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Removal Pharmaceutical Contaminants from Water and Wastewater: A review

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ABSTRACT


The use of pharmaceuticals is an integral part of global human health care and is used to treat diseases affecting humans and animals. However, in recent years there has been growing concern that they are present in sewage and are an environmental pollutant. Many studies refer to the toxicity and harmful effects of medicines, even at low concentrations, and it is therefore necessary to develop effective techniques for the removal of these pollutants. These studies indicate that biological treatment is not sufficient to decompose natural water and prevent contamination. Chemical processes such as advanced oxidation methods can disable, convert, or mineralize painkillers into simpler molecules and compounds. It is a very complex and costly technique that remains. For physical techniques, it is the most appropriate way to remove pharmaceuticals such as adsorption, which is the most efficient way to remove organic compounds from sewage. It is important to consider the efficiency of the removal and classification of pharmaceuticals during sewage and drinking water treatments. Analyzing view, methods capable of analyzing pharmaceuticals are explained with different efficiencies than those based on advanced biological and chemical adsorption and oxidation.

1. Introduction

Recent research has focused on the effective removal of such pollutants to reduce the threat they pose to organisms [1]. Different forms of organic pollutants such as pharmaceuticals have been detected in a wide range of water supply and because medicines are necessary to maintain human and animal health their production has increased significantly in recent decades. Pharmaceutical materials are released in large quantities into the environment mainly through water and soil and from a variety of sources including human activities, pharmaceutical industry, and hospitals [2] Pharmaceuticals can enter the surrounding areas from different sources such as pharmaceutical plants, hospitals and wastewater treatment plants as shown in the Fig. 1 [3]. The main source is discharge from wastewater treatment plants [4], in addition to pharmaceuticals that are often used as a human drug unchanged or as active metabolites [5] where medicines are introduced, absorbed and distributed into the body and are partially metabolized and eventually unchanged. This makes human secretion a combination of the original compound and its metabolites, all of which enter

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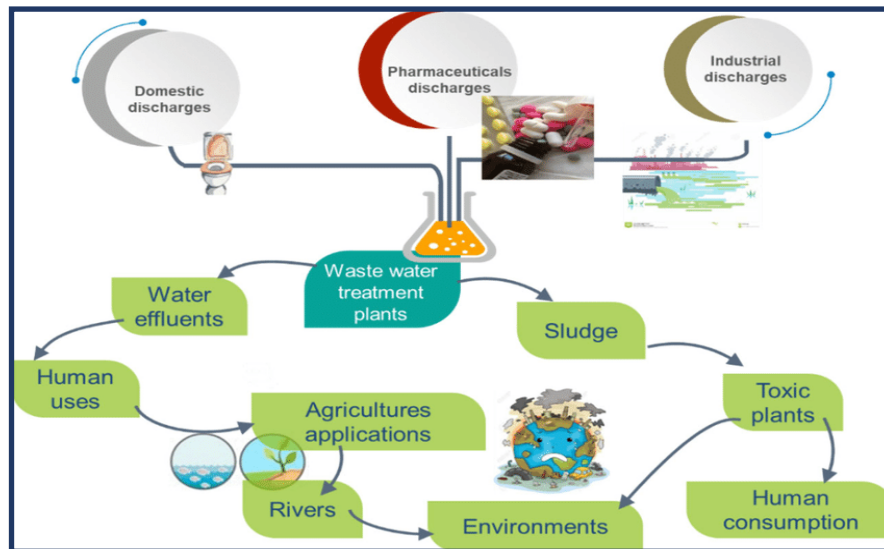


Figure 1: Sources of pharmaceutical contaminants

the sanitation system [6]. Another source of pharmaceutical contaminants is the improper disposal of unwanted or expired medicines and is usually disposed of in wastewater. Therefore, wastewater treatment plants, if not effective, can leak into surface water.[7] Many processes can occur in the hydrological environment of pharmaceuticals such as photo degradation, absorption, mitigation, and transport within the ecosystem with potential absorption of biological species [6]. The bacteria in wastewater can convert pharmaceuticals into products like human metabolites.

The mixture of these compounds makes their removal difficult from wastewater [8]. Many pharmaceuticals have been detected that are not completely removed during wastewater treatment processes and when they enter the environment are not easily degraded [9] This means that more effective therapeutic methods are needed to reduce the potential impacts of these contaminants [10] and therefore this review aims to explain the fundamentals of the methods used to remove pharmaceuticals from aquatic systems.

2- Pharmaceutical Contaminants

Pharmaceutical products are an essential part of life and are used in the treatment of many diseases affecting humans and animals.

Many studies have shown that direct and indirect human exposure to pharmaceuticals through water can cause negative effects. Raw wastewater is the main source of pharmaceuticals found in groundwater and drinking water [11]. In the past three decades, drug residues have been detected in all water sources [12] 3000 pharmaceutical substances have been used in the European Union that have been detected in groundwater, surface water and drinking water. The most widely used species are antibiotics and painkillers for human and veterinary medicines in the world [13] because of its low degradation canvas and high-water love, it is difficult to dispose of pharmaceuticals in water systems using traditional wastewater treatment techniques [14]. It is the most disturbing environmental contaminant, and the type and consumption of pharmaceuticals may vary from country to country [15]. Its consumption has reached approximately 12,500 tons per year. For example, non-centroidal anti-inflammatory drugs and analgesics such as (paracetamol, ibuprofen aspirin, naproxen) [12].

3- Methods of removing pharmaceutical contaminants from water and wastewater

Pharmaceuticals are consumed in different ways such as taking them under the tongue, injection, swallowing or absorption.

Pharmaceuticals are chemical compounds used to treat, prevent, or diagnose a condition or to promote health. Initially medicines were extracted from plants but currently they are organically manufactured, pharmaceuticals are taken either for a certain period of time or regularly as in the case of chronic diseases, it is applied to the ATC system and customized on a basis that met it on solubility, permeability or absorption properties and the type of class used for it. Pharmaceuticals are known to help treat many diseases, leading to their presence in huge quantities of wastewater, which has dangerous effects on the ecosystem and public health [16]. These methods include:

3.1 Conventional Methods

Conventional methods for removing pharmaceutical waste from wastewater include stages of physical, chemical, and biological processes to remove solids and organic materials. Treatment is carried out in primary, secondary, tertiary, or advanced stages to obtain a high level of treatment. Disinfection is performed to remove pathogens as a final treatment step [11].

