International Journal of Chemical and Biochemical Sciences
(ISSN 2226-9614)
Journal Home page: www.iscientific.org/Journal.html

Pharmaceuticals in Wastewater and Their Growing Concerns for Health: A review

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Abstract

Water pollution with substances, such as pharmaceutical residues, is also cited as a significant environmental threat that can endanger the well-being of aquatic organisms and human beings seriously. The pharmaceutical industry is one of the industries that engages in washing and production, which significantly contaminates water. The other sources occur in the forms of medicine manufacturing plants, municipal wastewater by leaching landfill, and hospital waste, which lead to the introduction of various pharmaceuticals to the water bodies. Most of these chemicals are poisonous, cancer-inducing, and bio-accumulative in small doses, leading to serious diseases. The elimination of pharmaceuticals in the wastewater point sources is compulsory in the remediation of the environment since they are hazardous, non-degradable, and everlasting. A few studies have been conducted on how to devise various mechanisms of managing pharmaceutical pollution in wastewater. AOPs are also observed to be efficient and more sophisticated methods for the deterioration of such contaminants. This review provides a discussion on sources of pharmaceutical pollution, water-related issues, and their changes, and their effect on health. It gives the need to conduct additional research to accelerate the effectiveness of decaying and mineralization by creating cost and efficiency-effective green initiators and the advanced integrated systems. This review presents the efficiency of the substances in the removal of pharmaceuticals, coping with the difficulties, constraints, and opportunities, suggesting the emphasis on the environmentally friendly methods and their effective application.

Keywords: Pharmaceutical Contaminants, Wastewater treatment technologies, Health hazards, Advanced removal of pharmaceuticals, Hybrid treatments

Full length article *Corresponding Author*, e-mail: mohamadfawzimahomoodally@duytan.edu.vn, *Doi #* https://doi.org/10.62877/13-IJCBS-26-29-23-13
Submitted: 27-01-2026; Accepted: 28-03-2026; Published: 11-04-2026

1. Introduction

Pharmaceuticals are released and discharged by industries on a large scale, including hospitals, manufacturing plants, and some institutes, making them a major reason of water contamination. As a result, a large concentration of pharmaceutical residues is disposed of in the environment [1]. The use of pharmaceuticals cannot be minimized because of the growing population and industrialization. Reducing their environmental release is very important to minimize risks to human health, ecosystems, and the climate. Studies have shown that these pharmaceutical compounds are intractable and cannot be simply removed via conventional wastewater treatment plants [2]. The detection of pharmaceutical compounds in small amounts in the water poses a major threat to the aquatic systems health [3]. The properties of pharmaceutical wastewater depend on the tools, raw materials, and the specific formulation methods employed [4]. The wastewater from manufacturing of medicines comprises considerable amounts of both biodegradable and non-biodegradable, toxic organic and inorganic chemicals [5]. Pharmaceutical pollutants are commonly removed using chemical, physical and biological methodology.

These methods include biological degradation, membrane bioreactors, adsorption, nano-filtration, phytoremediation, biological treatment, AOPs, and the combination of AOPs with biological treatment. While physical methods are typically employed for organic wastewater, their efficacy is mostly minimal [6]. The studies suggest that the efficiency of biological methods have been explored for pharmaceutical removal from wastewater, but it is very low. Adsorption techniques are an effective and cost-efficient approach for the removal of such pollutants from wastewater [7]. Even though emergence and concentration of these remains within those streams of water has not been of recent occurrence, their continued increase in the water sources and water treatment plants has been reported more often. Recent research on the world environment has been triggered by the high number of pharmaceutical residues in water bodies [8]. The supply of water in the world is limited, considering the fact that there are different pollutants in water, leading to drinkable and potable water. Indeed, the accumulation of pharmaceutical compounds (PhCs) in water and wastewater treatment plants has rendered treated effluents ineffective, as sludge formed is highly hazardous and has a high amount of PhCs. The presence of pollutants in the treated effluents indicates that the traditional methods of treatment were not able to get rid of them, and they are often discharged to the environment in untreated forms [9].

