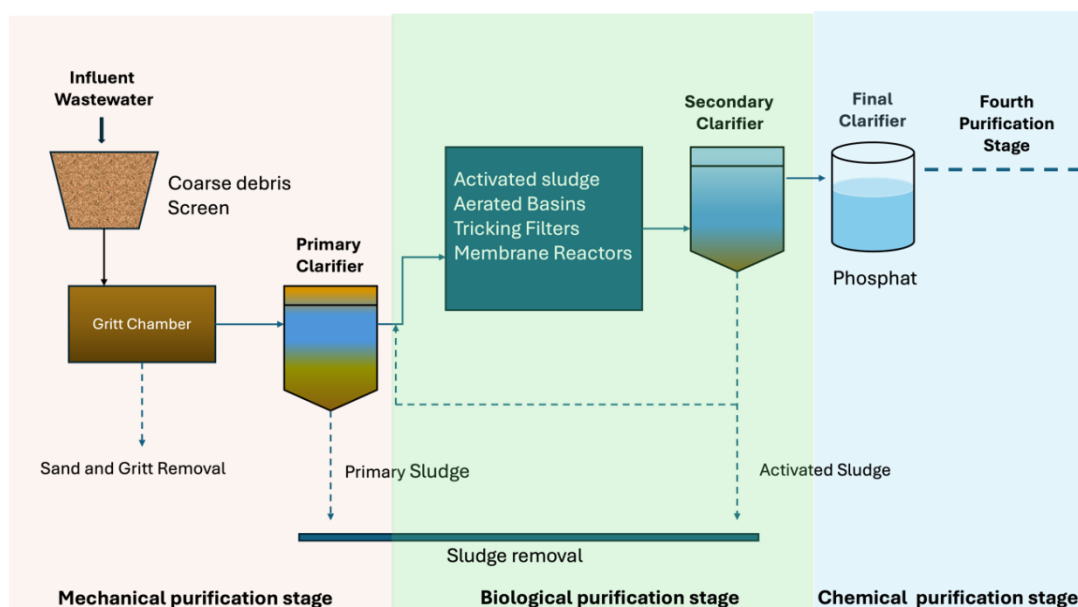


Figure 6. Schematic representation of a conventional wastewater treatment plant [231,232].

Jelic et al. (2011) conducted a study on the presence of 43 pharmaceuticals in the influent, effluent, and sludge of three WWTPs in Catalonia [233]. 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 PCs 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 [232–235]. 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 [234]. While the activated sludge process is commonly employed to treat wastewater containing NSAIDs, it has been shown that these drugs are not entirely removed [235].

Ongoing research and development have led to the exploration of various technologies that could serve as a fourth treatment stage in WWTPs [236–238]. These technologies are classified into biological, chemical, and physical processes, with potential combinations of these methods (Figure 7). The following sections provide an overview of these individual processes, emphasizing their functionality and effectiveness in eliminating PCs from wastewater [234,235,239].

