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Micropollutant Removal Efficiency of Advanced Wastewater Treatment Plants: A Systematic Review

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ABSTRACT

INTRODUCTION: Various review papers have been published regarding the occurrence and fate of micropollutants (MPs). MPs in the aquatic environment are still not well reviewed to generate comprehensive summaries with a special focus on their removal from wastewater using conventional and advanced treatment processes. Therefore, this review aimed to provide a synopsis of the efficiency of the advanced wastewater treatment plants in the removal of MPs.

MATERIALS AND METHODS: A systematic search of published literature was conducted on the National Library of Medicine (NLM) database, Web of Science, *Joanna Briggs Institute (JBI)* database, Scopus, and Google Scholar, based on studies with evidence of removal of MPs in the wastewater treatment process. Screening of the published articles was made using pre-specified inclusion and exclusion criteria.

RESULTS: Amongst the 1545 studies searched, 21 full-length articles were analyzed that showed 7 treatment options related to the removal of MPs from wastewater. MPs from wastewater effluents were successfully and effectively removed by advanced treatment techniques. Advanced Oxidation Processes (AOPs), membrane processes, and adsorption processes have all been shown to be potential solutions for the removal of MPs in advanced treatment plants (WWTPs). But, there are 2 critical issues associated with the application of the advanced treatment options which are high operational cost and the formation of dangerous by-products and concentrated residues.

CONCLUSION: This study identified that the removal of MPs using WWTPs was commonly incomplete with varying removal efficiency. Therefore, the adaptation and scale-up of the cost-effective and efficient combined wastewater treatment technology are vital to creating an absolute barrier to MPs emissions.

KEYWORDS: Advanced wastewater treatment plants, endocrine-disrupting chemicals, micropollutants, pharmaceuticals

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Introduction

A rising number of human and natural substances make up micropollutants (MPs), which are a cause of environmental contamination. They are made up of a variety of materials and may contain substances that do not disintegrate in the environment, such as plasticizers, insulating foams, insecticides, medicines, and drug residues. Endocrine disrupting chemicals (EDCs) and pharmaceuticals and personal care products (PPCPs) are 2 of the most often found anthropogenic contaminants in water.¹

A huge amount of water consumption in different working area releases a significant volume of wastewater loaded with complex mixtures of chemical and biological substances such as heavy metals, disinfectants, reagents, detergents, radioactive markers, X-ray contrast media, hormones antitumor, phenol, chloroform, pharmaceuticals, endocrine disrupting compounds, microorganisms (bacteria, viruses), and biodegradable organic materials (protein, fat, and carbohydrate).²

The quantity and type of wastewater released from working areas vary between and within the countries and this variation can be attributed to the size, activity, and nature of the working area, proportion of in and outpatients, type of institution and specialization, and the prosperity of the country. Hospitals discharge wastewater from medical wards and operating theaters (body fluids and excreta, anatomical waste), laboratories (microbiological cultures, stocks of infectious agents), pharmaceutical and chemical stores; cleaning activities (waste storage rooms), X-ray development facilities, autoclaves, microwave irradiation, chemical disinfection, and laundries.³

Wastewater in terms of quality is categorized as municipal wastewater or domestic sewage. However, due to the presence of hazardous, toxic, and pathogenic factors, this type of wastewater is considered to be a health and environmental issue. Recent studies indicated that hospitals may represent an incontestable release source of many toxic substances in the aquatic environment.⁴



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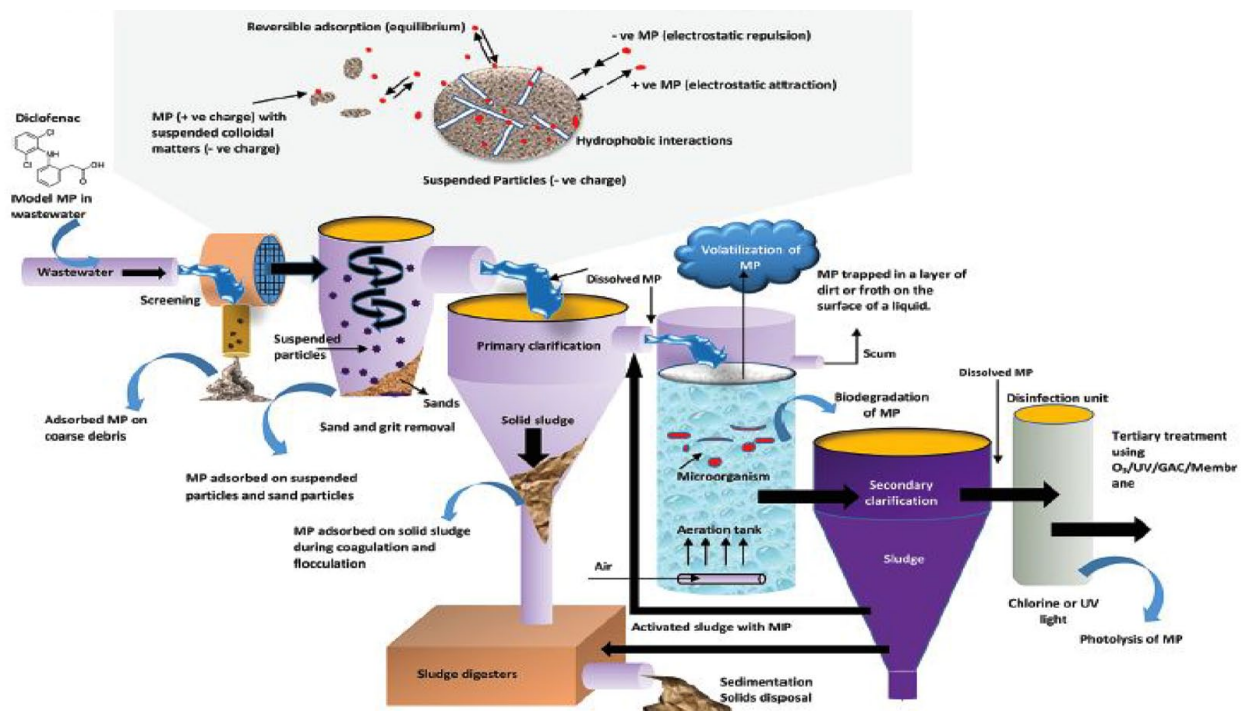