It has been shown that pharmaceuticals can only be partially removed as wastewater treatment plants are not designed for complete removal [12], initial treatment is needed using activated sludge systems or bioreactors with classic membranes and yet membrane contamination is a significant barrier to their widespread application [17]. Pharmaceutical compounds have volatile thresholds due to their low vapour pressure, some of which, such as ibuprofen and diclofenac, and carbamazine include biodegradable and absorbing limbs according to the [18] where diclofenac was removed 50 % while carbamazine was removed low and in its study [19] note the difficulties in removing various medicines (ibuprofen, TNA and diclofevac) with biological, physical and chemical treatments In addition, the study found that disposal rates differ from dry and humid seasons and in the same context as [20] in their study on palliative and anti-inflammatory drugs such as naproxen and ibuprofen). its limitations in final liquid

waste at 14 wastewater treatment plants from municipalities in Canada. The most important factor for drug degradation is the time of protective retention when protectors are 10 to 15 days old. The biodegradation of pharmaceuticals varies where protectors are aged for the degradation of ibuprofen, aspirin and sulfamethocazol from 2 to 5 days [21] and diclofenac and pyromide required from 5 to 15 day for significant degradation [22] while some are lost medications such as carbamazine and diazepam need more than 20 days [23] and estrogen needs 35 days to remove. From these studies it is concluded that the traditional wastewater treatment methods carried out have not led to desired water quality and pharmaceutical products have not been completely removed through these processes and therefore their survival is dangerous to humans and the environment

Conventional drinking water plants are characterized by multi-stage treatments including filtration, flocculation, and coagulation method. None of these processes have shown effective removal of pharmaceuticals [24]. Ineffective coagulation of sintering to eliminate pharmaceutical products [25]. Coagulation is effective for removing seven categories of antibiotics from tetracycline in optimal conditions with the addition of aluminum poly chloride at a rate of 94% generally. We note from these studies that traditional processing does not completely remove pharmaceuticals before discharging them into the environment, so it is necessary to introduce advanced processing techniques [11].

3.2 Biological Methods

Biological treatment methods depend on biological activity in the disposal of organic pollutants and researchers have studied the possibility of using different bacteria in the analysis of pharmaceuticals and personal care products [26] where bacteria possess an important place in the biosphere through their diverse metabolic capability, they support essential metabolic cycles of all forms of life on Earth [27] which can be an emerging alternative method for removing drug residues from the ecosystem [28] as well as a biological

technique that uses aerobic (protectors), steroid or granular is very effective in wastewater treatment [29]. In the evaluation study of the removal of pharmaceuticals such as caffeine, antienol, azithromycin. Paracetamol, anorfanstatin naproxen fluoxetine, dchlofenac ibuprofen valsartan using in vitro anabolic protectors at a concentration of 13.2 ng/L to 51.8 ng/L, The study showed that biodegradation by adding nutrients to the sludge of pharmaceutical contaminants by Nitrospirae, Actinoptria Verricomicrobia, Firmicutes, Acidobacteria, Proteobacteria, chloroflexi, Bacterioidetes and other types of bacteria involved in the process of removing pharmaceutical compounds at 90% or higher efficiency [30]. According to another study, the efficiency of a gram bacteria, identified as a B1 strain, could degrade ibuprofen and naproxen six days later to concentrate 20 mg/L and 35 days at a concentration of 6 mg/L [31]. Biodegradation can be a possible solution to eliminate these remnants. This process is done by microorganisms that are likely to degrade pharmaceutical waste into biomass, methane, CO₂, water, and various inorganic compounds. While the enzymes of organisms play a vital role and the removal of drugs by biodegradation depends on the desired composition. Simple reactors such as the sand column and membrane bioreactors that have been studied by many researchers can be formed [32]. Environmental factors of temperature, sunlight, humidity, and ultraviolet light were also shown to affect biodegradation [33];[34].

3.3 Electrocoagulation

It is a process in which cations are formed by metal electrodes in an electric field [35],[36]. It produces coagulation materials using metal electrodes, which is many advantages including reduced protectors, automatic treatment and efficient and low operating costs [36], The removal efficiency of the most consumed pharmaceutical products (carbamyzzatin 70%, diclofenac 90%, amoxislin 77%) [37] 0.5 mAh/cm² density, primary concentration of 10 mg/L, Hydraulic

time is 31 hours and in another study by[38]analysis of tetracycline oxide which is an antibiotic, The optimal current density was 20 mAh/cm² for both Anodians, the iron and aluminum had an efficiency of 93.2% and 87.75% respectively, The initial concentration efficiency of oxytetracycline hydrochloride has been studied up to 200 mg/L, It had no significant effect in removing it, the study found the removal of sperofloxin by electrolysis at the density of 15 mAh/cm² and pH of 7.5 and concentration of 60 mg/L, The electrolyte dosage of 0.07 molar Sodium chloride achieved complete removal within 20 minutes) [37],[39].

3.4 Advanced Oxidation

The advanced oxidation process is one of the most effective techniques for degrading and biodegradable hazardous organic pollutants from aquatic environments. One example is ultraviolet activation using hydrogen peroxide and photostimulation using a magnetite compound impregnated with activated carbon [40],[41]. Researchers have repeatedly led the analysis of pharmaceuticals effectively in wastewater and include optical Fenton processes generally more effective than ozone due to the split of the carbon-halogen association resulting from the photon and the increased generation of free hydroxyl roots [42]In addition to UV radiation can generate more effective cases of pharmaceutical removal, but the main problem associated with oxidation processes is that toxic oxidation products can remain in sterile water[43] studies found that different types of oxidation treatments and study[44] and [45] on the removal of paracetamol (astaminophen) two catalysts were used to get rid of the drug (astaminophen) by electrical photocatalysis. Nanotubes of photostimulators were shown and photodegradation of the ingutintestine converter enzyme was faster at pH 3 [44]. And remove paracetamol at 9 degrees. However, the use of photostimulation of paracetamol removed 98% [46] In another study, advanced light-based oxidation processes were used to treat three pharmaceutical products (ibuprofen, carbamyzzine and ciprofloxin) from wastewater.

The treatment showed the 80.4 removal of carbamazepine, an antidepressant drug after 40 minutes of treatment and 89.83% for ibuprofen and the complete removal of ciprofloxacin [47]. As for antibiotics, several studies [48],[49],[50],[51],[52]. The role and efficiency of photostimulation process to remove drugs and antibiotics such as ciprofloxacin which have been completely removed from wastewater at 20 minutes of treatment, when adding iron as a catalyst in wastewater treatment plants [50]. Thus, this drug was eliminated under UV light by combining TiO₂ and zinc oxide catalysts [48]. In a study [52] referred to the removal of antibiotic drugs amoxicillin, ampicillin and cloxacillin under UV and zinc oxide at pH 11.