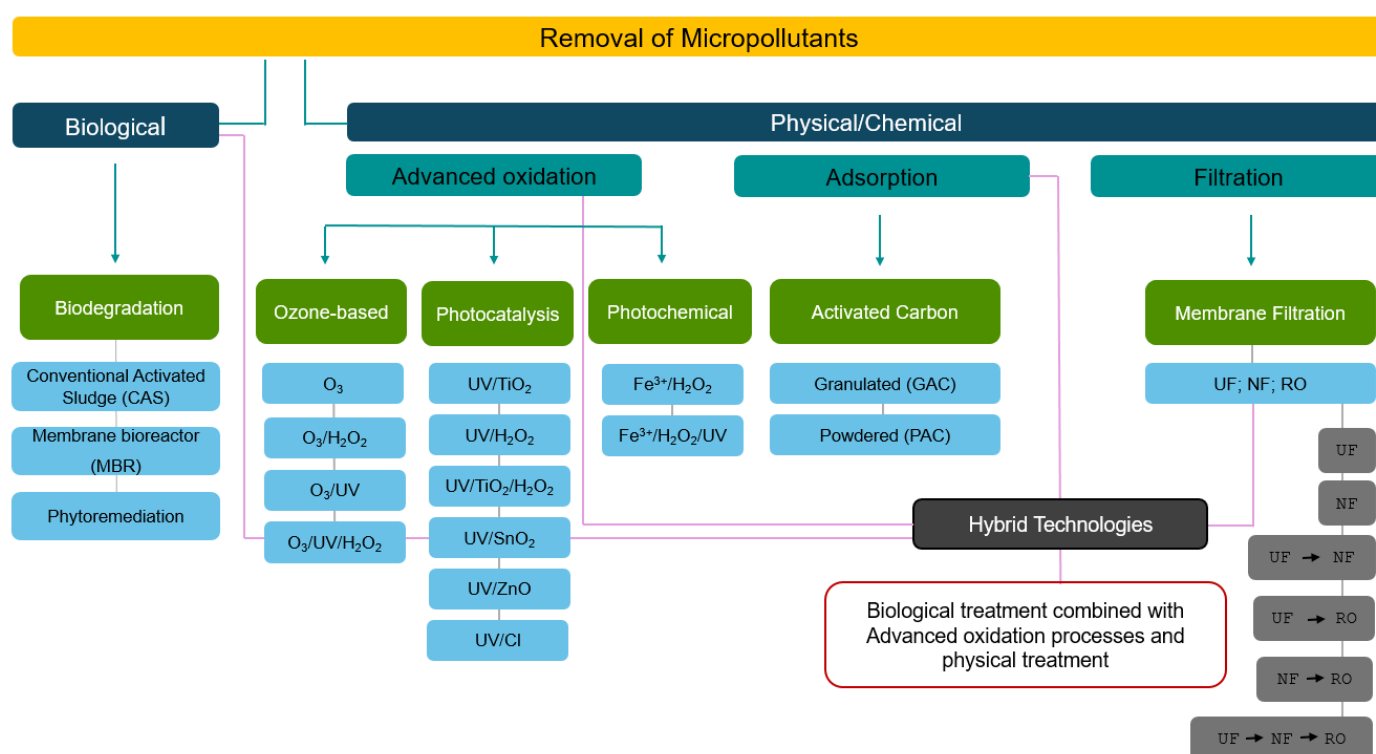


Figure 7. Processes for the removal of micropollutants [228,240].

Table 8 presents a summary of the advantages and disadvantages of the wastewater treatment processes discussed above, along with monitoring methods for selected micropollutants, particularly PCs, in the aquatic environment. 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 8. Advantages and disadvantages of various treatment processes for micropollutants removal and their analytical methods.

Treatment Processes	Advantages	Disadvantages	Category	Analytical Methods
Conventional biological treatment [241–243]	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	EDCs Bisphenol A (BPA) Nonylphenol (NP)	Centrifugation → Acidification → SPE → HPLC-UV at 280 nm [244,245] Liquid Extraction → Centrifugation → HPLC-UV at 274 [244,245]
			Organic matter, nitrate, and phosphorous, e.g., ammonium, sodium acetate	TC-IC (TOC) [241]
			PCs, e.g., carbamazepine and diazepam	SPE → LC-MS or GC-MS [241]
Advanced biological treatment [242,243,246]	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	Organic Matter, e.g., meat extract, peptide, sodium acetate, and glucose	TOC [247]
			PPCPs, EDCs, and pesticides	SPE → Derivatization → GC-MS [247]
			Organic and inorganic (C, N, and P)	TOC, TN and COD [248]
			Pharmaceuticals and herbicides	SPE → HPLC-MS [248]
Advanced oxidation processes (AOPs) [249–251]	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	Pharmaceuticals, e.g., diclofenac	UHPLC-QqQ-MS [249]
			Food additives and artificial sweeteners, e.g., acesulfame	UHPLC-QqQ-MS [250]
Adsorptive treatment [251–253]	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	Industrial chemicals	HPLC-UV at 230 nm and HPLC-FLD at 275 and 225 nm [245,254]
			Corrosion inhibitors and biocides/pesticides	SPE → UPLC-MS/MS and TOC [255]
			Anionic dyes, e.g., azo and anthraquinone	UV-Vis spectrophotometer [245,256]
			Pharmaceuticals, e.g., naproxen, ibuprofen, diclofenac, ketoprofen	HPLC-UV at 230 nm [245,257]
Membrane technology [250,255,256]	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	Heavy metals, organic and inorganic compounds	UV-Vis spectrophotometer, titrimetric determination, and oximeter [245,258]
			Pharmaceuticals, e.g., naproxen	UV-Vis spectrophotometer [259]
			Pesticides	SPE → LC-MS/MS [260]

4.2.1. 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 [261]. The biological methods used to remove micropollutants in WWTPs can be divided into conventional and advanced processes (Figure 8). 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 [262]. On the other hand, advanced biological processes, such as two-phase partitioning bioreactors, MBRs, immobilized cell bioreactors, and moving bed biofilm reactors, utilize enhanced technologies to improve treatment efficiency [263]. 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 [264].