Due to their high removal efficiency on pharmaceutical compounds, AOPs are superior among the selection of treatment methods aimed at the effective removal of emerging intractable molecules [10]. To produce active medicinal compounds, methods such as fermentation, chemical synthesis, and natural extraction are employed. Then, these compounds are turned into different forms that people can take, such as tablets, syrups, ointments, and capsules. During manufacturing, the ingredients are broken down, mixed, pressed into shape, encapsulated, and then packed. This process uses water, binding agents like corn syrup or starch, solvents for coating tablets, fillers to make medicine in dilute form, preservatives to keep it safe, flavoring to make it taste better, and antioxidants [11]. The reduction of human contact with wastewater and the removal of chemical waste and harmful microbes, and pathogens in wastewater may be the plans that could be implemented to limit the threat to human health. As the Environmental Protection Agency (EPA) and the World Health Organization (WHO) proposed that wastewater must be treated such that it meets a given water quality standard to avoid any risks associated with it, especially health risks [12].

The analytical technologies advancement for the checking of pharmaceutical pollutants at trace levels in aquatic environments have driven significant research in this area [13]. Painkillers, antibiotics, and anti-inflammatory drugs are some of the most commonly used drugs across the world. Sewage treatment plants typically extract a fraction of 20-30% of diclofenac because it is poorly biodegradable and has limited adsorption to the activated sludge. Their constant release into the environment exposes organisms to these medications for a long period of time due to their toxicity and activity even at low concentrations. This type of exposure can lead to health complications, such as cancer, weight gain, thyroid complications, and loss of memory, as per recent studies [14]. Traditional bioremediation and physicochemical remediation in infiltration of pharmaceuticals in influents and treated effluents procedures like coagulation, volatilization, adsorption, sedimentation, and filtration do not have the power to eliminate these emerging disruptors. Although chlorination and UV irradiation mechanisms are frequently employed to disinfect final effluents in the treatment plants, the strong oxidative systems/protocols are important in a bid to eliminate pharmaceutical pollutants [15].

2. Sources and Occurrence of Pharmaceuticals in Wastewater

Human use, personal hygiene products, inappropriate medicine disposal, pharmaceutical businesses, hospital waste, and human and animal excretion through urine are the major sources of pharmaceutical pollutants. Their chemical stability and resistance to biodegradation are the reasons for their longevity in water systems [16]. The antibiotics commonly found in wastewater treatment plants are sulfonamides (42.92 ng per liter), ciprofloxacin (390 ng per liter), and tetracycline (330 ng per liter); amoxicillin (2027 ng per liter); acetaminophen and diclofenac (6300 ng per liter). These compounds have been proven to be harmful to aquatic life and human health [17]. Scientists examined the presence of nine different therapeutically active chemicals (PACs) in treated water, drinking water, hospital wastewater, and sewer wastewater. The most common pollutants observed in hospital and sewer effluent were acetaminophen and caffeine, with concentrations up to 159 µg/L; however, drinking water did not contain any residues of these substances [18]. Agunbiade and Moodley conducted an investigation and examined eight acidic pharmaceuticals in wastewater, and sediments from the River in KwaZulu-Natal, South Africa. According to the study, the most common medications determined in the range of 117 µg/L and 25–29 µg/L, respectively, were aspirin and nalidixic acid. Although the effects of these pollutants on humans are still being studied, these findings show that pharmaceutical toxin pollution of water is a global disaster that requires attention because their presence in aquatic media may give rise to serious health-related concerns (Table 1) [19].