They noted that the amoxicillin had been removed after 90 minutes of ultraviolet (365-385 nm) treatment [53]. [52] found that the 70% elimination of amoxicillin and metradazol after 120 min of ultraviolet treatment showed the importance of photostimulation, especially in removing drugs in the dark and without light reaction, despite the high cost of this process, many researchers adapt these methods because of their effectiveness in removing most pharmaceutical drugs. (antibiotics, painkillers, non-steroid anti-inflammatory, anti-cancer and beta blockers, disinfectants) [54]. From these different studies we can observe the performance of photostimulation in pharmaceutical removal is important and the technique of using TiO₂ is characterized by inexpensive, low toxicity and high light stability [55]. In addition to the above, ozone therapy is one way of removing medicines. Carbamazepine has been removed at a 96% rate by ozone. It is a powerful oxidizing agent capable of working directly or indirectly. The removal of medicines such as antibiotics and neutral pharmaceuticals in wastewater has been treated. They found that more than 90% of the removal of all pharmaceutical products was done during in addition, the simple use of ozone for water treatment showed good results [56]. They indicated that the complete removal of the drugs (Bezavibrate) in the aqueous solution at a concentration (0.5 mmol/l) could

be reached after 10 min of ozone treatment. From these studies they concluded that ozone could be an economically viable solution for improving protective stability and thus removing anti-steroid drugs such as endometasin by [57], 7 minutes after the lowest dose of ozone.

3.4 Adsorption

Adsorption by solid adsorbents is one of the most efficient processes for treating a wide range of water and wastewater containing pharmaceutical products [13]. Indeed, there are studies of carbonaceous substances used to remove pharmaceutical pollutants that are a financially attractive alternative to wastewater treatment. Then different types are classified into natural and industrial materials. Natural accessories include coal, mud, clay minerals, zeolites, and ores, which are low-cost and abundant natural materials and have great potential to modify and improve their absorption capacity. Industrial accessories are absorbent materials prepared from agricultural products, household waste, industrial waste, sanitation protection, polymeric absorbent materials, and other metallic materials, such as metallic materials and fertilizer waste, sugar industry waste, algae, red clay, sediment, and raw soil [58]. Many adsorbents are used, including:

3.4.1 Activated Carbon

Activated Carbon is one of the most common adsorbent materials used in many industries to remove or restore organic compounds from invasive or liquid currents and has high absorption capacity due to its high surface area and carbonate-forming toxicity due to the presence of stimulant agents and carbonate conditions on the development of porous structures [59]. There has been increasing interest in the production of activated carbon from agricultural by products which has been used as an advantage in several studies. The study of the removal of naproxen by activated carbon from apricot waste as an absorbent substance using adsorption process was removed at a temperature of 50 [60]. In

addition, green synthesis of silver-reduced caraphene has been used to perform well to remove 92% naproxen for absorbents 20 mg and pH 2 and temperature 25[61] while green synthesis of nanoparticles as absorbents indicated the removal of 86% after 60 minutes [62] Also activated carbon was used to remove ibuprofen [63]. The absorbent substance was supplied, and the medicines removed using olive residues as a stimulating Carbone to remove dichlofenac medicines and more effective absorption was achieved at pH 4.5 [62] as for antibiotic drugs, the study indicated that the Amoxylene was completely removed from hospital wastewater and aqueous solutions using magnetic adsorption of carbon produced from the olive nucleus[64] . At maximum concentration of absorbent 0.5 mg/L and contact time 90 minutes and pH of 6 [64] . In a study, an active carbon transposed from pomegranate shells activated by KOH was used to remove spruoxacin from the aqueous system[65]. Experimental results revealed a score of pH 8, which accelerated the electrostatic attraction between sproxacin and activated charcoal and suggested that the optimum dose (0.5%) was achieved.

3.4.2 Clay

Clay is an inexpensive and widely available substance for pharmaceutical removal [66]. Where a lot of species and clay materials such as (elite, renolite, stapulite, caultite, fermiculite mentomoretite) are already used to remove a group of micro-organic pollutants. Mentomoretite is the most promising adsorbent for further studies using a clay-based absorbent substance to remove pharmaceutical compounds . In a study to remove ibuprofen, dichlofenac, cotophene and carbamyazine, fermcholate showed the best absorbent substance for these medicines for modified clay substances[67]. Moreover, adsorption of metaphic acid using two different mud substances showed that both adsorbents were able to remove drugs from the water medium while vermiculite provided a high absorption capacity of about 70%. A study was conducted on the adsorption of cyprusoxacin on a compound derived from medical waste fortified

with bentonite clay. The results showed an increase in removal efficiency of 40% from non-supported clay due to the entry of cyprooxacin molecules into the clay layer in the active pores present due to hydrostatic pressure The removal efficiency with clay increased at pH 6 [68].

5-4-3 Biochar

Biochar has been used as an absorbent substance for pharmaceutical compounds and has proved [13] that there are many factors that may affect pharmaceutical adsorption to biochar including pH and heat, raw material and preparation conditions and the presence of ions and pharmaceutical structures where biochar-dependent substances have been used by [69] to remove antibiotics such as (sulfanamides) and tetracycline in aquatic environments. Biochar has been shown to be a successful absorbent substance and can thus have excellent potential as an absorbent substance due to its ability to bloat in water leaving more space for adsorbed substances [70], so adsorption of pharmaceuticals with biochar often leads to high efficiencies and removal capability without generating toxic products.

3.5 Bionanotechnology

Recent progress in nanomaterials and nanotechnology has provided amazing results for the development of new strategies for the effective degradation of pharmaceutical contaminants with excellent physical and chemical propertie[71] . Nanotechnology is the new wave of innovation for the coming decades, and related technologies are being used to reduce pollution of groundwater, wastewater, and drinking water. Nanotechnology has great potential to prevent, treat, and clean pollution effectively, and nanomaterials play a fundamental role in recent research efforts to develop Effective treatment techniques: Pharmaceutical contaminants can be treated using nanosystem techniques such as adsorption of nanomaterials, photolysis, nanofiltration, etc [72].Currently, other nanomaterials are used such as nanosorbents,

nanocatalysts, nanomembranes, magnetic nanoparticles, beads, films, and molecular polymers. To remove toxic metals, microbes, and dissolved organic and inorganic materials from wastewater and drinking water [73]. Among the nanomaterials that are used are silver and titanium dioxide to disinfect and remove contamination with organic compounds, and the use of iron nanoparticles to treat wastewater. Likewise, a nanofiltration membrane can be used to reduce Hardness, color, odor, and heavy metals [74]. Nano technologies have great potential to make industrial wastewater more efficient, and they can be used for their unique properties, including efficiency, effectiveness, and being environmentally friendly as a promising alternative to water treatment. These technologies include:

3.5.1 Nanofiltration

Nanofiltration has been discovered and widely used to remove pharmaceutical contaminants in wastewater, because it is highly efficient, easy to operate, and provides good retention of ionic salts. The choice of these membranes depends largely on the nature and size of the contaminants, and nanomaterials are incorporated into these membranes to enhance flow and mitigate membrane contamination as a sustainable approach [75],[71].