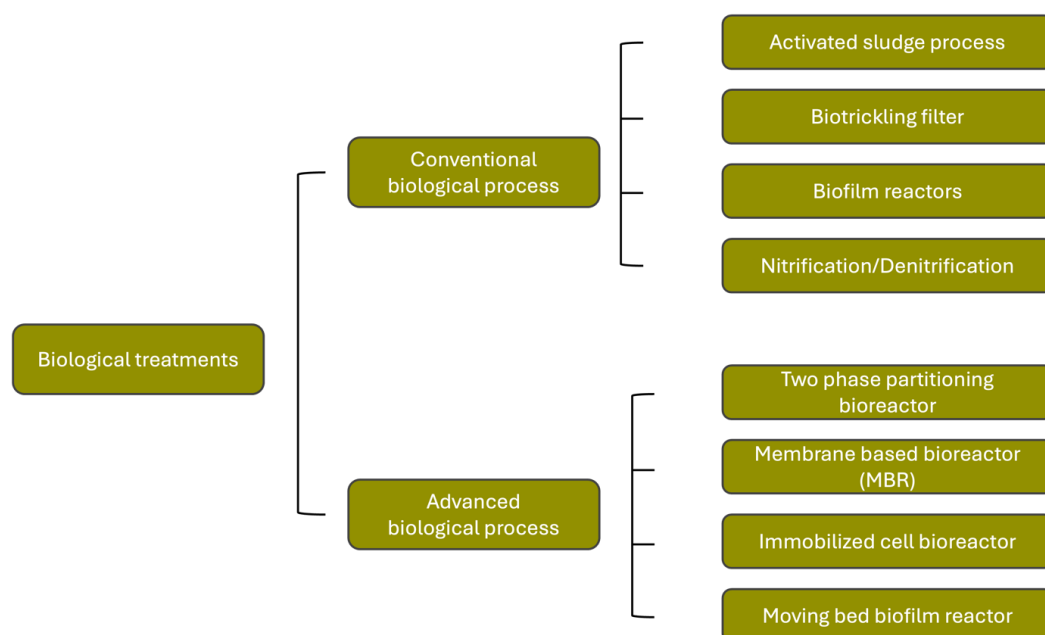


Figure 8. Traditional and advanced biological treatment methods for wastewater treatment plants contaminated with pharmaceutical compounds [262,263].

Activated Sludge

The removal of pharmaceuticals entering a WWTP primarily takes place during the biological treatment stage [218,261,262]. 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 [58]. 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 [241,265].

Treatment efficiencies for pharmaceuticals in the biological process of WWTPs can range from 20% to 99% [218,241], with some compounds, like carbamazepine, showing removal rates as low as below 20% [176,241,266,267]. This low removal rate for carbamaz-

epine can be attributed to its persistent nature and water-soluble properties [176]. 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 [176,268]. Typically, diclofenac removal rates range around 40%, but biodegradation can occur with varying efficiencies depending on the conditions [269]. A study by Sim et al. (2010) found the removal rates of naproxen and gemfibrozil to be $82 \pm 20\%$ [270], while Clara et al. (2005) reported a 99% removal of ibuprofen during biological treatment in a WWTP [271].

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 [176]. 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, consistent with findings by Zwiener and Frimmel, who observed that anoxic-oxic processes enhance diclofenac degradation [272].

Pharmaceutical removal in biological treatment relies on two primary mechanisms: biodegradation and sorption (including absorption and adsorption) [57,265,273]. Biodegradation can occur via co-metabolism, where other substances serve as the primary carbon or energy source, or through microbial metabolism [274]. Sorption efficiency depends on electrostatic interactions [265,273,275] and compound hydrophobicity, with higher hydrophobicity often resulting in increased removal efficiency [266]. For example, Chen et al. (2022) observed strong adsorption of antibiotics like erythromycin and azithromycin during biological treatments, primarily driven by these interaction mechanisms [276]. Conversely, compounds with high water solubility (low hydrophobicity), such as sulfamethoxazole, are expected to exhibit lower sorption and removal rates [265].