3. Distribution and Types of Pharmaceuticals Remains in Wastewater

Antibiotics are among the several medicinal substances that are most commonly found in different countries' waterways. The majority of antibiotic cases were identified in European nations like Greece and Spain, where rates ranged from 6 to 10%. The detection rates were greater, ranging from 6 to 30%, in Asian countries like China and Singapore. Antibiotics were found in four rivers in Hanoi, Vietnam, in extremely high concentrations ranging from 3050 to 16,700 (median 7800) ng/L, which seemed to be far higher than those found in rivers from countries around the globe. Clofibric acid, which is routinely used to regulate blood lipids, has also been documented. This metabolite is biologically active, discovered in wastewater treatment plants, groundwater, and tap water at concentrations of tens of milligrams per litre. This is mainly due to its lasting tendency and strong flotation in aquatic environment [20]. In sample from the downstream site Snitz Creek in Lancaster County, antibiotic concentrations ranged from 130 to 328 ng/L [21]. Vidal-Dorsch et al. studied abundance of emerging concern compounds (CECs) in municipal effluents from Southern California's coastal waterways. Naproxen, gemfibrozil, and atenolol were most commonly found PhCs at quantities more than 1.5 µg/L [22]. Direct release of contaminated municipal wastewater, manufacturing contaminants, & reckless disposal of leftover drugs continue to most significant routes for pharmaceuticals in environment [23]. Elimination of hazardous organic pharmaceuticals from various water sources through innovative oxidation methods is an important aspect of research and development in water and wastewater treatment industries due to increasing retention of these chemicals (Table 2) [24].

4. Pharmaceuticals-Drivers of Human Health Risk

4.1. Antibiotic Resistance

Antibiotic-resistant bacteria are linked to the existence of antibiotics in the environment, which can pose a serious threat to the health of humans. Toxicity is an undesirable consequence of many medications, which are explicitly formulated to produce desired therapeutic benefits. Although pharmaceutical waste can be found in minimal quantities in water bodies like rivers, wells, ponds, and other streams, ongoing exposure to various medicinal drug components, even in trace amounts, can have serious negative effects on one's health [25]. Antibiotics cause selection pressure on microbial communities, with the resistant ones that thrive even at low-concentration conditions. The effluents with residual antibiotics are treated and then have an opportunity to spread resistant bacteria into rivers and streams, which later can intertwine with human populations. The emergence of multidrug-resistant pathogens across the world highlights the severity of this effect [26].

4.2. Long-Lasting Health Concerns

The presence of carcinogenic medications in water treatment facilities may eventually impede the transfer of clean water to recipients because pharmaceuticals can change into endocrine disruptors (EDCs), which are known to block human genetic systems. Pharmaceuticals must completely decompose to sustain human health [27]. It is aimed at filling the gap of research on the elimination of low levels of endocrine harmful steroid hormones, even though scientific studies regarding catalytic and photocatalytic ozone elimination are presented in literature. The human excretions containing medications that have been defecated move through sewage system and end up in wastewater treatment facilities (WWTPs), according to María et al. This has been considered to be among main causes of water contamination. According to Jones et al.and Sirés and Brillas, approximately 90% of pharmaceutical residues made up of unmetabolized and metabolized bio-recalcitrant fragments were retained in the final effluents of water and wastewater treatment plants (WWTPs) following biodegradation, de-conjugation, sorption, and photodegradation processes (Figure 1) [28].

5. Limitations of Conventional Wastewater Treatment Technologies

The composition of wastewater contaminants has changed significantly during the last few decades, reflecting changes in industrial processes and human activity. Several complex and resistant chemicals are gradually replacing traditional wastewater contaminants, including suspended particles and biodegradable organic materials. Since these pollutants like Pharmaceuticals and personal care products (PPCPs), endocrine-disrupting chemicals (EDCs), and microplastics, are frequently ineffectively eliminated by traditional treatment techniques, their presence in wastewater has made treatment even more difficult [29]. Although physical techniques are frequently used to treat high-concentration organic wastewater, their efficacy is restricted. Similarly, biological methods have been investigated for the removal of pharmaceuticals from wastewater; research indicates that their efficacy is limited. Biological treatment is an eco-friendly choice, but it needs a considerable land area, has a slower removal speed, and is not effective for non-degradable contaminants [30]. To meet more than half of global water demand in 2020, over 55% of all wastewater was recycled (United Nations, 2021). Nevertheless, present wastewater treatment technologies were not aimed at removing emerging contaminants (ECs), despite their excellent treatment efficiency for traditional pollutants [31].