3.5.2 Carbon nanotubes

Carbon nanotubes have unique physical, chemical, mechanical and electrolytic structure properties and have high adsorption capacity to remove pharmaceutical pollutants from wastewater. Studies have shown the removal of pefloxacin, sulfamethasone, amoxicillin, thorfloxacin, and tetracycline. Adsorption models of activated carbon and carbon nanotubes are considered highly effective in removing pharmaceutical contaminants [76].

4. Membrane bioreactor

The MBR membrane bioreactor has become important in the field of sewage treatment because it is a process that has a

higher efficiency than biodegradation, occupies less space and produces a small amount of sludge [77]. The MBR method is considered a combination of the membrane microfiltration process and ultrafiltration in treatment biological wastewater using activated sludge. In recent years, MBR has been widely used to treat municipal, industrial, and pharmaceutical wastewater [78],[79]. It was used to treat pharmaceutical wastewater using MBR with a size of 20 m³ for a period of 140 days, and a COD removal of 96% was achieved. The BOD was 99% and the concentration of various liquid suspended solids in the tank was maintained. In turn [80], demonstrated the effectiveness of the MBR system in removing pharmaceutical waste particles from wastewater with a stable and satisfactory operating system [81], where he conducted an experiment on the ultrafiltration bioreactor and showed an efficiency of 82% for hydroxybupropione, and 98 % for ibuprofen, 95% for metformin, and 92% for valsartan, respectively. This indicates the effectiveness of the MBR technology in removing pharmaceutical contaminants from wastewater.

In addition to the above, recent studies indicate the feasibility of using MBR on a large scale in treating and removing pharmaceutical contaminants, showing promising results, as a study [82] was found in which eight pharmaceutical preparations and two polycyclic perfumes were separated using MBR, and a comprehensive comparison was conducted. With wastewater treatment technology, the MBR method is the best compared to traditional systems and has shown a better retention time of solids within the reactors. In a study [83], MBR was used to remove acetaminophen, caffeine, metformin, 2-hydroxyibuprofen, paraxanthin, ibuprofen, naproxen, clarthyrombin, atenol, and carbamazine from different water streams from a wastewater plant.

5. Advantages and disadvantages of pharmaceutical removing methods.

The removal of pharmaceutical substances from wastewater depends on

different treatment processes leading to different efficiencies, as sewage treatment plants are not designed to remove pharmaceutical substances form a complete [12]. When comparing processing methods, we find that adsorption due to its simple design can involve a low investment in cost and space required [84] The adsorption process is characterized by a processing process with applicability to low concentrations of absorbed material and its suitability for continuous treatment and the ability to reuse and regenerate absorbent material. Adsorption is a low-energy operation that can be very effective leading to a removal rate of up to 90%. Large quantities of wastewater can be treated with photostimulation (which needs electrodes and cells), The greatest advantage of photostimulation is that it uses TiO

semiconductor material, which is non-hazardous, environmentally friendly and inexpensive [85] . However, this process has many defects, including the need for an acidic medium or pH. (0_3) resulting in the production of a large amount of protectors, While the advanced oxidation process leads to a high degradation of pharmaceuticals but is difficult to apply widely in wastewater plants due to oxidized by-products and high operating cost, A common problem between advanced oxidation removal processes and methods is their high cost due to the increased demand for electric power and as in the figure (2), which illustrates the advantages and disadvantages of different processing methods.

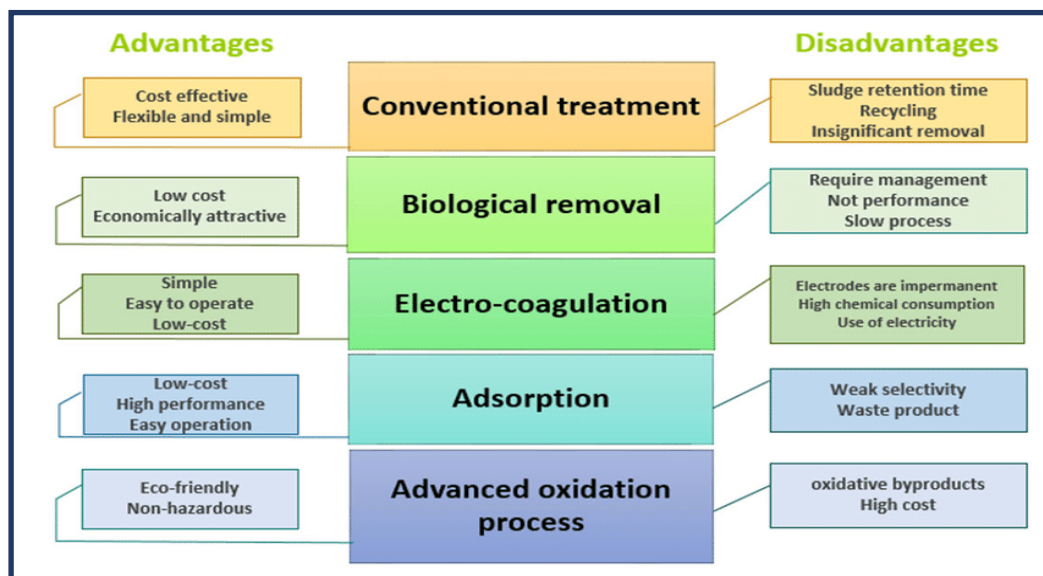


Figure 2 Advantages and disadvantages of pharmaceutical processing methods.

Table 1 showing pharmaceutical contaminants and removal methods.

