Chapter

An Overview of Occurrence and Removal of Pharmaceuticals from Sewage/Wastewater

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Abstract

Nowadays, the occurrence of pharmaceuticals in sewage/wastewater is a major environmental concern. Their precise characterization and suitable treatment/disposal is a must else it pollutes the surface water bodies and causes major distress on aquatic lives and human health. Also, the up-gradation of the sewage/wastewater treatment plant (WWTP) is a must to consider the removal of these pollutants and to provide the best quality effluent for various reuse purposes. Mostly, the conventional treatment methods are inefficient for their removal, and hence, the most advanced and refined treatment options are needed for their effective treatment. In this chapter, we have highlighted the occurrence of pharmaceuticals in various water samples and their treatment options are reviewed. It was recommended that integrated treatment systems are more efficient, economical, and environmental friendly than single stand-alone treatment. Further advancement and modifications in the treatment options are required to overcome the shortcomings regarding pharmaceutical removal to achieve the legal standard discharge limit.

Keywords: advanced oxidation process, biological treatment, emerging contaminants, wastewater, recalcitrant compounds, sewage

1. Introduction

Nowadays, the problems associated with the widespread occurrence of pharmaceuticals in the aquatic environment have been recognized as an emerging environmental issue [1–3]. The increasing usage of pharmaceuticals and their improper discharge is one of the major environmental concerns. Pharmaceuticals are a large and diverse group of compounds designed to prevent, cure, and treat disease and improve health. Their usage and consumption are increasing consistently due to the discoveries of new drugs, the expanding population, etc. [2, 3]. After intake, these pharmaceutically active compounds undergo metabolic processes in the organism. Significant fractions of the parent compound are excreted in un-metabolized form into raw sewage and wastewater treatment systems. The most commonly occurring pharmaceuticals in the environment are given in **Table 1** [4]. Thus, body metabolization and excretion followed by wastewater treatment are considered to be the primary pathway of pharmaceuticals to the environment [1–3, 5–7]. Disposal of drug leftovers into sewage and trash is another source of entry [8]. In addition, sewer leaking [9], sewer overflow [10], and surface runoff [11] are also considered

S. no.	Class of drugs	Name of drugs
1.	Antibiotics	Erythromycin, ofloxacin, streptomycin, flumequine, ciprofloxacin, trimethoprim, sulfamethoxazole, lincomycin, penicillin, and amoxicillin
2.	Antidepressants	Mianserin
3.	Anticancer drugs	Cyclophosphamide and ifosphamide
4.	Anti-inflammatory drugs	Acetylsalicylic acid (aspirin), diclofenac, ibuprofen, acetaminophen, naproxen, and phenazone
5.	Beta-blockers	Metoprolol, propranolol, nadolol, and atenolol
6.	Diuretics	Furosemide
7.	Lipid regulators	Bezafibrate, gemfibrozil, clofibric acid, and fenofibrate
8.	Steroids and related hormones	17-β-estradiol, estrone, and diethylstilbestrol
9.	Tranquilizers	Diazepam

Table 1.
Some common pharmaceuticals are found in the environment [4].

as additional sources contributing to the presence of pharmaceuticals in the aquatic environment [5].

Their detection techniques and proper characterization are relatively difficult which required distinctive procedures and sophisticated instruments due to their low concentration levels in different environmental matrices [7, 11, 12]. Several studies investigated the occurrence and distribution of pharmaceuticals in soil irrigated with reclaimed water [13, 14] and soil that received biosolids from urban sewage treatment plants [15, 16]. These studies confirmed that the conventional systems are not enough to completely remove such micro-pollutants from wastewater and sludge, and as a result, they find their way into the environment [17]. Once entered the environment, pharmaceutically active compounds can produce subtle effects on aquatic and terrestrial organisms. Therefore, the occurrence of pharmaceutical compounds and the extent to which they can be eliminated during wastewater treatment have become the active subject matter of actual research [1, 3–7].

Domestic sewage is relatively simple to treat with conventional methods due to the absence of any recalcitrant compounds. The conventional treatment options are widely applicable for their effective treatment [1, 18–20]. The sewage/wastewater treatment plants are generally not designed to consider the specific pharmaceuticals, emerging compounds, etc., during the treatment. Hence, their presence in the sewage water is very problematic for the treatment performance of the plant [1, 5–7, 21]. Furthermore, the presence of pharmaceuticals in the effluents of sewage/wastewater treatment plants is very toxic in many ways to the soil and surrounding water bodies [1–5, 21]. To overcome the abovementioned problems, firstly, we have to stop the improper disposal of pharmaceuticals and their proper monitoring/collection system should be designed [3]. The accurate characterization and suitable treatment options should be provided to obtain the legal effluent discharge standards. The constant discharge of various pharmaceuticals into the water bodies and their persistent nature and bioaccumulation potential cause serious effects to aquatic lives and human health [21–23]. Therefore, in this chapter, we have highlighted the occurrence and some of the removal techniques specifically for the pharmaceuticals from sewage/wastewater. The scope for future research directions is also highlighted in the conclusion part.