To identify the best approach for each medication, the removal effectiveness of six distinct medications using various separation strategies was investigated. Except for acetaminophen and ibuprofen, the NF process yielded the best removal efficiency for all investigated chemicals in membrane processes, while UF was essentially worthless for all of them. With SBR, both compounds showed the maximum removal percentages; however, this treatment had lower removal efficiency for the remaining medications, while the NF method performed better. Except for acetaminophen, nearly every medication tested in the AC tests demonstrated outstanding clearance efficiency, particularly at high AC concentrations of 55 mg/L [32]. When space is not an issue, a constructed wetland is the best treatment option for eliminating physicochemical characteristics. Additionally, it has been discovered that the Electro Bio-Reactor and Sequencing Batch Reactor technologies are highly effective in lowering the BOD, COD, and TSS. Following the tertiary treatment, the values do fluctuate and are more reliant on the age and quality of media filters, which are not always reliable. Therefore, it is better to choose the most effective technique while taking the removal following secondary treatment into account (Figure 2) [33].

6. Emerging treatment technologies for Pharmaceuticals Removal

6.1. Advanced Oxidation Processes

Because advanced oxidation processes (AOPs) are effective at breaking down emerging APIs, they are now being used to remove active pharmaceutical ingredients (APIs) from pharmaceutical or drug products. AOPs use the high reactivity of hydroxyl radicals (HO·) to oxidize organic compounds to nontoxic products. A variety of AOPs, such as Fenton and Photo-Fenton, ozonation, electrochemical, ultrasonic or sonolysis, photo-catalysis, and hybrid AOPs, have been used thus far to remove APIs [34]. Results of study demonstrate that when treating pharmaceutical wastewater, enhanced Fenton process can eliminate up to 85% of COD. When treating industrial wastewater that contains refractory contaminants, photocatalytic AOP technology shows greater degradation efficiency. These sophisticated oxidation technologies can greatly lower secondary pollution produced during treatment process in addition to efficiently breaking down refractory contaminants and lowering reliance on conventional biological treatment techniques (Figure 3) [35].

6.2. Hybrid and Sustainable Treatment Technologies

Emerging technologies focusing on nanocomposite-based adsorbents and biochar to replace them for sustainability. Tunable surface chemistry of agricultural and industrial biomass waste, high porosity, & low environmental footprint Biochar.