Figure 1. Conceptual model of fate and removal processes of MPs in a typical WWTP.

Many pollutants are not completely metabolized and are disposed into the wastewater. MPs, in particular, are often highly complex products or mixtures of active substances. Some non-biodegradable materials may pass through the sewage of wastewater treatment plants into surface water or reach underground water after the use of sludge as fertilizer.⁵

The disposal of untreated wastewater which contains MPs is also a matter of concern since it constitutes a health risk to the population. Furthermore, pathological, radioactive, chemical, infectious, and pharmaceutical wastes, if left untreated could lead to acute and chronic illness.⁶

These activities may also lead to a risk directly related to the existence of hazardous substances which could have potential health risks and negative effects on the biological balance of the aquatic ecosystem. On the one hand, the toxic metabolites of waste residues passed into the marine ecosystem can cause direct hazards to both the aquatic environment and organisms that reside in it; on the other hand, the impacts can further extend to the land and air arena, in which the terrestrial organisms including human beings and vegetation can be affected by the toxic outcomes.⁶

MPs can be found in the environment in trace amounts between $\mu\text{g/L}$ and ng/L . The low concentration, complexity, and diversity of MPs make it difficult to completely remove them from water and wastewater treatment facilities. Despite being treated at wastewater treatment plants, wastewater from companies and homes is a significant source of pollutants entering the aquatic environment in low- and middle-income countries with established sewage networks. Additionally, combined sewers or leaking sewers may be used to discharge untreated effluent.⁷

Studies on the occurrence and disposition of MPs have grown in recent years as a result of their potential toxicological consequences on both human health and the environment. Wastewater treatment plants (WWTPs) that accept wastewater from home sewage, hospital sewage, agriculture, etc. are the main sources of MPs. These newly developing contaminants are dispersed throughout several environmental matrices after they enter the ecosystem (Figure 1).

MPs can modify their chemistry in response to changing temperatures and surroundings since they are biologically active molecules. This increases their chronic long-term influence and allows them to wind up in natural waters that are utilized by food chains.⁸

In aquatic systems, MPs are frequently present, which is a significant worry everywhere. By using traditional wastewater treatment, the majority of MP is not eliminated, even while a few hydrophobic, biodegradable, or volatile chemicals are effectively removed. As a result, the scientific community and water treatment experts evaluate various ways for reducing contamination. For instance, implementing tertiary or advanced therapies intended to remove MPs.⁹

Wastewater management is now very concerned about the health and environmental effects of MPs on wastewater. Public worries are growing, especially when wastewater effluent is discharged into areas of the environment (such as rivers and streams) that are later used as household water sources by people downwind. Pharmaceuticals are one of the types of MPs that are typically found in aquatic ecosystems. They come from pharmacies, hospitals, and convenience stores, and some of these medications are sold over the counter (eg, acetaminophen, ibuprofen, naproxen, and aspirin). These medications are

created for both human and animal healthcare, but they are not entirely digested by the body. Both leftover medicines and their metabolites are excreted by both humans and animals in wastewater. Waste from the manufacturing process and expired drugs can also be sources of pharmaceuticals.¹⁰

Additionally, significant MPs are the EDCs, which include natural hormones, nonylphenol, pesticides, bisphenol-A, and perfluorooctanesulfonic acids (PFOS). Raw materials like plastic goods and flame retardants discharge these chemicals into the water. As well as being indirectly produced by people and animals. It has been established that these hormones harm human health, similar to how EDC chemicals behave. EDCs produced by the human body are expelled in sewage and dumped into lakes and rivers. Because of this, sewage effluent is regarded as a significant source of MPs pollution.¹¹

Standards and recommendations for discharge do not yet exist for the majority of these chemicals. However, a list of 45 priority compounds or groups of contaminants is announced in the European Union (EU) water Framework Directive 2000/06/CE. The list, which also includes metals, pesticides, phthalates, PAHs, and endocrine disruptors, requires the removal of these substances by 2015 to preserve the purity and good ecological condition of water, as well as to reduce their Ecotoxicity in receiving waterways. Some organic MPs may be toxic and Bioaccumulation due to their persistence, which could have a negative influence on both human health and the environment.¹²