Process	Pharmaceutical contaminants	Requirements of process	Ref.
Conventional Methods	diclofenac carbamazine	physical, chemical, and biological processes to remove solids and organic materials	[18]; [19]
	ibuprofen		[20]; [21]
	naproxen		[22]; [23]
	aspirin sulfamethocazol		[24]
Biological treatment	caffeine, antienol, azithromycin.	depend on biological activity	[30]
	Paracetamol, anorfanstatin		[31]
	naproxen fluoxetine, dchlofenac		
Electrocoagulation	ibuprofen valsartan	It is a process in which cations are formed by metal electrodes in an	[37]; [38] [39]
	carbamyzzatin , diclofenac, amoxislin		

The advanced oxidation process	oxytetracycline hydrochloride, sperofloxin paracetamol ibuprofen, carbamyzine, ciprofloxin antibiotics amoxicillin, ampicillin, cloxacillin metradozol . (antibiotics, painkillers, non-steroid anti-inflammatory, anti-cancer and beta blockers, disinfectants)	electric field. It produces coagulation materials using metal electrodes one of the most effective techniques for decomposing and removing dangerous and biologically resistant organic pollutants from aquatic environments. Examples are activation with ultraviolet radiation using hydrogen peroxide and photocatalysis using a magnetite compound impregnated with activated carbon. ozone therapy	[44]; [45] [46]; [47] [48]; [49] [50]; [51] [52]; [53] [54]; [55] [56]; [57]
Adsorption	Naproxen, Ibuprofen Dichlofenac the Amoxylene spruoxacin cotophene carbamyzine cyprusoxacin sulfanamides tetracycline	Activated Carbon Clay Biochar	[60]; [61] [62]; [63] [64]; [65] [67]; [68] [69]; [70]
Bionanotechnology	of pefloxacin, sulfamethasone, amoxicillin, thorfloxacin tetracycline	nanosorbents, nanocatalysts, nanomembranes, magnetic nanoparticles, beads, films, molecular polymers	[76]
Membrane bioreactor	hydroxybupropione, ibuprofen, metformin, valsartan acetaminophen, caffeine, metformin, 2-hydroxyibuprofen, paraxanthin, ibuprofen, naproxen, clarthyrombin, atenol, and carbamazine	combination of the membrane microfiltration process and ultrafiltration	[81] [82] [83]

6. Conclusion

We conclude from this review that the presence of pharmaceuticals significantly in the wastewater of homes, hospitals and pharmaceutical industries has resulted in significant pollution in the water sources available to us. This has raised concern for human health and the environment and has been suggested many ways to reduce the presence of medicines in water. Saving the environment and living organisms from it and maintaining natural balance. Many ways of removing pharmaceuticals from contaminated wastewater have been introduced and discussed, such as traditional methods of biodegradation, coagulation, ozone, photostimulation and how to treat each of them. Many methods have been considered, such as adsorption and advanced oxidation.

Advanced oxidation is an excellent method of removing pharmaceutical products such as antibiotics, non-steroid anti-inflammatory and painkillers even when micro-concentrations as well as the advantages and disadvantages of each technique to reduce the toxicity of certain byproducts of treatment methods, through this review we find that, based on many studies, one technique cannot be relied upon to treat water contaminated with medicines.

In addition, we conclude from this review that the adsorption process is one of the most efficient methods for removing pharmaceutical preparations, as it relies on environmentally friendly and inexpensive materials and compounds that do not contain toxic by-products.

References

- [1] A. Jahanban-Esfahlan, R. Jahanban-Esfahlan, M. Tabibiazar, L. Roufegarinejad, and R. Amarowicz, "Recent advances in the use of walnut (*Juglans regia* L.) shell as a valuable plant-based bio-sorbent for the removal of hazardous materials," *RSC advances*, vol. 10, pp. 7026-7047, 2020.
- [2] R. C. Ferreira, O. Couto Junior, K. Carvalho, P. Arroyo, and M. Barros, "Effect of solution pH on the removal of paracetamol by activated carbon of dende coconut mesocarp," *Chemical and Biochemical Engineering Quarterly*, vol. 29, pp. 47-53, 2015.
- [3] A. Deegan, B. Shaik, K. Nolan, K. Urell, M. Oelgemöller, J. Tobin, et al., "Treatment options for wastewater effluents from pharmaceutical companies," *International Journal of Environmental Science & Technology*, vol. 8, pp. 649-666, 2011.
- [4] J. L. Tambosi, L. Y. Yamanaka, H. J. José, R. d. F. P. M. Moreira, and H. F. Schröder, "Recent research data on the removal of pharmaceuticals from sewage treatment plants (STP)," *Química Nova*, vol. 33, pp. 411-420, 2010.
- [5] P. N. Karungamye, "Methods used for removal of pharmaceuticals from wastewater: A review," *Applied Journal of Environmental Engineering Science*, vol. 6, pp. 6-4 (2020) 412-428, 2020.
- [6] G. McEniff, W. Schmidt, and B. Quinn, "Pharmaceuticals in the aquatic environment: a short summary of current knowledge and the potential impacts on aquatic biota and humans," 2015.
- [7] Nielsen, L., & Bandosz, T. J. (2016). *Chemical Engineering Journal*, 287(November), 139–147.
- [8] E. Zuccato, S. Castiglioni, R. Fanelli, G. Reitano, R. Bagnati, C. Chiabrando, et al., "Pharmaceuticals in the environment in Italy: causes, occurrence, effects and control," *Environmental Science and Pollution Research*, vol. 13, pp. 15-21, 2006.
- [9] S. Karthikeyan, M. A. Kumar, P. Maharaja, T. Partheeban, J. Sridevi, and G. Sekaran, "Process optimization for the treatment of pharmaceutical wastewater catalyzed by poly sulphate sponge," *Journal of the Taiwan Institute of Chemical Engineers*, vol. 45, pp. 1739-1747, 2014.
- [10] P. Amouzgar and B. Salamatnia, "Journal of Molecular and Genetic Medicine," *Journal of Molecular and Genetic*, vol. 4, p. 001, 2015.
- [11] F. Mansouri, K. Chouchene, N. Roche, and M. Ksibi, "Removal of pharmaceuticals from water by adsorption and advanced oxidation processes: State of the art and trends," *Applied Sciences*, vol. 11, p. 6659, 2021.
- [12] M. Patel, R. Kumar, K. Kishor, T. Mlsna, C. U. Pittman Jr, and D. Mohan, "Pharmaceuticals of emerging concern in aquatic systems: chemistry, occurrence, effects, and removal methods," *Chemical reviews*, vol. 119, pp. 3510-3673, 2019.
- [13] A. H. Naeini and S. A. H. Moradi, "Adsorption Method for Removal of Pharmaceuticals from Wastewater," *Iranian Journal of Materials Science & Engineering*, vol. 20, 2023.
- [14] R. Szabó, C. Megyeri, E. Illés, K. Gajda-Schranz, P. Mazellier, and A. Dombi, "Phototransformation of ibuprofen and ketoprofen in aqueous solutions," *Chemosphere*, vol. 84, pp. 1658-1663, 2011.
- [15] M. Petrovic, M. J. L. De Alda, S. Diaz-Cruz, C. Postigo, J. Radjenovic, M. Gros, et al., "Fate and removal of pharmaceuticals and illicit drugs in conventional and membrane bioreactor wastewater treatment plants and by riverbank filtration," *Philosophical Transactions of the Royal Society A: Mathematical, Physical and Engineering Sciences*, vol. 367, pp. 3979-4003, 2009.
- [16] B. Liao, A. Bokhary, L. Cui, and H. Lin, "A Review of membrane technology for integrated forest biorefinery," *Journal of Membrane Science and Research*, vol. 3, pp. 120-141, 2017.
- [17] B. Petrie, E. J. McAdam, M. D. Scrimshaw, J. N. Lester, and E. Cartmell, "Fate of drugs during wastewater treatment," *TrAC Trends in Analytical Chemistry*, vol. 49, pp. 145-159, 2013.
- [18] A. Tauxe-Wuersch, L. F. De Alencastro, D. Grandjean, and J. Tarradellas, "Occurrence of several acidic drugs in sewage treatment plants in Switzerland and risk assessment," *Water research*, vol. 39, pp. 1761-1772, 2005.
- [19] C. D. Metcalfe, B. G. Koenig, D. T. Bennie, M. Servos, T. A. Ternes, and R. Hirsch, "Occurrence of neutral and acidic drugs in the effluents of Canadian sewage treatment plants," *Environmental Toxicology and Chemistry: An International Journal*, vol. 22, pp. 2872-2880, 2003.
- [20] B. Strenn, M. Clara, O. Gans, and N. Kreuzinger, "Carbamazepine, diclofenac, ibuprofen and bezafibrate-investigations on the behaviour of selected pharmaceuticals during wastewater treatment," *Water Science and Technology*, vol. 50, pp. 269-276, 2004.
- [21] T. A. Ternes, M. Meisenheimer, D. McDowell, F. Sacher, H.-J. Brauch, B. Haist-Gulde, et al., "Removal of pharmaceuticals during