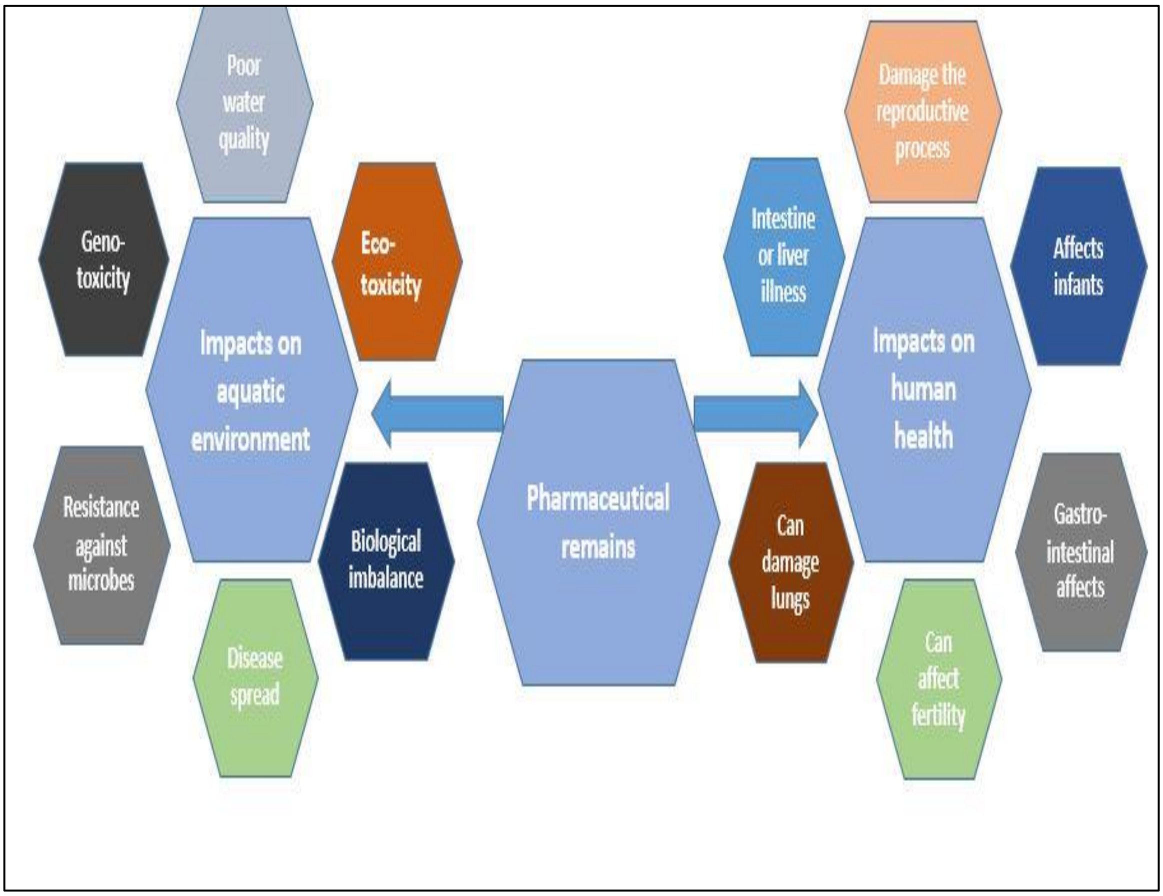

Figure 1: Impacts of Pharmaceuticals on Life

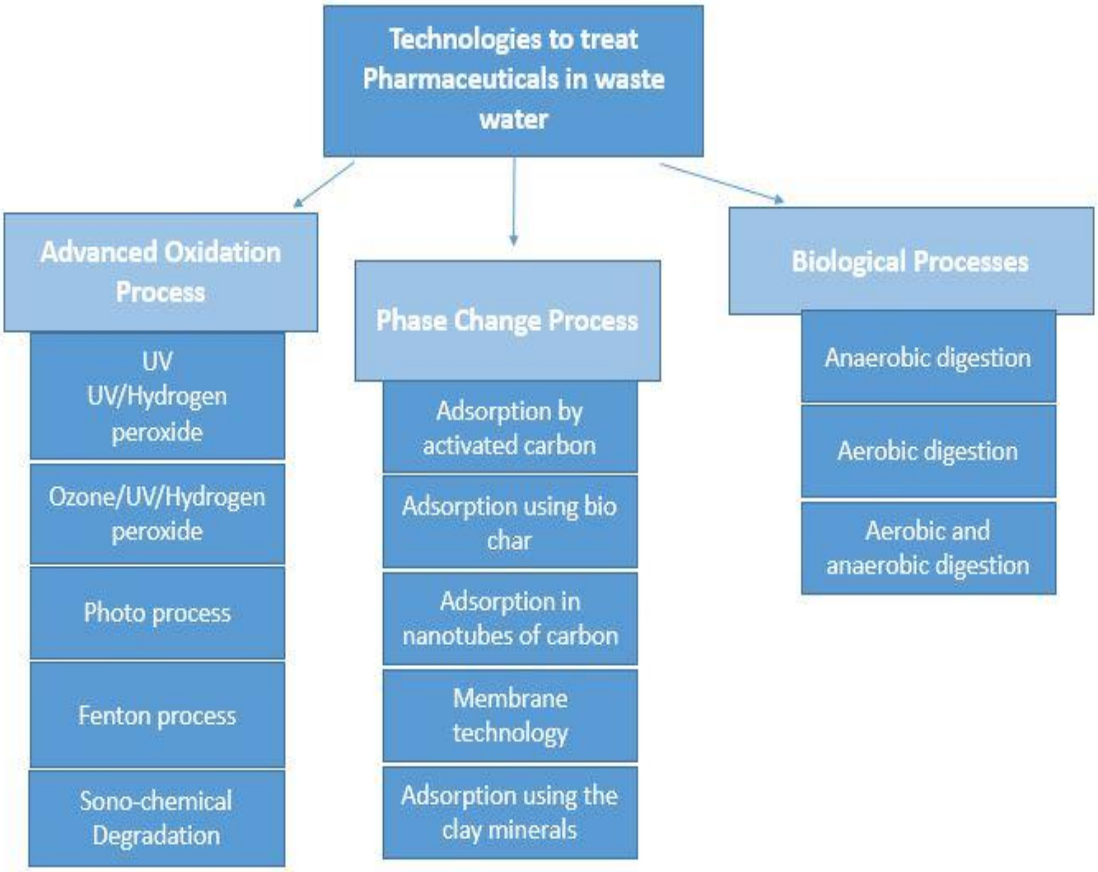

Figure 2: Treatment Technologies for Removal of Pharmaceuticals

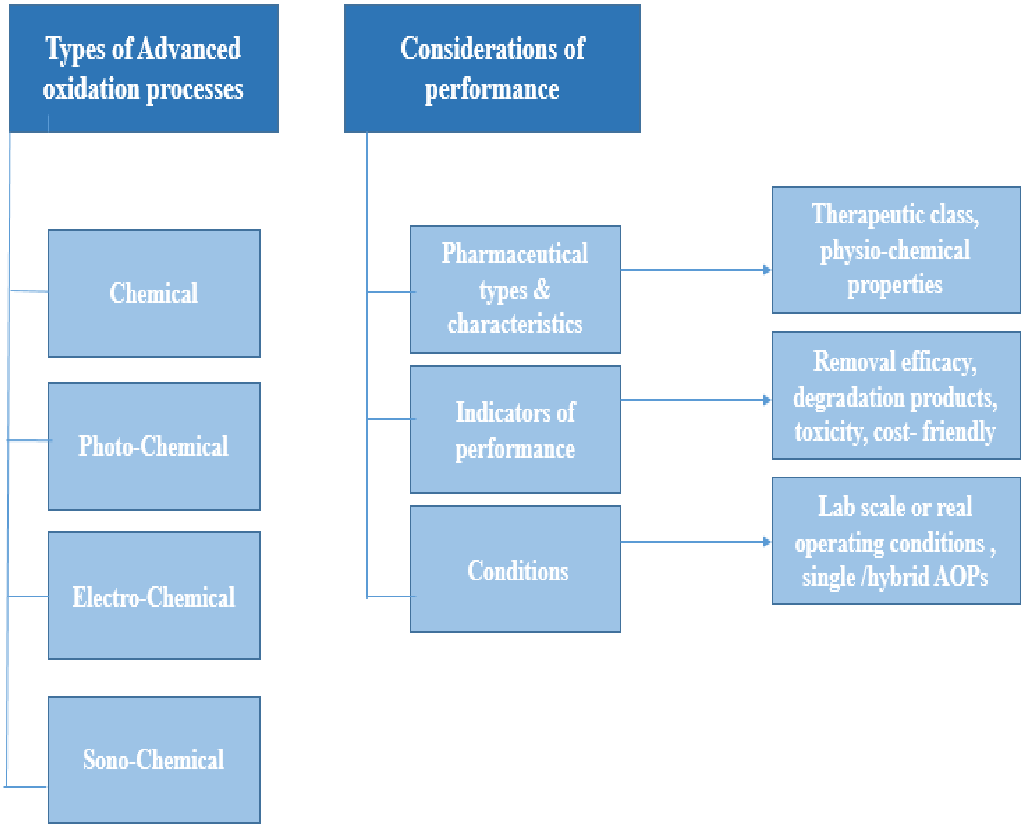

Figure 3: Types of Advanced Oxidation Process

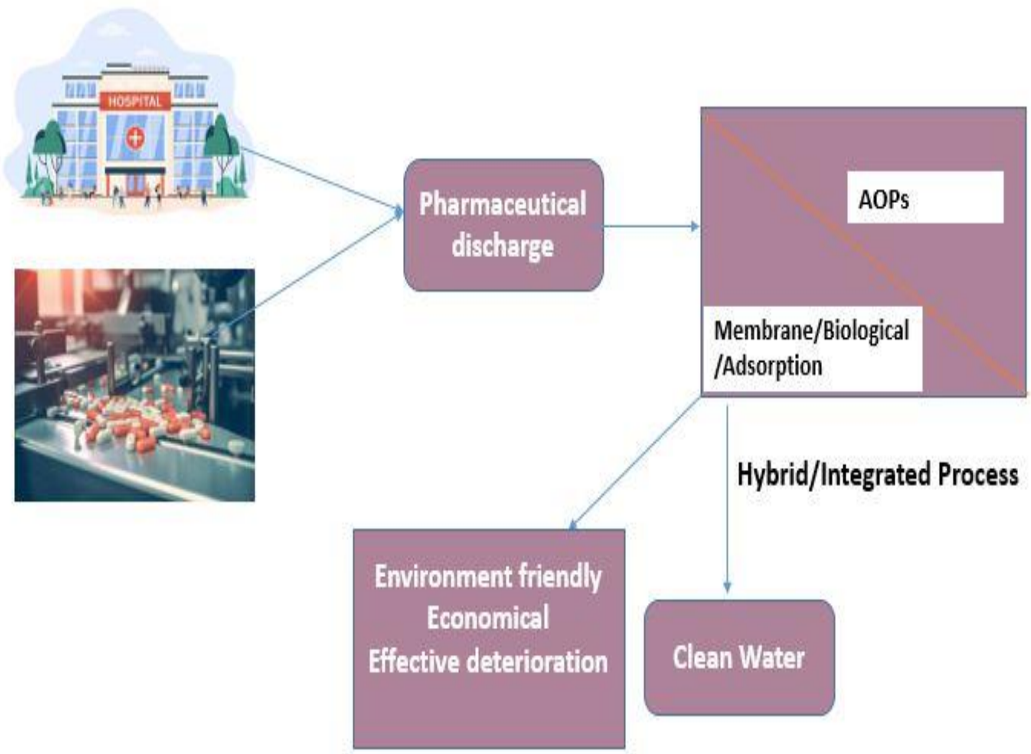

Figure 4: Efficiency of Hybrid treatment technology

Table 1: Occurrence of Pharmaceuticals in Water

Detected Pharmaceutical Compounds	Matrix	Methods of detection	References