Particulates, carbonaceous chemicals, nutrients, and pathogens are just a few of the many pollutants that WWTPs are made to handle. While it is possible to effectively and consistently eliminate these compounds, it is impossible to control the elimination of MPs. To optimize the treatment procedures and minimize the discharge of these potentially dangerous MPs, it is crucial to evaluate the fate and removal of MPs during wastewater treatment. The best ways to remove MPs are through advanced therapeutic procedures. Ozone, ultrasonic, UV, Fenton reactions, and membrane systems are a few of these techniques. The physicochemical characteristics of the compound, wastewater treatment technology, and process-specific parameters including temperature, sludge retention time (SRT), and organic loading rates all have an impact on the MPs removal by WWTPs (OLR).¹³

This issue can be remedied by identifying and implementing WWTPs that use bio-based materials. These bio-based materials have many uses for micropollutant removal because they include a variety of functional groups, a wide surface area, high stability, and reusability. Two frequently used procedures in the removal of micropollutants utilizing biomaterials are adsorption and degradation.¹⁴

Numerous researchers have demonstrated elevated levels of MPs in the aquatic environment. But, the limited number of available studies demonstrates MPs removal of a single conventional and advanced treatment technologies.^{12,13} Therefore, the current research outlined the overall removal efficacies of advanced treatment technologies for MPs.

These technologies are aimed at minimizing environmental pollution, mainly water and the terrestrial environment. To explore wastewater treatment technology which (1) combining anaerobic treatment method with aerobic system so that Residual and anaerobic pathogen will be removed metabolized by physical and bacteriological activities respectively, and available pathogens will be inactivated and minimized by elevated temperature and other environmental conditions (eg, pH) during the anaerobic digestion process, (2) the SFCW system will be filled with crushed bricks as well as compost, where the remaining pathogens and residue, received from the community will be further eliminated through biological and adsorption process; (3) effective wetland plants will be isolated and planted for further clarification of the wastewater before it is discharged into the municipal system or final disposal site; and (4) The sludge accumulated at the bottom of the anaerobic baffled reactor will be used to make compost (mixed with other non-hospital refuse) so as the humus then will be used to remove pathogens.

Methods

Search strategy and data extraction

This review was conducted according to the recommendations from the Preferred Reporting Items for Systematic Reviews and Meta-Analysis (PRISMA).¹⁵ A systematic search was conducted using NLM database, JBI database, Scopus, and Google Scholar. Three steps of searching were used. In the first step, initial database searches and analyzed the text words contained in the title and abstract, and the index terms used to describe articles. Secondly, all included databases with all identified keywords and index terms were searched. Thirdly, the references of all identified articles were searched to get additional studies (Figure 2).

As inclusion criteria, each study had to: (1) be written in English, (2) report the removal of MPs in the wastewater treatment process. The exclusion criteria were: (1) absence of clear results on the removal efficiency of different treatment processes, (2) specific conditions which couldn't represent the treatment process as it is implemented routinely in treatment systems, (3) articles with no abstracts and/or full text, duplicate studies, and studies with poor quality or not sufficient were excluded.

To minimize bias, the reviewers independently extracted data from the papers included in the review using JBI mixed methods data extraction form. The data extraction form was piloted on randomly selected papers and modified accordingly. For each study, 2 reviewers independently assessed the quality of the included studies. Information like the author, year of publication, type of study, and focus of the study was extracted.

Methodologically quality assessment

Three authors (BB, CY, and BD) independently screened all included titles and abstracts of the entire list of studies identified and reviewed the full texts of articles that met

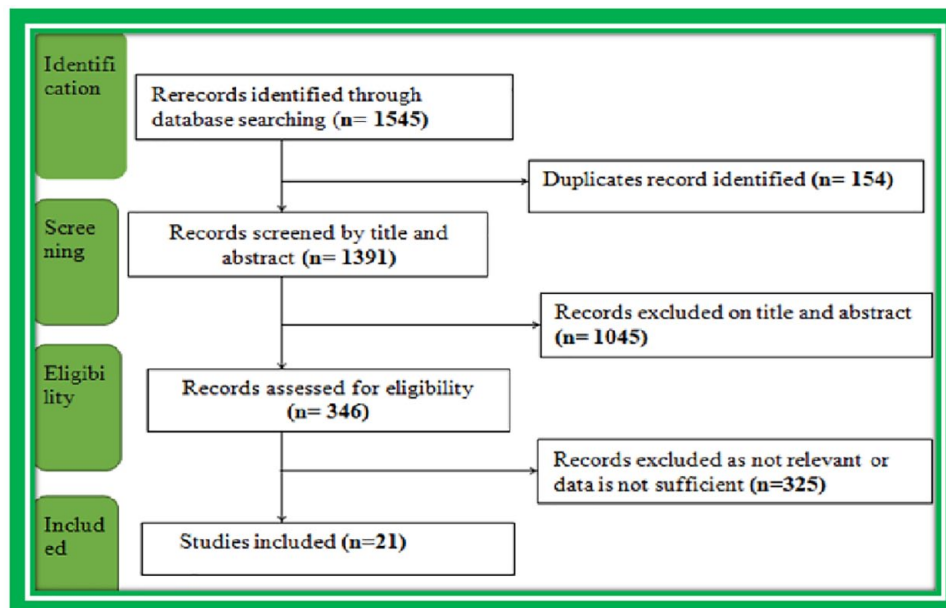


Figure 2. PRISMA flow chart of articles searched and selected for the study.