- drinking water treatment," *Environmental science & technology*, vol. 36, pp. 3855-3863, 2002.
- [22] N. Kreuzinger, M. Clara, B. Strenn, and H. Kroiss, "Relevance of the sludge retention time (SRT) as design criteria for wastewater treatment plants for the removal of endocrine disruptors and pharmaceuticals from wastewater," *Water Science and Technology*, vol. 50, pp. 149-156, 2004.
- [23] G. Kooijman, M. De Kreuk, C. Houtman, and J. Van Lier, "Perspectives of coagulation/flocculation for the removal of pharmaceuticals from domestic wastewater: A critical view at experimental procedures," *Journal of Water Process Engineering*, vol. 34, p. 101161, 2020.
- [24] K.-J. Choi, S.-G. Kim, and S.-H. Kim, "Removal of antibiotics by coagulation and granular activated carbon filtration," *Journal of hazardous materials*, vol. 151, pp. 38-43, 2008.
- [25] M. C. Molina, L. F. Bautista, M. Catalá, M. R. de las Heras, P. Martínez-Hidalgo, J. San-Sebastián, et al., "From laboratory tests to the ecoremedial system: the importance of microorganisms in the recovery of PPCPs-disturbed ecosystems," *Applied Sciences*, vol. 10, p. 3391, 2020.
- [26] R. M. Martínez-Espinosa, "Microorganisms and their metabolic capabilities in the context of the biogeochemical nitrogen cycle at extreme environments," *International journal of molecular sciences*, vol. 21, p. 4228, 2020.
- [27] Y. Nancharaiah, M. Sarvajith, and T. K. Mohan, "Aerobic granular sludge," *Current science*, vol. 117, pp. 395-404, 2019.
- [28] O. Muter, P. Ingus, T. Selga, A. Berzins, D. Gudra, I. Radovica-Spalvina, et al., "Removal of pharmaceuticals from municipal wastewaters at laboratory scale by treatment with activated sludge and biostimulation," *Science of the total environment*, vol. 584, pp. 402-413, 2017.
- [29] A. Marchlewicz, D. Domaradzka, U. Guzik, and D. Wojcieszynska, "Bacillus thuringiensis B1 (2015b) is a gram-positive bacteria able to degrade naproxen and ibuprofen," *Water, air, & soil pollution*, vol. 227, pp. 1-8, 2016.
- [30] K. M. Onesios, J. T. Yu, and E. J. Bouwer, "Biodegradation and removal of pharmaceuticals and personal care products in treatment systems: a review," *Biodegradation*, vol. 20, pp. 441-466, 2009.
- [31] F. Raza, H. Zafar, Y. Zhu, Y. Ren, A. -Ullah, A. U. Khan, et al., "A review on recent advances in stabilizing peptides/proteins upon fabrication in hydrogels from biodegradable polymers," *Pharmaceutics*, vol. 10, p. 16, 2018.
- [32] A. George, P. A. Shah, and P. S. Shrivastav, "Natural biodegradable polymers based nano-formulations for drug delivery: A review," *International journal of pharmaceutics*, vol. 561, pp. 244-264, 2019.
- [33] R. E. Neale, P. W. Barnes, T. M. Robson, P. J. Neale, C. E. Williamson, R. G. Zepp, et al., "Environmental effects of stratospheric ozone depletion, UV radiation, and interactions with climate change: UNEP Environmental Effects Assessment Panel, Update 2020," *Photochemical & Photobiological Sciences*, vol. 20, pp. 1-67, 2021.
- [34] N. Yasri, J. Hu, M. G. Kibria, and E. P. Roberts, "Electrocoagulation separation processes," in *Multidisciplinary Advances in Efficient Separation Processes*, ed: ACS Publications, 2020, pp. 167-203.
- [35] A. Aitbara, A. Khelalfa, M. Bendaia, R. Abrane, A. Amrane, and S. Hazourli, "Treatment of dairy wastewater by electrocoagulation using A-U4G (2017-AI) alloy and pure aluminum as electrode material," *Euro-Mediterranean Journal for Environmental Integration*, vol. 6, pp. 1-10, 2021.
- [36] T. K. Dey, M. E. Uddin, and M. Jamal, "Detection and removal of microplastics in wastewater: evolution and impact," *Environmental Science and Pollution Research*, vol. 28, pp. 16925-16947, 2021.
- [37] B. M. B. Ensano, L. Borea, V. Naddeo, V. Belgiorno, M. D. G. De Luna, and F. C. Ballesteros Jr, "Removal of pharmaceuticals from wastewater by intermittent electrocoagulation," *Water*, vol. 9, p. 85, 2017.
- [38] E. Nariyan, A. Aghababaei, and M. Sillanpää, "Removal of pharmaceutical from water with an electrocoagulation process; effect of various parameters and studies of isotherm and kinetic," *Separation and Purification Technology*, vol. 188, pp. 266-281, 2017.
- [39] M. Yoosefian, S. Ahmadzadeh, M. Aghasi, and M. Dolatabadi, "Optimization of electrocoagulation process for efficient removal of ciprofloxacin antibiotic using iron electrode; kinetic and isotherm studies of adsorption," *Journal of Molecular Liquids*, vol. 225, pp. 544-553, 2017.
- [40] R. Shokri, R. Jalilzadeh Yengejeh, A. A. Babaei, E. Derikvand, and A. Almasi, "Removal of azithromycin from wastewater using advanced oxidation processes (UV/H₂O₂) and moving-bed biofilm reactor (MBBR) by the response surface