2. Occurrence of pharmaceuticals in sewage/wastewater

The huge variation in the concentrations of pharmaceutically active compounds (PhACs) was observed due to various factors viz. environmental persistency, dilution, treatment efficiency [21, 24, 25]. In some studies, the reported amounts of pharmaceuticals are estimated to be 5.6, 2.0, and 0.4 g/day/1000 equivalent inhabitants [1, 21]. In one of the studies, the highest levels at the influent of WWTPs were observed for nonsteroidal anti-inflammatory drugs (NSAIDs) that were expected due to their high consumption [1]. Lower but still significant levels of lipid-modifying agents (7–12%), diuretics (8–10%), and beta-blockers (5–9%) were detected entering the WWTPs [1]. Atenolol and carbamazepine were quantified in the influent samples of WWTPs in average concentrations ranging from 0.4 to 1.4 mg/L [1]. The amount found in effluent or sludge depended on the removal efficiency of the plant and/or the physicochemical properties of the compounds. In the effluent waters, NSAIDs were present in the highest percentage (35–44%), followed by the lipid-modifying agents (8–29%) and psychiatric drugs (17–30%) [1]. The highest concentrations in the effluents were found for naproxen, diclofenac, and carbamazepine [1].

It has been reported that from the list of detected samples of the emerging contaminants about 70% are PACs and personal care products (PCPs) [26]. Globally, more than 200 PhACs have been reported in river waters with a maximum concentration of 6 mg/L for ciprofloxacin antibiotics [27]. Similarly, tamoxifen was detected in the range of 25–38 ng/L [28]. Also, the concentrations of antibiotics, hormones, antidepressants, and chemotherapy drugs range from 0.04 to 6.3 μg/L [29]. Out of the various sources of the PhACs into the environment, the domestic discharge and effluents of the manufacturing units of pharmaceuticals are well-thought-out major sources [22]. Various categories for the occurrence of the PhACs have been reported viz. wastewater treatment plants (WWTPs), wastewater,

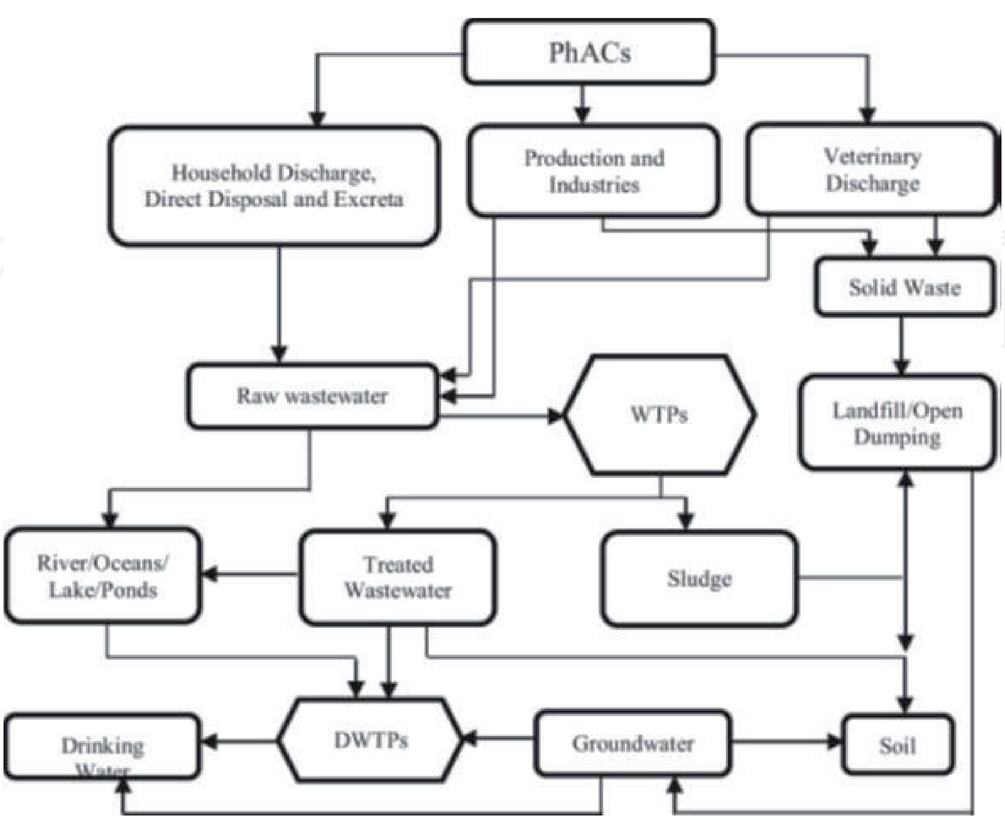

Figure 1.
Flowchart showing PhACs pathways in the environment [30, 31].