predetermined inclusion criteria. All references identified through the search were uploaded into the citation manager software Zotero and duplicates were removed. Data extracted for each publication was identified by the title and abstract of the study. Discrepancies were solved by a third person. The reviewers sat together to resolve disagreements during the review process. The methodological quality of the included studies was assessed using the mixed methods appraisal tool (MMAT) version 2018.¹⁸ This method explains the detail of each criterion. The rating of each criterion was done as per the detailed explanations included in the method. Almost all included full-text articles fulfilled the criteria and all included full-text articles were found to be of better quality.

Results and Discussion

Characteristics of the study

A total of 1545 studies were identified, 154 were excluded due to duplication, 1045 were excluded after title, abstract, and full-text screening, and 346 were selected for data extraction. But, 325 studies hadn't sufficient data. So, only 21 selected studies were identified and purported to be eligible for analysis that showed 7 treatment options related to the removal of MPs from wastewater as summarized in the PRISMA flowchart of the method part (Figure 2).

This study discusses recent studies on MP removal in aquatic environments and during water treatment procedures at advanced water treatment plants (AWTPs). Untreated wastewater from sewage and wastewater treatment facilities frequently contains MPs that are dumped into surface waterways. The presence of MPs in surface waters is a serious problem since surface water is often transferred to water treatment facilities (WTF) to make drinking water. Many MPs can stay in tap

water and withstand conventional WTF systems. Drugs and endocrine disruptors are 2 examples of MPs that can be detected in drinking water, ppb, or even ppb levels. Numerous techniques and procedures have been employed to decrease the concentration of MPs in water and prevent the contamination of drinking water, particularly advanced oxidation processes.¹⁸⁻²¹

Coagulation-flocculation

Processes for flocculation and coagulation are frequently employed to improve wastewater treatment facilities by removing suspended particles, colloids, and some dissolved organics that do not settle naturally. Using coagulants such metal salts and synthetic organic polymers, the coagulation process destabilize colloids or emulsions. pH, dosage of the coagulant, and the solution's ionic strength are the variables that have an impact on how well coagulation works.²²

Substantial reduction (about 80%) of musks (such as celestolide and tonalide) in a coagulation-flocculation process for treating secondary effluent as demonstrated by studies on hospital wastewater.²³

The benefits of the flocculation and coagulation processes include better-quality effluent, chemical-free water treatment, the ability to recover metals from solutions, and low electrical current requirements. But the drawbacks include the fact that electrodes are temporary, that numerous circumstances might alter the outcome, that active fine-tuning is necessary, and that an excessive amount of poisonous sludge is produced.²⁴

According to reports, the majority of MPs are ineffectively eliminated during coagulation-flocculation procedures, as seen in Table 1. Apart from a few musks, some medications (such as diclofenac and nonylphenol, a common anti-inflammatory drug frequently found in waste waters) are known to adversely

Table 1. Removal of some MPs by coagulation-flocculation process.

PERFUME AND PHARMACEUTICALS	AMOUNT OF COAGULANT USED	REMOVAL (%)	REFERENCES
Aldrin	100 mg/L Al ₂ (SO ₄) ₃	34	Balest et al ²⁰
	200 mg/L Al ₂ (SO ₄) ₃	46	
	300 mg/L Al ₂ (SO ₄) ₃	43	
Bentazon	100 mg/L Al ₂ (SO ₄) ₃	15	
	200 mg/L Al ₂ (SO ₄) ₃	7	
	300 mg/L Al ₂ (SO ₄) ₃	12	
Diclofenac	50 mg/L FeCl ₃	21	Azizi et al ²¹
Ibuprofen	25 mg/L FeCl ₃	12	
Galaxolide	25 mg/L FeCl ₃	79.2 ± 9.9	
Celestolide	25 mg/L FeCl ₃	77.7 ± 16.8	
Tonalide	25 mg/L FeCl ₃	83.4 ± 14.3	
Naproxen	25 mg/L FeCl ₃	21.8 ± 10.2	
Bisphenol A	200 mg/L FeCl ₃	20	Mungondori et al ²²
Nonylphenol	Not mentioned	90	

affect a number of environmental species already at concentrations of less than 1 µg/L due to their bioconcentration and bioaccumulation nature. The elimination of pesticides was not significantly influenced by the coagulant dose or the operating temperature.²⁵ Despite the modest variations among several types of coagulants at various doses, Fenyvesi et al found that the addition of 25 mg/L FeCl₃ generally produced excellent outcomes. Other than dose, a number of operating parameters, such as mixing conditions, pH, alkalinity, temperature, the presence of divalent cations, and concentrations of destabilizing anions, can affect how well coagulation-flocculation processes function. In general, most MPs are not effectively eliminated by coagulation-flocculation processes (Table 2).²⁶