The biological treatment of PCs can lead to mineralization (conversion into CO₂, water, and inorganic ions), degradation into smaller molecules, or minor structural modifications [277]. The formation of metabolites or biotransformation products depends on the PCs nature and the type of microorganisms involved [278]. 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 [279].

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 [280]. 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 8000 and 12,000 mg/L, while CAS typically operates at around 5000 mg/L, as higher mixed liquor suspended solids (MLSSs) concentrations can lead to settling issues in the sedimentation basin [281].

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 [280]. CAS is often insufficient for removing organic pollutants due to the quantity and variability of micropollutants [282]. Studies have shown that MBRs can effi-

ciently 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 [280,283,284].

As Mert et al. (2018) [284] and Nghiem et al. (2020) [280] 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 [285]. 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 [285]. To address this, high-retention membrane bioreactors (HRMBRs), such as osmotic membrane bioreactors (OMBRs) [286], membrane distillation bioreactors [287], and bio-electrochemical membrane reactors (BEMRs) [288], can be used to enhance the removal of persistent, high molecular weight micropollutants [289].

Research by Song and Luo et al. (2018) [248] 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 ($\text{Log } D > 3.2$) and hydrophilic ($\text{Log } D < 3.2$). 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.

Cornelissen et al. (2011) [290] investigated the OMBRs, which integrate 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 [290]. Nguyen et al. (2013) [291] 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 [291].

In conclusion, Pathak et al. (2020) [289] 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 [289].

Phytoremediation

Phytoremediation technology is an innovative, eco-friendly approach used to degrade and eliminate various micropollutants from the environment, including pharmaceuticals, personal care products (PPCPs), pesticides, dyes, heavy metals, and inorganic elements, through the metabolic processes of plants. This method utilizes specific plants—whether aquatic, terrestrial, or marsh species—in specialized systems known as constructed wetlands (CWs). The mechanisms involved in phytoremediation include photolytic degradation, sorption, plant uptake, accumulation, and microbial degradation, making CWs a promising alternative to traditional wastewater treatment plants (WWTPs). CWs have gained widespread recognition due to their sustainable nature, strong pollutant

removal capabilities, minimal waste production, soil improvement, greenhouse gas reduction, ease of operation and construction, exceptional stability, cost-effectiveness, and significant potential for nutrient and water recycling. CWs can be broadly categorized into (a) free water surface flow (FWS-CW), (b) subsurface flow (SSF-CW), and (c) hybrid constructed wetlands (see Figure 9). For more information on the different types of constructed wetlands, further details are available [277].

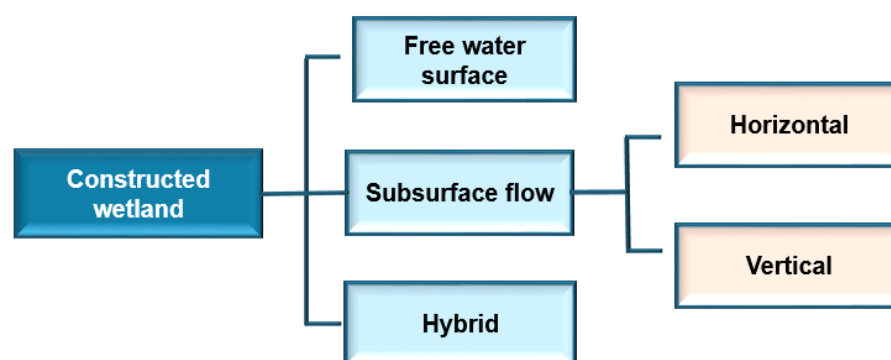


Figure 9. Different type of constructed wetlands.