- methodology (RSM)," *Journal of Advances in Environmental Health Research*, vol. 7, pp. 249-259, 2019.
- [41] F. Gashtasbi, R. J. Yengejeh, and A. A. Babaei, "Photocatalysis assisted by activated-carbon-impregnated magnetite composite for removal of cephalixin from aqueous solution," *Korean journal of chemical engineering*, vol. 35, pp. 1726-1734, 2018.
- [42] F. Gashtasbi, R. J. Yengejeh, and A. A. Babaei, "Adsorption of vancomycin antibiotic from aqueous solution using an activated carbon impregnated magnetite composite," *Desalination and water treatment*, vol. 88, pp. 286-297, 2017.
- [43] K. Ikehata, N. Jodeiri Naghashkar, and M. Gamal El-Din, "Degradation of aqueous pharmaceuticals by ozonation and advanced oxidation processes: a review," *Ozone: Science and Engineering*, vol. 28, pp. 353-414, 2006.
- [44] W. Aloulou, H. Aloulou, A. Jadda, S. Chakraborty, and R. B. Amar, "Characterization of an asymmetric ultrafiltration membrane prepared from TiO₂-smectite nanocomposites doped with commercial TiO₂ and its application to the treatment of textile wastewater," *Euro-Mediterranean Journal for Environmental Integration*, vol. 5, pp. 1-9, 2020.
- [45] J. Borràs-Ferrís, R. Sánchez-Tovar, E. Blasco-Tamarit, M. J. Muñoz-Portero, R. M. Fernández-Domene, and J. García-Antón, "TiO₂ nanostructures for photoelectrocatalytic degradation of acetaminophen," *Nanomaterials*, vol. 9, p. 583, 2019.
- [46] A. Gómez-Avilés, M. Peñas-Garzón, J. Bedia, D. Dionysiou, J. Rodríguez, and C. Belver, "Mixed Ti-Zr metal-organic-frameworks for the photodegradation of acetaminophen under solar irradiation," *Applied Catalysis B: Environmental*, vol. 253, pp. 253-262, 2019.
- [47] N. Jallouli, L. M. Pastrana-Martínez, A. R. Ribeiro, N. F. Moreira, J. L. Faria, O. Hentati, et al., "Heterogeneous photocatalytic degradation of ibuprofen in ultrapure water, municipal and pharmaceutical industry wastewaters using a TiO₂/UV-LED system," *Chemical Engineering Journal*, vol. 334, pp. 976-984, 2018.
- [48] Y. He, N. B. Sutton, H. H. Rijnaarts, and A. A. Langenhoff, "Degradation of pharmaceuticals in wastewater using immobilized TiO₂ photocatalysis under simulated solar irradiation," *Applied Catalysis B: Environmental*, vol. 182, pp. 132-141, 2016.
- [49] A. Silva, P. Martins, S. Teixeira, S. Carabineiro, K. Kuehn, G. Cuniberti, et al., "Ciprofloxacin wastewater treated by UVA photocatalysis: Contribution of irradiated TiO₂ and ZnO nanoparticles on the final toxicity as assessed by *Vibrio fischeri*," *RSC advances*, vol. 6, pp. 95494-95503, 2016.
- [50] A. Monteoliva-García, J. Martín-Pascual, M. Muñío, and J. Poyatos, "Removal of carbamazepine, ciprofloxacin and ibuprofen in real urban wastewater by using light-driven advanced oxidation processes," *International journal of environmental science and technology*, vol. 16, pp. 6005-6018, 2019.
- [51] I. R. Bautitz and R. F. P. Nogueira, "Degradation of tetracycline by photo-Fenton process—Solar irradiation and matrix effects," *Journal of Photochemistry and Photobiology A: Chemistry*, vol. 187, pp. 33-39, 2007.
- [52] A. G. Trovó, S. A. S. Melo, and R. F. P. Nogueira, "Photodegradation of the pharmaceuticals amoxicillin, bezafibrate and paracetamol by the photo-Fenton process—application to sewage treatment plant effluent," *Journal of Photochemistry and Photobiology A: Chemistry*, vol. 198, pp. 215-220, 2008.
- [53] M. L. Tran, C.-C. Fu, and R.-S. Juang, "Removal of metronidazole and amoxicillin mixtures by UV/TiO₂ photocatalysis: an insight into degradation pathways and performance improvement," *Environmental Science and Pollution Research*, vol. 26, pp. 11846-11855, 2019.
- [54] E. S. Elmolla and M. Chaudhuri, "Degradation of amoxicillin, ampicillin and cloxacillin antibiotics in aqueous solution by the UV/ZnO photocatalytic process," *Journal of hazardous materials*, vol. 173, pp. 445-449, 2010.
- [55] Q. Chen, L. Chen, J. Qi, Y. Tong, Y. Lv, C. Xu, et al., "Photocatalytic degradation of amoxicillin by carbon quantum dots modified K₂Ti₆O₁₃ nanotubes: Effect of light wavelength," *Chinese Chemical Letters*, vol. 30, pp. 1214-1218, 2019.
- [56] P. Calza, V. Sakkas, C. Medana, C. Baiocchi, A. Dimou, E. Pelizzetti, et al., "Photocatalytic degradation study of diclofenac over aqueous TiO₂ suspensions," *Applied Catalysis B: Environmental*, vol. 67, pp. 197-205, 2006.
- [57] M. Valari, A. Antoniadis, D. Mantzavinos, and I. Poullos, "Photocatalytic reduction of Cr (VI) over titania suspensions," *Catalysis Today*, vol. 252, pp. 190-194, 2015.