Advanced oxidation processes

Due to the refractory nature of pollutants, conventional wastewater treatments are unable to adequately remove MPs (Table 3). Advanced oxidation processes (AOPs) can be taken into consideration to solve this issue. Advanced wastewater treatment and water recycling increasingly utilize hydroxyl radical-based advanced oxidation techniques (OH•). These processes are non-selective, fast, and efficient. However, the construction need for more land.³⁶ Multiple AOPs are more efficient than a single oxidation process, according to studies. The combination systems include titanium dioxide, hydrogen peroxide, ozone, and UV light. AOPs based on ultraviolet (UV) light are efficient at removing numerous persistent organic pollutants (POPs) from drinking water.^{37,38}

High degradation rates and non-selectivity are 2 characteristics of advanced oxidation processes, which show certain advantages over traditional therapies. Shown that these procedures have the ability to disinfect, which is crucial for reuse applications involving direct human contact, such as domestic reuse applications.^{32,39-42} Two techniques exist for the UV-based advanced oxidation procedure to eliminate organic contaminants. Some organic pollutants can absorb UV light, which causes their chemical connections to be severed. These pollutants are eliminated; however certain organic MPs can't be broken by UV light alone. According to Odabasi and Buyukgungor,²⁷ oxidizing agents must be applied to these contaminants.

Ozonation

The issue related with ineffective MPs elimination using traditional physicochemical and biological treatments can be helped by ozonation. Both directly and indirectly, ozone can destroy pollutants (mostly by forming the more potent and non-selective oxidizing agent, OH). The disadvantages of using ozone treatment are high cost, toxicity, reactivity and byproducts. Mecha et al⁴² looked assessed the effectiveness of ozonation for the removal of a variety of MPs (UV-filters, perfumes, biocides, and surfactants) from biologically treated gray water.

At an applied ozone concentration of 15 mg/L, the majority of chemicals were considerably eliminated (>79%) from the biologically treated effluent. In a related investigation, Ben et al found that most of the targeted MPs had high removal

Table 2. Removal of some MPs by advanced oxidation process.

TREATMENT APPLIED	PHARMACEUTICALS	REMOVAL (%)	REFERENCE
Chlorine dioxide (ClO ₂) Ferrate (VI)	Naproxen	50	Odabasi and Buyukungor ²⁷
UV/chlorine	17β-estradiol(E ₂), sulfamethoxazole, diclofenac	100 removal E ₂ , sulfamethoxazole, diclofenac	Nam et al ²⁸
UV lamp	Carbamazepine	>99	Choubert et al ²⁹
UV lamp	Diclofenac	95	Lester et al ³⁰
UV/H ₂ O ₂	İbuprofen	100	Meiczinger et al ³¹
	Diclofenac	100	
	Carbamazepine	75	
UV lamp	Ketoprofen	>99	Guillossou et al ³²

Table 3 Removal of some MPs by ozonation.

TREATMENT APPLIED	PERFUME AND PHARMACEUTICALS	REMOVAL (%)	REFERENCE
O ₃ (15 mg/L)	Tonalide	79	Hernández-Leal et al ³³
	Galaxolide	>87	
	Nonylphenol	>79	
O ₃ (5 mg/L): 15 min	Trimethoprim	>90	Silva et al ³⁴
	Carbamazepine	>90	
	Diclofenac	>90	
	Metoprolol	80-90	
	N,N-diethyl-meta-toluamide	50-80	
	Bezafibrate	0-50	
	Sulpiride	>90	
	Mefenamic acid	80-90	
O ₃ (Conc. not mentioned)	İbuprofen	83	Polińska et al ³⁵
	Diclofenac	99	
	Carbamazepine	80	

efficiencies at a lower ozone exposure of 5 mg/L. More than 95% less of the drugs such as carbamazepine, diclofenac, indomethacin, sulpiride, and trimethoprim were present. Metoprolol dosage decreases were not significant. Bezafibrate, on the other hand, was exceedingly resistant to ozonation and was only eliminated by 14%.⁴³

Activated carbon adsorption

The removal of organics from water metrics by adsorption as a unit operation employing either granular or powder-activated carbon (GAC and PAC) is well known. A correlation created by Polińska et al³⁵ by fusing the Polanyi potential theory and

linear solvation energy relationships (LSERs) can be employed in the absence of experimental data on adsorption isotherms.

Both powdered activated carbon (PAC) and granular activated carbon (GAC) have been widely used in adsorption processes. PAC has been considered as an effective adsorbent for treating persistent or non-biodegradable organic compounds. An advantage of employing PAC is that it can provide fresh carbon continuously or can be used seasonally or occasionally when the risk of trace organics is present at a high level. A main disadvantage of powdered activated carbon is that after use it cannot be reactivated and is also sometimes difficult to dig out of water treatment reservoirs. Hernández-Leal et al, evaluated the effectiveness of GAC in treating 2 wastewaters: (a) spiked

Table 4. Removal of some MPs by adsorption processes.