The removal of PPCPs occurs through a simultaneous and complex interaction of physical, chemical, and biological processes. The primary biotic pathways responsible for eliminating organic compounds include bioaccumulation, bioadsorption, and biodegradation. PPCPs can enter plants through their root tissues, where ionic uptake is driven by diffusion during mass stream application in soil, or through aerial tissues, such as leaves, from the air. The efficiency of uptake is also influenced by various physical and chemical factors, such as molecular mass (compounds with a molecular mass of less than 1000 are more easily absorbed), hydrophobicity, pollutant concentration, and solubility [277]. In a study by Ávila et al. (2015) [278], it was reported that a full-scale hybrid system combining vertical flow constructed wetlands (VFCWs) and horizontal flow constructed wetlands (HFCWs) planted with *Phragmites australis*, along with a free water surface flow constructed wetland (FWS-CW) featuring a mix of plant species, achieved a removal efficiency of over 80% for PPCPs. In another study by Yi et al. (2017) [279], the removal efficiency for 10 selected PPCPs was found to exceed 90% in hybrid systems. Further studies can be referenced in Table 9.

Table 9. Different CWs and plants used for micropollutants removal.

CW Design	Plant Used	PPCPs	Removal (%)	Ref.
Free water constructed wetland (lab-scale)	<i>Spirodela polyrhiza</i>	Paracetamol,	>95	[292]
		Caffeine,	>95	
		Triclosan	>95	
VFCW (lab-scale)	<i>Phalaris arundinacea</i>	Diclofenac, Sulfamethoxazole	52–91 47–74	[293]
VFCW	<i>Phalaris australis</i>	Bisphenol A (BPA),	60–73	[294]
		Metoprolol,	77	
		Diclofenac	28–30	
Subsurface flow CW	<i>Phragmites australis</i>	17 α -ethinylestradiol	81.4	[295]

4.2.2. Physical Treatments Technologies

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 [29]. 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 [296].

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 m²/g), and strong surface interactions between pharmaceuticals and the adsorbent surface [297]. 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 [298]. 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) [299].

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 [300]. 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 [301–303]. Table 10 demonstrates the high removal efficiency of certain pharmaceutical compounds from water and wastewater.

Table 10. Removal efficiency of selected pharmaceuticals by activated carbon from various precursors and their analytical methods.

Pharmaceuticals	Water Type	Concentration (mg/L)	AC	Removal Efficiencies (%)	Analytical Methods	Ref.
Diclofenac	Various	10–30	AC from cocoa pod husks	76.0–93.6	UV-Vis at 380 nm	[304]
Carbamazepine	Wastewater	2	PAC	93	HPLC-UV	[305]
Naproxen	Wastewater	1–30	PAC	67.2–89.2	UV-Vis at 230 nm	[259]
Sulfamethoxazole	Distilled	50–500	AC	90	HPLC-DAD at 254 nm	[306]
Penicillin G	Distilled	50–1000	AC	12.0–78.3	Hydroxylamine method → UV-Vis at 515 nm	[307]
Atenolol	Various	5–900	GAC	88	UV-Vis at 224 nm	[308]

The AC/GAC process is limited by the specific properties of AC material and the contact time with the water, making it less adaptable [309]. 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 [310]. 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 [311]. 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 [312]. 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 [299].

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 [252]. 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) [313]. In contrast, compounds that adsorb weakly, such as sulfamethoxazole, require significantly higher PAC dosages, typically ranging from 10 to 50 mg/L or even up to 100 mg/L [314].

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 [315–317]. 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) [318,319].

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 [3,320].

Membrane selectivity is governed by several mechanisms, including size exclusion, electrostatic interactions, hydrophobic interactions, adsorption, diffusion, solute–solute interactions, and fouling. Simon et al. [321] 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 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) [322], who observed that NF and RO membranes achieve higher removal rates for ionic substances (97%) compared to non-ionic 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 neg-

atively 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% [323]. Similarly, Albergamo et al. (2019) [324] 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 [324]. Both studies emphasize the significant role of size and hydrophobicity in PCs removal, with electrostatic repulsion enhancing the rejection of negatively charged PCs.

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) [325], 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 K_{ow} (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 [325].

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 [326,327]. 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 [327]. Yoon et al. (2006) confirmed this by stating that once steady-state operation is reached, size exclusion becomes the dominant mechanism in retaining EDCs and pharmaceutical and personal care products (PPCPs) by UF membranes [328,329]. 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 11.

Table 11. An overview of membrane processes using organic ultrafiltration membranes for micropollutant removal and their analytical methods.