- [58] R. F. Dantas, M. Canterino, R. Marotta, C. Sans, S. Esplugas, and R. Andreozzi, "Bezafibrate removal by means of ozonation: Primary intermediates, kinetics, and toxicity assessment," *Water Research*, vol. 41, pp. 2525-2532, 2007.
- [59] Y. Zhao, J. Kuang, S. Zhang, X. Li, B. Wang, J. Huang, et al., "Ozonation of indomethacin: kinetics, mechanisms and toxicity," *Journal of hazardous materials*, vol. 323, pp. 460-470, 2017.
- [60] M. N. Rashed, "Adsorption technique for the removal of organic pollutants from water and wastewater," *Organic pollutants-monitoring, risk and treatment*, vol. 7, pp. 167-194, 2013.
- [61] O. Odubiyi, A. Awoyale, and A. Eloka-Eboka, "Wastewater treatment with activated charcoal produced from cocoa pod husk," *Int. J. Environ. Bioener*, vol. 4, pp. 162-175, 2012.
- [62] Y. Önal, C. Akmil-Başar, and Ç. Sarıcı-Özdemir, "Elucidation of the naproxen sodium adsorption onto activated carbon prepared from waste apricot: kinetic, equilibrium and thermodynamic characterization," *Journal of Hazardous Materials*, vol. 148, pp. 727-734, 2007.
- [63] S. Mondal, S. Patel, and S. K. Majumder, "Bio-extract assisted in-situ green synthesis of Ag-RGO nanocomposite film for enhanced naproxen removal," *Korean Journal of Chemical Engineering*, vol. 37, pp. 274-289, 2020.
- [64] D. Z. Husein, R. Hassanien, and M. F. Al-Hakkani, "Green-synthesized copper nano-adsorbent for the removal of pharmaceutical pollutants from real wastewater samples," *Heliyon*, vol. 5, 2019.
- [65] H. Guedidi, L. Reinert, Y. Soneda, N. Bellakhal, and L. Duclaux, "Adsorption of ibuprofen from aqueous solution on chemically surface-modified activated carbon cloths," *Arabian Journal of Chemistry*, vol. 10, pp. S3584-S3594, 2017.
- [66] K. Jafari, M. Heidari, and O. Rahmanian, "Wastewater treatment for Amoxicillin removal using magnetic adsorbent synthesized by ultrasound process," *Ultrasonics Sonochemistry*, vol. 45, pp. 248-256, 2018.
- [67] T. Thiebault, M. Boussafir, L. Fougère, E. Destandau, L. Monnin, and C. Le Milbeau, "Clay minerals for the removal of pharmaceuticals: Initial investigations of their adsorption properties in real wastewater effluents," *Environmental nanotechnology, monitoring & management*, vol. 12, p. 100266, 2019.
- [68] K. Styszko, K. Nosek, M. Motak, and K. Bester, "Preliminary selection of clay minerals for the removal of pharmaceuticals, bisphenol A and triclosan in acidic and neutral aqueous solutions," *Comptes Rendus. Chimie*, vol. 18, pp. 1134-1142, 2015.
- [69] C. Peiris, S. R. Gunatilake, T. E. Mlsna, D. Mohan, and M. Vithanage, "Biochar based removal of antibiotic sulfonamides and tetracyclines in aquatic environments: a critical review," *Bioresource Technology*, vol. 246, pp. 150-159, 2017.
- [70] J. Wang and S. Wang, "Preparation, modification and environmental application of biochar: A review," *Journal of Cleaner Production*, vol. 227, pp. 1002-1022, 2019.
- [71] N. Hilal, H. Al-Zoubi, N. Darwish, A. Mohamma, and M. A. Arabi, "A comprehensive review of nanofiltration membranes: Treatment, pretreatment, modelling, and atomic force microscopy," *Desalination*, vol. 170, pp. 281-308, 2004.
- [72] R. Rienzie, S. Ramanayaka, and N. M. Adassooriya, "Nanotechnology applications for the removal of environmental contaminants from pharmaceuticals and personal care products," in *Pharmaceuticals and personal care products: waste management and treatment technology*, ed: Elsevier, 2019, pp. 279-296.
- [73] A. Cincinelli, T. Martellini, E. Coppini, D. Fibbi, and A. Katsoyiannis, "Nanotechnologies for removal of pharmaceuticals and personal care products from water and wastewater. A review," *Journal of nanoscience and nanotechnology*, vol. 15, pp. 3333-3347, 2015.
- [74] S. Irvani, "Nanomaterials and nanotechnology for water treatment: recent advances," *Inorganic and Nano-Metal Chemistry*, vol. 51, pp. 1615-1645, 2021.
- [75] J.-F. Li, Z.-L. Xu, H. Yang, L.-Y. Yu, and M. Liu, "Effect of TiO₂ nanoparticles on the surface morphology and performance of microporous PES membrane," *Applied Surface Science*, vol. 255, pp. 4725-4732, 2009.
- [76] W. Xiong, Z. Zeng, X. Li, G. Zeng, R. Xiao, Z. Yang, et al., "Multi-walled carbon nanotube/amino-functionalized MIL-53 (Fe) composites: remarkable adsorptive removal of antibiotics from aqueous solutions," *Chemosphere*, vol. 210, pp. 1061-1069, 2018.
- [77] C.-Y. Chang, J.-S. Chang, S. Vigneswaran, and J. Kandasamy, "Pharmaceutical wastewater treatment by membrane bioreactor process—a case study in southern Taiwan," *Desalination*, vol. 234, pp. 393-401, 2008.

- [78] J. Ji, A. Kakade, Z. Yu, A. Khan, P. Liu, and X. Li, "Anaerobic membrane bioreactors for treatment of emerging contaminants: A review," *Journal of Environmental Management*, vol. 270, p. 110913, 2020.
- [79] M. Prado, L. Borea, A. Cesaro, H. Liu, V. Naddeo, V. Belgiorno, et al., "Removal of emerging contaminant and fouling control in membrane bioreactors by combined ozonation and sonolysis," *International Biodeterioration & Biodegradation*, vol. 119, pp. 577-586, 2017.
- [80] R. M. Sterritt, M. J. Brown, and J. N. Lester, "Metal removal by adsorption and precipitation in the activated sludge process," *Environmental Pollution Series A, Ecological and Biological*, vol. 24, pp. 313-323, 1981.
- [81] D. Mousel, D. Bastian, J. Firk, L. Palmowski, and J. Pinnekamp, "Removal of pharmaceuticals from wastewater of health care facilities," *Science of the Total Environment*, vol. 751, p. 141310, 2021.
- [82] M. Clara, B. Strenn, O. Gans, E. Martinez, N. Kreuzinger, and H. Kroiss, "Removal of selected pharmaceuticals, fragrances and endocrine disrupting compounds in a membrane bioreactor and conventional wastewater treatment plants," *Water research*, vol. 39, pp. 4797-4807, 2005.
- [83] M. Kim, P. Guerra, A. Shah, M. Parsa, M. Alaei, and S. Smyth, "Removal of pharmaceuticals and personal care products in a membrane bioreactor wastewater treatment plant," *Water science and technology*, vol. 69, pp. 2221-2229, 2014.
- [84] J. Wang and C. Chen, "Biosorbents for heavy metals removal and their future," *Biotechnology advances*, vol. 27, pp. 195-226, 2009.
- [85] D. Friedmann, C. Mendive, and D. Bahnemann, "TiO₂ for water treatment: parameters affecting the kinetics and mechanisms of photocatalysis," *Applied Catalysis B: Environmental*, vol. 99, pp. 398-406, 2010.