sewage, sewage sludge, groundwater, surface water, and drinking water [24, 30, 31]. So, WWTPs are considered as one of the prominent anthropogenic sources emitting pharmaceuticals into the environment along with industrial discharges, hospital effluents, etc. [22, 25]. Furthermore, the inefficient management and treatments of PhACs risk the prospect of sustainable reuse of treated wastewater and sludge [21]. **Figure 1** shows the flowchart showing PhACs pathways in the environment [30, 31].

3. Removal of pharmaceuticals from sewage/wastewater

It was well recognized in the literature that the conventional biological treatment systems alone are not sufficient enough to completely remove the pharmaceuticals and therefore, some additional steps are required for their proper treatment. It was reported that among the conventional activated sludge process (ASP) and membrane bioreactor (MBR) systems, the MBR system appeared to have a higher removal efficiency for many of the pharmaceuticals [1, 32]. On the other hand, carbamazepine and hydrochlorothiazide showed poor removal efficiencies in either ASP or MBR systems [1, 32]. O'Brien et al. [33] have found that compounds such as atenolol, carbamazepine, and ibuprofen appeared to be persistent in the sewer system. It was recommended to use a composite sampling approach in wastewater treatment plants [5, 33]. The superior performance of MBR in the removal of some target pharmaceuticals is due to the result of the higher biomass concentration, longer solid retention time (SRT), and better-retaining capacity of solids and microbes [32, 34]. On the other hand, the integration approach of membrane technology is called electrochemical membrane bioreactors (EMBR). It was observed that EMBRs are more efficient with low energy consumptions as compared to MBRs and ASPs [21, 35]. But the common problem with the advanced technologies is their limited applications only at laboratory and pilot scales. Besides, membrane fouling, high energy demand, and costly membrane materials are some limitations of MBRs, which need to be overcome for their extensive full-scale applications [21].

The term removal of pharmaceuticals used here means the conversion of the parent compound. Thus, the overall removal refers to the losses of a parent compound by different mechanisms of chemical and physical transformation, biodegradation, and sorption to solid matter [1]. The most analyzed carbamazepine showed very low removal (<25%) regardless of the treatment applied [32]. The pharmaceuticals removal efficiencies are based on the characteristics of the wastewater, treatment types used, and other operational conditions [1, 21]. The addition of the occasional tertiary treatment improves the removal efficiencies of the pharmaceuticals. The lower removal efficiency of diclofenac was reported in some studies [1, 36, 37]. Better performances of WWTP may be due to longer both hydraulic and solid retention times. As a compound spends more time in reactors wherein bacteria growth is promoted, the biological transformation may occur to a greater extent [38]. It has been proven that longer SRT, especially, improves the elimination of most of the pharmaceuticals during sewage treatment [1, 39].

A variety of treatment techniques for pharmaceuticals removal have been considered in the past studies such as natural, conventional and advanced treatment approaches. Dilution, volatilization, photolysis, sorption, biodegradation, etc., are cost-effective and natural processes [21]. However, the natural processes are proved less efficient [22]. On the other hand, the conventional approaches viz. adsorption, ozonation, membrane filtration, showed high pharmaceuticals removal efficacies [23]. But these approaches are having some disadvantages like oxidation by-products formation in the ozonation process may be more toxic than the parent compounds, and high operational costs in addition to the concentrate disposal are required in the membrane filtration process [25]. The widespread applications of various advanced treatment approaches viz. advanced oxidation processes (AOPs), constructed wetlands, bioelectrical systems, enzymatic treatment, have been recommended in the past few years [21]. Also, the up-gradation of the conventional WWTPs might further minimize the environmental release of the various pharmaceuticals [21, 23, 40]. Although the AOPs are considered one of the most effective treatment options for a variety of pharmaceuticals removal, their full-scale applications are still limited due to the number of challenges [18–21, 25, 41].

The WWTPs generally considered the primary, secondary, and sometimes tertiary treatment stages. The pharmaceuticals entered into the plants undergo several treatment stages, and their fraction is degraded/removed [21, 24, 42]. In the secondary stage, the pharmaceuticals are subjected to several processes such as biodegradation, sorption, dispersion, dilution, photodegradation, and volatilization [21, 22, 24]. Likewise, the tertiary treatment steps are reported to exhibit significant pharmaceuticals removal efficiency via ozonation-like conventional oxidation processes [21, 43, 44].