PHARMACEUTICALS	ADSORBENT		REMOVAL (%)	REFERENCES
	DOSAGE (MG/L)	TYPE		
Ibuprofen	Not mentioned	PAC	98	Fenyvesi et al ²⁶
Diclofenac	"	"	98	
Carbamazepine	"	"	75	
Bisphenol A	29g/70.6mL bed volume	GAC	66	Hernández-Leal et al ³³
Tonalide	"		67	
Galaxolide	"		79	
Nonylphenol	"		84	

Table 5. Removal of some MPs by nanofiltration and reverse osmosis.

MEMBRANE	PHARMACEUTICALS	REMOVAL (%)	REFERENCES
Nanofiltration	Diclofenac	60	Balest et al ²⁰ and Silva et al ³⁴
	Naproxen	60	
	Acetaminophen	23	
	Atrazine	97	
Reverse osmosis	Ibuprofen	>99	Balest et al ²⁰ and Poliška et al ³⁵
	Sulfonamides	>93	
	Diclofenac	95	
	Macrolides	>99	
	Bisphenol A	>99	

(0.1-10 µg/L) aerobic effluent in a GAC column operated at low flow and (b) aerobic effluent with real concentrations (40 ng/L to 7.9 µg/L) of MPs in a GAC column. In the first case, removals for all compounds were generally high (>67%), particularly for ethyl, propyl, and butyl paraben, triclosan, caffeine, BP3, PBSA, and 4MBC (>90%). In the second case, most compounds were also effectively eliminated. Specifically, the removal efficiency ranged from 50% (tonalide and nonylphenol) to more than 90% (galaxolide and PBSA). The MPs removal efficiency of activated carbon adsorption, specifically by using PAC (which is the most effective), as summarized in Table 4.

Membrane processes

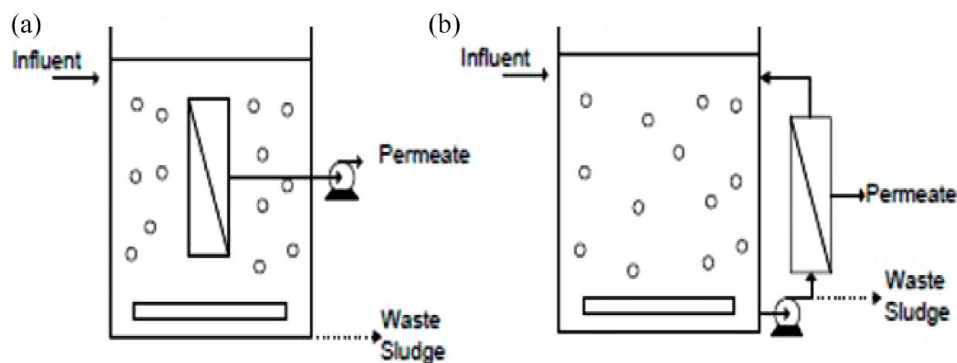
The nature of the membrane, the properties of the solute, the operating environment, and membrane fouling are only a few of the variables that affect the removal of MPs by membrane process. Although adsorption owing to electrostatic repulsion, hydrophobic interactions, and adsorption on the fouling layer can all play a role, size exclusion is the primary mechanism of membrane filtration.^{47,48} The size exclusion method mostly applies to uncharged MPs, although molecular shape should

also be taken into account. MPs adhere to membrane surfaces by hydrogen bonding and hydrophobic contact. By altering the membrane's pore size and surface properties, the presence of dissolved organic carbon and membrane fouling may also boost adsorption. Electrostatic exclusion for membrane surfaces with like charges results from the electrostatic interaction between the compound and membrane surface for a charged MP. Membrane-based processes have a number of benefits, including high efficiency, good adaptability, resistance, and no hazardous intermediates are produced.⁴⁷

Although turbidity can be effectively removed by microfiltration (MF) and ultrafiltration (UF), MPs are typically ineffectively removed by MF and UF because the membrane pore diameters are much bigger than the molecular sizes of MPs. Reverse osmosis (RO) (Table 5) and nanofiltration (NF) have substantially tighter structures than MF and UF. Due to their superior ability to remove pollutants, reverse osmosis and nanofiltration are 2 processes that are frequently utilized in the water reuse sector. For the removal of pharmaceuticals from WWTP effluent, Röhrich et al, tested 2 distinct types of submerged NF flat sheet modules. Compared to carbamazepine, naproxen, and diclofenac (60%) were largely retained (slight removal). As a result, the negatively

Table 6. Removal of some MPs during MBR processes.