Carbamazepine clofibrate; phenazone, aminopyrine;	Wastewater	LC-MS	[39]
Clofibric acid; diclofenac; fenofibrate; fenoprofen, naproxen	Wastewater	GC-MS	[39]
Ibuprofen, clofibric acid; diclofenac; triclosan	Wastewater	GC-MS	[40]
Metoprolol; propranolol; paroxetine	Wastewater	LC–MS–MS	[40]

Table 2: Types of contaminants and their distribution

Group of Contaminants	Detection of compound	Amount	Region	References
Pharmaceuticals/analgesics/ Anti-inflammatory	Naproxen Tylenol Ibuprofen *Diclofenac*	297 ng/L0 293.8 µg/L 14.6 µg/L 0.24–15 µg/L	Colombia Italy Czech Republic	[41]
Antibiotic	Ciprofloxacin Clarithromycin Tetracycline Amoxicillin	0.03 – 125 µg/L 10 – 250 ng/L 8.41 – 69.5 10	Italy Czech Republic Korea	[42]
Personal care products	Triclosan Octocrylene	2µg/L 13.5 µg/L	China Spain	[43]
Industrial chemicals	2-Butoxyethanol Dibenzofuran Longifolene Isophorone	28ong/L 51 ng/L 26 ng/L 39 ng/L	China China China China	[44]

