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Drug Contamination in the Environment: A Review of Sources, Occurrence and Remediation Technologies

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Abstract. The existing treatment technologies for new types of environmental pollutant, such as Pharmaceuticals and personal care products (PPCPs), still have significant deficiencies in terms of removal efficiency, operating costs, and the coordinated control of resistance genes. This article systematically reviews the main pollution sources of PPCPs, typical drug categories, and their migration and transformation behaviors in multi-media environments, with a focus on comparing the removal efficiency and applicable conditions of conventional biochemical treatment, physicochemical separation, and advanced oxidation technologies. The results show that the removal rate of carbamazepine and other refractory drugs by the conventional process is less than 40%. Although the advanced oxidation technology can achieve a removal rate of over 90%, it is restricted by engineering bottlenecks, such as the high cost and toxicity of by-products. The synergistic effect of the mixed technology is regarded as an important direction to balance efficacy and economy. Furthermore, the dependence of photolysis rate on the water matrix and the inhibitory effect of the low-temperature environment on microbial activity highlight the limitations of extrapolating laboratory parameters to real scenarios. The synergistic removal of antibiotic resistance genes poses higher requirements for the treatment process than conventional pollutant removal. This paper looks forward to future directions such as multi-technology coupling and climate change adaptation strategies, with the aim of providing a reference basis for the selection of water pollution control technologies and the formulation of environmental management policies.

Keywords: Pharmaceuticals and personal care products (PPCPs), Emerging contaminants, Occurrence and fate, Advanced oxidation processes (AOPs), Antibiotic resistance genes (ARGs)

1. Introduction

Pharmaceuticals and personal care products (PPCPs), as a new type of environmental pollutant, include antibiotics, non-steroidal anti-inflammatory drugs, hormone substances and personal care products, etc. They are continuously introduced into the water environment through multiple channels such as domestic sewage, hospital wastewater, pharmaceutical industry emissions and aquaculture runoff [1]. Studies on agricultural drainage in the Yellow River Basin have shown that PPCPs are widely present in surface water bodies, with the detection frequency of some antibiotics

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exceeding 80%. Their spatial distribution is closely related to the type of discharge source [2]. However, traditional pollution perceptions are mostly based on the steady-state discharge assumption, which holds that PPCPs mainly enter the environment through continuous discharge from sewage treatment plants. This paradigm is no longer capable of comprehensively depicting the actual pollution characteristics. Combined sewer overflow (CSO) incidents triggered by heavy rain can release untreated wastewater in a short period of time, forming high-intensity intermittent pollution pulses, with instantaneous concentrations often several times that of conventional discharges. Such dynamic processes are often overlooked in existing risk assessments [3]. At the technical level, the removal efficiency of PPCPs by traditional sewage treatment processes varies greatly, with the removal rate of refractory drugs such as carbamazepine being less than 40% [2]. Although advanced oxidation technology (AOPs) can achieve a removal rate of over 90%, its high operating costs and the potential toxicity of disinfection by-products have restricted its engineering promotion [1]. In addition, the problem of the co-transmission of antibiotic residues and resistance genes (ARGs) in the environment has not been effectively curbed [3, 4]. Take South Africa as an example. The superposition of agricultural drainage and pharmaceutical industry emissions has led to the widespread detection of various PPCPs residues in surface water bodies. Moreover, the fragmentation of relevant management policies and the insufficiency of law enforcement capabilities have further exacerbated the difficulty of pollution prevention and control [5]. Based on the above research gaps, this paper intends to systematically review the source apportioning, occurrence characteristics and main treatment technologies of drug pollutants. By sorting out the contributions of different pollution sources and comparing the efficiency of various treatment technologies, it will analyze the key challenges currently faced and make a forward-looking outlook on the future governance direction. With the aim of providing a reference basis for the selection of water pollution control technologies and the formulation of environmental management policies.