WATER TYPE	PHARMACEUTICALS	REMOVAL (%)	REFERENCES
Raw wastewater	Ibuprofen	≈100	Boehler et al ⁴⁴
	Diclofenac	43	
	Carbamazepine	24	
	Sulfamethoxazole	60	
	Trimethoprim	30	
	Estrone	≈100	
	Estriol	≈100	
	Bisphenol A	≈100	
Synthetic wastewater	Bisphenol A	>93.7	Mert et al ⁴⁵
Hospital wastewater	Ibuprofen	>80	
	Diclofenac	<20	
	Carbamazepine	20	

**Figure 3.** Configuration of MBR systems: (a) submerged (immersed) MBR and (b) side stream (external) MBR configuration adopted from El-Sheekh et al¹⁶ and Iorhemen et al¹⁷ as shown in the above figure).

charged membrane surface might reject naproxen and diclofenac, but not carbamazepine.⁴⁹

After applying CAS-UF and MBR processes (Table 6), Sahar et al evaluated the effectiveness of RO. The removal of micropollutants. The elimination efficiencies for the 2 procedures, CAS-UF/RO and MBR/RO, were nearly identical and high: >99% for macrolides, medicines, cholesterol, and BPA; 95% for diclofenac; 97% for SMX; and >93% for both SMZ and TMP. About 28 to 223 ng/L residuals of ibuprofen, cholesterol, diclofenac, salicylic acid, and BPA were found in the permeates from both units despite the extremely efficient RO treatment. This demonstrated that RO wasn't a complete roadblock for MPs. The RO membranes' elimination efficiency and that of NF membranes were extremely similar. While RO was able to remove ionic contaminants with a 99% clearance rate, tight NF had an average retention efficiency of 97% for ionic contaminants. Overall, reverse osmosis exhibits excellent ability to remove MPs in part or in large amounts.⁵⁰

Membrane bioreactor

The membrane bioreactor (MBR) technique is a promising alternative to traditional wastewater treatment, and it is being used more and more for municipal wastewater treatment and reuse (Figure 3). However, several newly emerging MPs in the field of aquatic environment treatment have expressed grave concerns.⁵¹ The advantage of MBR over traditional wastewater treatment, according to El-Sheekh et al,¹⁶ is its effective microbial separation capabilities, excellent effluent quality, absolute control of HRTs, reduced rate of sludge production, flexibility, and little space required.

Membrane bioreactor technology is becoming more and more popular as an innovative technology thanks to numerous full-scale installations for treating municipal and other types of wastewater.^{17,52} The nature of the wastewater, floc size, sludge age and concentration, the presence of anoxic and anaerobic compartments, operating parameters like solid retention time (SRT), high retention time (HRT), pH, temperatures, and

Table 7. Removal of some MPs during attached growth treatment processes.

SYSTEM	PHARMACEUTICALS	REMOVAL (%)	REFERENCES
BAC filter	Diclofenac	≈91	Kovalova et al, ¹⁹ Azizi et al, ²¹ and Falàs et al ⁴⁶
	Carbamazepine	≈95	
	Sulfamethoxazole	≈90	
	Gemfibrozil	≈90	
SBBGR	Bisphenol A	91.8	
ASFBBR	Bisphenol A	27	
MBBR	Diclofenac	≈80	
	Ibuprofen	≈100	
	Naproxen	≈100	
	Ketoprofen	≈100	
	Mefenamic acid	>80	
	Clofibric acid	>60	

Abbreviations: ASFBBR, aerated submerged fixed bed bioreactor; BAC, biological activated carbon; SBBGR, sequencing batch biofilter granular reactor; MBBR, moving bed biofilm reactor.

conductivity contamination can all have an impact on the removal of MPs in MBR.⁵³

The effectiveness of a full-scale MBR's MPs elimination was examined by Boehler et al. For the majority of the MPs, high efficiency (>90%) was noted. However, other substances, such as carbamazepine, diclofenac, sulphamethoxazole, and trimethoprim, were only partially eliminated (24%-60%). As a result, these substances were taken into consideration as prospective markers for assessing the removal of MPs utilizing MBR methods. Boehler et al⁴⁴ and Mert et al⁴⁵ looked at full-scale MBR experiments for hospital wastewater treatment and recommended that rainwater collecting should be separated from water streams with low pharmaceutical contents and that sludge age more than 100 days should be maintained.

Attached growth treatment processes

A viable alternative to traditional wastewater treatment is attached growth technology, which involves attached growth on inert carriers that are either mobile or fixed in the suspension of the reactor (Table 7). With higher oxygen transfer, greater efficiency, suitability for tiny reactor sizes, and the ability to generate microorganisms with relatively low specific growth rates, the connected growth techniques offer an advantage over conventional wastewater treatment.²¹

According to Rattier et al, biofiltration is an effective biological method for removing MPs. In the treatment of water and wastewater, trickling filters, sand filtration, and biological activated carbon (BAC) are often employed techniques. A fixed bed of GAC normally serves as the carrier for bacterial adhesion and growth in BAC filters.⁵⁴ According to Yang et al, BAC

had a remarkable potential for removing PPCPs (such as diclofenac, carbamazepine, sulfamethoxazole, and gemfibrozil) by more than 90% and lowering the risk to human health and the environment. On the other hand, sand filters could only partially remove PPCPs. While empty bed contact time (from 30 to 120 minutes) did not significantly alter the removal of chemicals, dissolved oxygen was reducing the effectiveness of BAC filters. Additionally, the long-term investigation revealed that biodegradation, not adsorption, was the primary mechanism for organic matter and PPCP removal in biofiltration.⁵⁵