The importance of the tertiary treatment in the WWTPs is versatile as it supplements the secondary treatment and those pollutants that are not removed in the second stage are removed in the tertiary stage. Several advanced technologies are employed to remove the pharmaceuticals in the WWTPs themselves to produce high-quality effluent for reuse purposes [21, 44, 45]. Among the tertiary treatment, AOPs have been considered that oxidize/mineralize the various pharmaceuticals and their by-products to CO_2, H_2O, and simple inorganic ions [18, 21]. The various types are AOPs are now widely applied for various applications of high strength and pharmaceuticals removal viz. Fenton process, Photo-Fenton process, Electro-Fenton process, Sono-Fenton process, ozonation process, UV-based treatment [21, 46]. Also, a range of commercially available adsorbents, such as activated carbon (AC), biochar, carbon nanotubes, clay minerals, are used for the adsorption of various pharmaceuticals [21, 47]. The usages of AC for a broad-spectrum pharmaceutical adsorption were found most suitable due to reduced interference from the organic materials for the adsorption active sites [21, 48]. The adsorption efficiency depends on the types of PhACs, properties of AC, and other environmental conditions [21, 24].

Among the mentioned options, ozonation and AC treatment are found to be the economically feasible option and utilized in some WWTPs [21, 25]. The main reactive species in AOPs for the degradation/mineralization of the pharmaceuticals are hydroxyl radicals ($OH^{\bullet}$) and the number of parallel reactions is reported in their mechanism [18–20, 49, 50]. The suitability of the various adoption of the AOPs is mainly based on wastewater characteristics, recalcitrant nature of the target compounds, available resources, and economic conditions [50]. It was well recognized in the literature that the integrated processes are more efficient and environmental friendly [18, 50]. A very high removal efficiency (>95%) of diclofenac, carbamazepine, sulpiride, at an ozone dose of 5 mg/L, was observed [51]. All the AOPs are having their limitations/disadvantages as well; hence, the suitable/optimized treatment options should be designed and implemented to achieve the target removal efficacies etc. [18, 41, 50]. Some of the disadvantages of the Fenton process are low-working pH requirement and high sludge production, the chances of the pharmaceuticals accumulate in the iron sludge produced after the treatment [41]. On the other hand, when the applied ozone dosages are inadequate, it will result in the formation of transformation products [18], and the toxicity can further be reduced by a subsequent biological treatment [21, 52]. The combined approach of the ozonation-biological process is found most efficient for the removal of pharmaceuticals from secondary urban wastewater [21, 52, 53]. Currently, many treatment technologies are available as mentioned in **Table 2** [54, 55].

Treatment technologies	Classification
Physical treatment	Primary treatment
Aerobic process Anaerobic process	Secondary biological treatment
Activated carbon Membrane distillation Membrane technology	Tertiary treatments
Fenton process Ozone/hydrogen peroxide treatment Photocatalysis Electrochemical oxidation Ultrasound irradiation Wet air oxidation	Advanced oxidation processes
Mixed primary, secondary and tertiary treatments	Hybrid technologies

Table 2.
Some treatment technologies for pharmaceutical wastewater treatment [54, 55].

4. Conclusion

This chapter provides a brief overview regarding the problems associated with the pharmaceuticals present in the sewage/wastewater and their suitable treatment options. From the literature, it was understood that the problems related to the emerging contaminants and particularly for the pharmaceuticals are of great concern and require specific attention to protecting the environment and public health. Out of the various categories of pharmaceuticals, different treatment options are required and one single option is not sufficient to remove all the types of pharmaceuticals. The challenges associated with their accurate analysis, detection, and extraction due to their low concentration are also an important domain for further research. Regarding the treatment options in various studies, it was reported that the integrated processes are more advantageous in many ways for pharmaceuticals removal. For example, the post-biological treatment option after the ozonation process significantly improves the pharmaceutical removal. The other options like ASP and MBR are also considered useful but not efficient enough for their complete mineralization and removal. Also, the activated carbon-adsorption process is just a phase change mechanism system and required extensive research for further improvement. Various transformation/intermediate products are formed in AOPs treatment, hence required more advancements to remove those toxic intermediates from the water matrix. The up-gradation of the WWTPs is a very important step to improve the effluent quality considering the problems of the pharmaceuticals. The single and combined AOPs are limited to lab/pilot scale only and their full-scale applications are required, which should be focused on in future research for the best-fit alternative both economically and environmental friendly.

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Conflict of interest

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