Biochar has been reported as magnetic modified and biochar metal-oxide functionalized biochar to have higher adsorption capacity and high adsorption kinetics, and easy recovery following treatment [36]. It has been demonstrated that nanocomposite adsorbents (graphene, metal-organic frameworks (MOF), and polymer-metal oxide hybrids) can be utilized to achieve extraordinarily adsorbing antibiotics, antivirals, and analgesics. Their huge surface area, variable functional groups, and selective interactions enable them to be readily eliminated at trace concentration levels. However, being aware of potential nano-toxicity, and before such application is widely used, a life-cycle assessment has to be performed [37]. The combined systems are biological-AOP systems, which utilize biological treatment to take away bulk organic matter and AOPs to take away pharmaceutical residues and their metabolites, which are not eliminated by biological treatment. Studies indicate that either pre- or post-integration of AOP makes significant contributions to degradation of pharmaceuticals and reduces utilization and toxicity of the oxidants in effluent. The interest in energy-efficient, scalable means, including solar-driven photo-catalysis, low-voltage electrochemical, and modular hybrid reactor, possible to be used in decentralized treatment of wastewater [38]. Pilot-scale trials that have been carried out during last few years suggest that integrated treatment policies are more effective in real wastewater matrix as compared to individual process systems of a laboratory scale. Nevertheless, despite the possible improvement, there exist challenges to the complexity of the systems, stability in their operation, and financial sustainability. Further research should prioritize optimization of the processes, validation of the process on the actual wastewater, as well as techno-economic and environmental analysis of the same in order to make the full-scale implementation a possibility (Figure 4) [45].

7. Research gaps and future directions

Although AOPs have proven extremely successful in the laboratory, they still present a problem with their application in practice, such as the complexity of the equipment required, high energy costs, catalyst degradation, and secondary pollutants. Fenton-based AOPs have the advantage of operating at a high oxidation capacity and having a relatively low operation cost; still, they have a limited operating pH and result in the formation of iron sludge that increases the cost of disposal and threatens a second contamination [46]. Despite research showing that catalytic ozonation may be effective in eliminating medications, the treatment's byproducts still pose a problem. The development of more stable heterogeneous catalysts, including iron-based composites on carbon material or metal-organic frameworks, can reduce the production of sludge and increase the application. Further research ought to focus on developing cost-effective and cleaner catalysts, optimization of process combination, greater energy efficiency, and enhancement of the capacity of AOPs to respond to real wastewater conditions. The concept of sustainable development and green chemistry will expand, and thus, energy efficiency and environmental friendliness in AOPs are going to gain importance [47]. In future studies, photocatalytic AOPs that driven by sun and electrochemical AOPs in conjunction with renewable energy, e.g., will be addressed. It is expected that AOPs will clean more complex wastewater and new pollutants at a lower cost through development of new catalysts and reaction conditions. It would vigorously promote sustainable utilization of the global water resources, besides aiding in overcoming the current pharmaceutical water pollution problems [48].

8. Conclusions

The sources that produce pharmaceutical residues lead to the outcome that they are deposited in the environment in numerous locations, particularly in factories, labs, and hospitals. These chemicals interfere with the well-being of water catchments and bring about adverse transformations in life forms. Research has established that the levels of these chemicals in the rivers, lakes, and even in the drinking water are gradually rising. These pollutant sources can be removed well using advanced oxidation processes (AOPs). A waste-to-resource approach where biochar and MOFs are used as catalysts in AOPs, i.e., Fenton, persulfate, ozonation, and photo-catalysis, can be of great assistance in breaking down and eliminating pharmaceuticals. AOPs have become more realistic in real-life applications as they have low cost and sustainable catalysts. They are simple to prepare and recycle, and they perform well and can be recycled in actual wastewater environments, hence offering a good solution to large-scale water cleanup. The results may be improved when various AOPs are used concomitantly, as more reactive oxygen species might be generated throughout the treatment. It is significant to test AOPs to eliminate various types of pollution in various water, such as municipal, household, and industrial waste. Another important thing to do is to determine what becomes of the chemicals upon decomposition and whether the results are toxic or not. Very little research has been conducted on the toxicity of these degraded chemicals. Work in the future should focus on finding out these byproducts and evaluating their relative harmfulness to the original chemicals. Of relevance in the gauging of the effectiveness of AOPs in the treatment of wastewater is hydrolysis, the extent to which the chemicals are degraded, a measure of which is the removal of total organic carbon (TOC). Although AOPs are excellent in degrading pharmaceuticals, they can be enhanced by using other techniques, such as adsorption, membrane filtration, and other AOPs in a hybrid or integrated mode.

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