Membrane Processes	Matrix	Target MP (PPCPs)	Removal Efficiencies (%)	Analytical Methods	Ref.
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	HPLC-PAD at 220, 250, and 280 nm	[326]
UF (8000 Da)	Synthetic (model) water and natural freshwater sources	52 EDCs/PPCPs	Up to 80% removal efficiency for hydrophobic compounds	LC/MS/MS and GC/MS/MS	[328]
UF (100 kDa)	Synthetic (model) water	Estradiol and ibuprofen	25% ibuprofen, 80% estradiol	UV spectrometry + TOC + scintillation counter	[327]

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 [316,330]. 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 [331].

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 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 12) 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 12. Rejection of selected pharmaceuticals by polymeric NF membranes and their analytical methods.

PCs	pH	Membrane Name	Materials	MWCO (Da)	Removal Efficiencies (%)	Analytical Methods	Ref.
PARA*	6.5	NF270	Polyamide	200–300	44	HPLC-DAD	[332]
PARA	7	NF 200	Aromatic polyamide	~ 300	22	HPLC-MS	[330]
IBU *					89		
PARA					75		
IBU					96		
PARA	7.4–7.6	NF 270	Thin aromatic or semi-aromatic polyamide	200–300	0	TOC, TN and HPLC-MS	[333,334]
IBU					99		
DIC *					95		
PARA					99		
IBU					99		
DIC					90		
PARA	7	NF 270 NF 90	Polypiperazine with polymeric active layer polyamide supported by polysulfone	220 102	20–30 ~ 90	LC-MS/MS	[334]
DIC	8	FM	Hydrophilic polyethersulfone	1000	61	LC-MS/MS	[335]
IBU	8	NP010			55		
PARA	12	NF 50	Sulfonated polyethersulfone	1000	36.16	UV-Vis at 243, 222 and 276 nm	[317]
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 [317,336,337].

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 Al₂O₃ and TiO₂. 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 Al₂O₃ and TiO₂, hold promise for the removal of pharmaceutical micropollutants from water [69,228,338,339].

Additionally, research by Radeva et al. (2021) [339] 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 13. 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 [339].

Table 13. Rejection of selected pharmaceuticals by ceramic membranes and their analytical methods.

Membranes	MWCO (Da)	PCs	Removal Efficiencies (%)	Analytical Methods	Ref.
TiO ₂	200	41 organic compounds (N-nitrosamine and PCs, e.g., IBU, DIC, and CARBA *)	95–100	Nitrosamine: SPE → GC-MS/MS PCs: SPE → HPLC-MS/MS	[69]
LC1/LC2 *	630/440	PCs, e.g., SUL * and CBZ	50–80	UPLC-MS/MS	[338]
TiO ₂ (UF-membrane)	3.0 nm	DIC and IBU	32–47	HPLC-MS-MS	[228]
Al ₂ O ₃ /LBL coating with polyelectrolytes (polystyrene sulfonic acid)	~200	IBU, DIC, SUL, and Clofibric Acid	56% für SUL, up to 84% für DIC	HPLC	[339]

* CARBA: carbamazepine; * SUL: sulfamethoxazole; * LC1 and LC2 manufactured with active layers of TiO₂/ZrO₂.

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 [340].

Incorporating inorganic membranes into NF systems could improve the efficiency and sustainability of water treatment, especially in the removal of persistent pharmaceutical pollutants. Their longevity and reduced maintenance needs can lead to more cost-effective and reliable water treatment solutions, particularly in industrial and municipal applications.

4.2.3. Chemical Treatment Technologies

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 [341]. 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 [342].

Studies have demonstrated that ozone can effectively target electron-rich pharmaceutical aromatic compounds such as sulfamethoxazole, ciprofloxacin, carbamazepine, azithromycin, clarithromycin, diclofenac, erythromycin, and metoprolol [314,343–345]. 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, cephalixin, and penicillin [343,345]. Table 14 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 [346]. 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 [312,347]. According to Östman et al., ozone was found to be less effective at removing pharmaceuticals compared to GAC [348].

Table 14. Ozonation-based removal of selected pharmaceuticals and their analytical methods.