Balest et al looked at the Sequencing Batch Biofilter Granular Reactor (SBBGR), another biofilter, for the removal of a number of chosen EDCs. The results showed that in a municipal WWTP, SBBGR outperformed the traditional activated sludge process in terms of EDC removal efficiency. In comparison to the conventional activated sludge process, the SBBGR system had removal efficiencies of 91.8%, 62.2%, 68%, and 77.9% for Bisphenol A, estrone, estradiol, and 4-tert-octylphenol at the municipal WWTP, respectively. The extremely high sludge age was the cause of the SBBGR's remarkable performance (about 160 days). Biofiltration was recommended as an effective treatment technology due to its high performance, which could be used in advanced treatment processes to provide water of higher quality for reuse and lessen the impact of effluent discharge into the environment.²⁰

In order to assess the efficacy of a hybrid moving bed biofilm-activated sludge process for the removal of different MPs, Falàs et al carried out a series of batch studies. It was discovered that the presence of carriers might facilitate the complete biological clearance of some substances. For instance, mefenamic acid, clofibric acid, and diclofenac were not eliminated in the activated

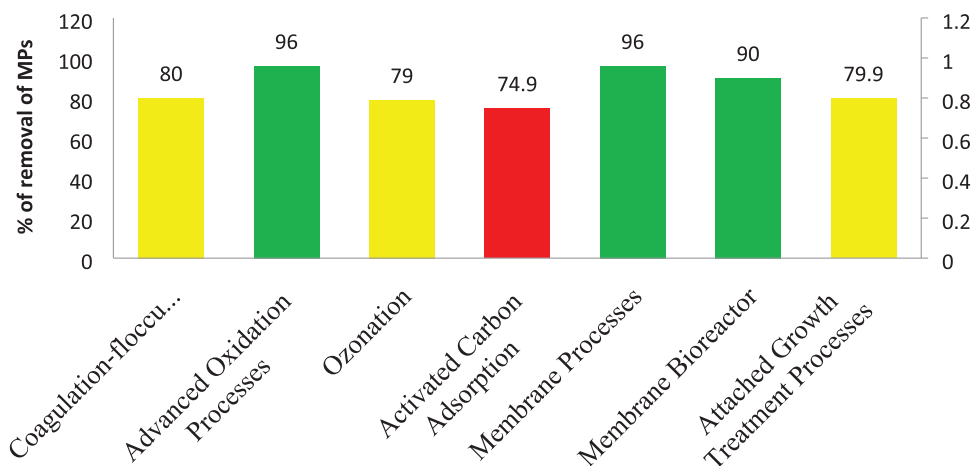


Figure 4. The overall efficacy of WWTPs to remove MPs in %.

sludge reactors, but they were more clearly and quickly removed (at least 60% after 24 hours) in the carrier reactors.⁵⁶

The results of some recent pilot-scale research generally showed that attached growth treatment processes are promising for the removal of MPS, even if they have not been widely and particularly utilized to form MPs removal. The enlarged microbial population can be kept in the system by adding moving carriers or packaging, which promotes the growth of slowly developing microbes for MPs elimination.⁵⁷

Membrane Processes, Advanced Oxidation Processes, or Membrane Bioreactor would be relative useful WWTPs for MPs expulsion with variable rate of diverse MPs (Figure 4). However, those effective technologies are not applicable in low-income countries due to their cost. Overall, MPs elimination via connected growth processes is a method that has a high likelihood of success and is likely to receive greater attention in future studies. Specifically, the adaptation and scale-up of the cost-effective and efficient combined wastewater treatment technology could be relevant for low-income countries to remove MPs.

Conclusion

Due to significant obstacles such the MPs' varied sources, physico-chemical features, accumulation of MPs at environment, and rising concentration in the aquatic environment, single WWTPs have not been able to successfully and efficiently remove the MPs from wastewater effluent. Although it has been shown that modern treatment technologies, including as adsorption processes, AOPs, and membrane processes, are viable alternatives for the removal of MPs, their use is complicated by 2 problems: high operating costs and the production of by-products and concentrated residues. Given their wide range of characteristics (hydrophobicity and biodegradability), low concentrations, and partial removals, MPs are often and unevenly removed in existing WWTPs. Therefore, in order to completely block MPs emissions, it is essential to adapt and scale up the cost-effective and efficient combined wastewater treatment technology, which should be a top priority in contemporary WWTPs. This suggested the use of combined technologies that can eliminate MPs using low-cost physical, chemical, and

biological methods. For instance, anaerobic baffled reactor (ABR) combined with subsurface-flow constructed wetland (SFCW) filled with compost and crushed brick. The anaerobic baffled reactor digests the organic matter anaerobically with the high-temperature generation which will mineralize or degrade long-chain molecules and kill microorganisms with elevated temperature. The effluent discharged from the ABR will be received by SFCW. At this stage, the mineralized antibiotic residues and AMR will be further degraded by the microorganisms in the compost, selected wetland plants, and adsorption into the crushed bricks.

Author Contributions

BB and CY were involved in the conception of the study, reviewing article, and manuscript writing. AA and BD was involved in reviewing articles and manuscript writing. all authors read and approved the manuscript.

Data Availability

Data is available from the corresponding author upon reasonable request.

Ethical Approval and Consent to Participate

Not applicable.

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