Applied Treatment (Concentration and Duration)	Pharmaceuticals	Elimination Efficiencies (%)	Analytical Methods	References
O ₃ (5 mg/L, 15 min)	Carbamazepine	>90	GC-MS/MS, LC-MS/MS	[8]
	Diclofenac	>90		
	Metoprolol	80–90		
	Trimethoprim	>90		
O ₃ (n/a, n/a) *	Ibuprofen	83	GC-MS/MS, LC-MS/MS	[349]
	Diclofenac	99		
	Carbamazepine	80		
O ₃ (33 mg/L, 20 min)	Tetracycline	95	UV–Vis at 357 nm and TOC	[350]

* n/a: not available.

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 CO₂ and H₂O. Various methods are used to produce hydroxyl radicals (-OH), including ozone (O₃)-based AOPs [351], hydrogen peroxide (H₂O₂)-based approaches [352], heterogeneous photocatalysis [249,255,353,354], sonochemical and electrochemical processes (EAOPs) [355], and combinations of these techniques. These processes aim to effectively eliminate pollutants. Table 15 highlights recent studies on the development and application of AOPs for wastewater treatment and the removal of micropollutants, mostly pharmaceuticals.

Table 15. The most widely used types of advanced oxidation processes for micropollutant removal and analytics.

AOPs-Type	Micropollutants	Removal Efficiencies of PCs (%)	Analytical Methods	References
Peroxone	Artificial sweetener, acesulfame	~100	UHPLC-QqQ-MS	[250]
UV/H ₂ O ₂	Anticancer drug, fluorouracil (5-FU)	>99	LC-IT-MS/MS	[352]
Photo-Fenton	Pharmaceuticals, corrosion inhibitors, and biocides/pesticides	97–98	SPE → UPLC-MS/MS and TOC	[255]
Electrochemical Oxidation	Antibiotics, e.g., ofloxacin	~90	UV Spectrophotometry	[356]
TiO ₂ -Solar Photocatalysis	Pharmaceuticals, diclofenac	~90	HPLC-QqQ-MS	[249]

Gahrouei et al. (2024) [357] present significant findings on the effectiveness of various AOPs for removing antibiotics from water. They highlight that AOPs, including ozonation, photo-Fenton processes, UV/H₂O₂, TiO₂ 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 by-products 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 [357].

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 H₂O₂, and (2) GAC. The average removal efficiencies for micropollutants were $76.4 \pm 6.2\%$ for AOP and $90.0 \pm 4.6\%$ 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 2184 bed volumes. For AOP, optimizing UV and H₂O₂ doses significantly improved performance, achieving a removal efficiency of 97.1% with 40 ppm H₂O₂ 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 [310].

4.2.4. 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.

Recent applications of hybrid membrane/adsorption processes have increased, particularly the combination of AC adsorption with MF or UF [358]. 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% [359]. However, a challenge is that some AC fines may be carried over with the treated water, necessitating an additional post-treatment separation process [360]. 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 [361,362]. This approach also improves permeate flux [363], reduces transmembrane pressure (TMP), and lowers energy consumption [183]. Both GAC [185,364,365] and PAC [187,189] 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 [190].

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 [188]. MBRs are particularly effective for removing degradable hydrophilic contaminants through biological processes [285], while NF and RO primarily target hydrophobic contaminants via electrostatic interactions and resistance effects [366]. The integration of MBRs with NF/RO technologies effectively removes both hydrophilic and hydrophobic contaminants from wastewater [285,366]. 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 [367].

Moreover, Asheghmoalla et al. (2024) reviewed integrated and hybrid processes for MPs 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 [368].

5. Conclusions

The daily release of persistent micropollutants has increased significantly worldwide in recent decades due to industrialization and population growth. Among the most concerning of these microcontaminants are microplastics, pharmaceutical compounds, personal care products, industrial chemicals, and endocrine-disrupting compounds (EDCs) in (waste)water—at concentrations ranging from ng/L to µg/L—posing a significant threat to human health and aquatic ecosystems. This issue is further exacerbated by the diverse sources and complex physicochemical properties of organic micropollutants, as well as the inability of conventional water and wastewater treatment systems to effectively remove these contaminants. For instance, they can only be partially eliminated during biological treatment in wastewater treatment plants and remain detectable in effluents even with the latest available technology.

The removal of micropollutants is therefore becoming increasingly important, leading to extensive research into various physicochemical, biological, and hybrid treatment methods aimed at minimizing their environmental impact. Of particular concern in this context are pharmaceutical contaminants, which are frequently found in hospital, household, and industrial wastewater, as well as in natural water bodies. Given the persistence and low biodegradability of pharmaceutical compounds, a single treatment approach is insufficient for complete removal. This highlights the need for advanced and hybrid treatment systems. The detection of pharmaceutically active compounds in aquatic environments has recently intensified research into effective removal methods, including membrane bioreactors (MBRs), ozonation, other advanced oxidation processes (AOPs), physical separation techniques, adsorption using activated carbon, and membrane filtration technologies.

Similarly, the elimination of microplastics from aquatic environments presents a major challenge for conventional treatment processes. Individual microplastic particles can exhibit highly diverse properties and may interact with other micropollutants, such as pharmaceutical residues, pesticides, and other contaminants. This makes microplastics and their removal one of the most complex research areas and challenges in environmental and public health protection.

This review provides an overview of the classification, occurrence, and associated environmental and health risks of micropollutants in aquatic systems, as well as potential remediation methods.

It is a well-established fact that persistent micropollutants cannot be effectively or efficiently removed using conventional treatment methods, despite extensive research in this field. Therefore, there remains an urgent need for the development of efficient eco-technologies to purify various water systems contaminated with micropollutants. In order to achieve efficient micropollutant removal in current wastewater treatment plants, the potential of an additional “fourth treatment stage” is currently being intensively explored, particularly for large-scale treatment facilities.

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Abbreviations

The following abbreviations are used in this manuscript:

AC	Activated Carbon
AnMBRs	Anaerobic Membrane Bioreactors
AOP	Advanced Oxidation Products
AS	Activated Sludge
BEMRs	Bio-electrochemical Membrane Bioreactors
BPA	Bisphenol A
CAFOs	Concentrated Animal Feeding Operations
CAS	Conventional Activated Sludge
CW	Constructed Wetlands
COD	Chemical Oxidation Demand
DAD	Diode Array Detector
EAOPs	Electrochemical Processes
ECs	Emerging Contaminants
EDCs	Endocrine-Disrupting Chemicals
FC	Flocculation
FLD	Fluorescence Detector
FO	Forward Osmosis
FWS-CW	Free Water Surface Constructed Wetlands
GAC	Granular Activated Carbon
GC	Gas Chromatography
HFCW	Horizontal Flow Constructed Wetlands
HRMBRs	High-Retention Membrane Bioreactors
HRMBRs	High-Retention Membrane Bioreactors
IC	Inorganic Carbon
IT-MS/MS	Ion trap mass spectrometer
KOW	Octanol–Water Partition Coefficient
LC	Liquid Chromatography
MBR	Membrane Bioreactor
MD	Membrane Distillation
MF	Microfiltration
MIPs	Molecular-Imprinted Polymers

MLSS	Mixed-Liquor Suspended Solids
MOF	Metal–Organic Framework
MS/MS	Tandem mass spectrometry
MW	Molecular Weight
MWCO	Molecular Weight Cut-Off
NF	Nanofiltration
NP	Nonylphenol
NP	Nonylphenol
NPs	Nanoplastics
NSAIDs	Non-Steroidal Anti-Inflammatory Drugs
OMBRs	Osmotic Membrane Bioreactors
PCs	Pharmaceuticals Contaminants
PAC	Powdered Activated Carbon
PAD	Photodiode Array Detector
PCPs	Personal Care Products
PE	Polyethylene
PET	Polyethylene terephthalate
PhACs	Pharmaceutically Active Compounds
PKa	The Acid Dissociation Constant
PPCPs	Pharmaceutical and Personal Care Products
PP	Polypropylene
PS	Polystyrene
PVC	Polyvinyl chloride
QqQ-MS	Triple-Quadrupole Mass Spectrometry
RO	Reverse Osmosis
SPE	Solid Phase Extraction
SPME	Solid Phase Microextraction
SRT	Sludge Retention Time
SSF-CW	Subsurface Flow Constructed Wetlands
TMP	Transmembrane Pressure
TN	Total Nitrogen
TOC	Total Organic Carbon
TrOCs	Trace Organic Contaminants
UF	Ultrafiltration
UHPLC	Ultra-High-Performance Liquid Chromatography
UPLC	Ultra-Performance Liquid Chromatography
UV–Vis	Ultraviolet–Visible Spectrophotometry
VFCW	Vertical Flow Constructed Wetlands
WWTPs	Wastewater Treatment Plants

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