2. The sources and destinations of pharmaceutical pollutants in the environment

2.1. Major pollution source apportionment

The input pathways of pharmaceutical pollutants in the environment are characterized by diversification and complexity, and there are significant differences in the contribution ratios and pollutant compositions of different pollution sources. Overall, pharmaceutical pollutants mainly enter the environment through four major channels: municipal sewage, hospital wastewater, industrial wastewater and aquaculture wastewater. Municipal sewage is the main channel for the collection and discharge of pharmaceutical pollutants. Domestic sewage contains a large amount of drug residues that have been metabolized by the human body or directly discarded, including antibiotics, anti-inflammatory and pain-relieving drugs, hormones and various personal care products. The effluent from urban sewage treatment plants is the main source of PPCPs in surface water bodies. However, the removal rate of most PPCPs by conventional treatment processes is less than 40%, and the removal rate of refractory substances such as carbamazepine is even lower than 20% [1]. Even treated municipal effluent remains a continuous input source for PPCPs entering the environment. Although the hydraulic flow of hospital wastewater is far less than that of municipal sewage, the concentration of pharmaceutical pollutants per unit volume is often several orders of magnitude higher. Hospital wastewater contains a large amount of high-concentration antibiotics, anti-tumor drugs, contrast agents and disinfectants, etc., which have a strong inhibitory effect on conventional biological treatment processes, making hospital wastewater an important point source of highly active drug residues in the environment. The pollution contribution of industrial

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wastewater mainly comes from the production drainage of pharmaceutical enterprises. This type of wastewater may contain high concentrations of active pharmaceutical ingredients, intermediates and by-products. The water quality fluctuates greatly and has strong biological toxicity. If it is discharged into the municipal water network or directly discharged without pretreatment, it will cause serious local pollution to the receiving water body. Chemical analysis of 132 wastewater samples from 20 industrial sectors by Keane et al. [6] indicated that wastewater from different industries had characteristic PPCPs fingerprint patterns. Among them, the concentration of acetate in the beverage manufacturing industry was significantly increased, and the concentrations of multiple new pollutants in landfill and waste facilities reached extremely high levels. The random forest model based on fingerprint recognition can effectively identify approximately 50% of pollution sources [6]. This indicates that the input of PPCPs from industrial sources shows significant industry dependence, and source identification and classification control are the key links in the treatment of industrial wastewater. Aquaculture wastewater is an important source of antibiotic pollutants. In aquaculture and livestock breeding, antibiotics are widely used for disease prevention and growth promotion. A large number of drugs enter the environment in their original form or metabolite form along with excreta. Monitoring shows that the concentration of some antibiotics in the effluent of aquaculture wastewater can be thousands of times higher than that in municipal sewage, and the discharge has regional aggregation, posing a continuous threat to the surrounding surface water and groundwater [1].

2.2. The main pharmaceutical pollutants in the environment

Based on the existing monitoring data, the drug pollutants with high detection frequency and significant concentration levels in the environment can be classified into the following categories. Antibiotics are the most diverse and highly concerned category detected in the environment, covering sulfonamides, quinolones, tetracyclines and macrolides, etc. Typical substances include sulfamethoxazole, norfloxacin, ciprofloxacin, tetracyclines and amoxicillin, etc. Monitoring data on agricultural drainage in the Yellow River Basin shows that the detection frequency of some antibiotics in surface water exceeds 80%, and their concentrations in aquaculture wastewater and hospital wastewater are particularly prominent [2]. The core risk of antibiotics lies in their selective pressure on ARGs. Even at extremely low concentrations, their long-term presence may accelerate the spread of drug-resistant bacterial colonies. Anti-inflammatory and pain-relieving drugs are another type of widely present drug contaminants. Ibuprofen, diclofenac, naproxen and acetaminophen have been detected in various water bodies around the world. Among them, the photodegradation half-life of acetaminophen in estuarine water can be as short as 1.2 hours, but its continuous input makes it exhibit a "pseudo-persistence" characteristic [1]. Diclofenac has been placed on the priority watch list by the European Union due to its chronic toxic effects on aquatic organisms. Hormone substances mainly include natural estrogens and synthetic hormones. At extremely low concentrations (ng/L level), they can interfere with the reproductive endocrine system of fish, leading to feminization and changes in population structure. Estrogen-like substances are detected at a relatively high frequency in the effluent of sewage treatment plants, and the removal capacity of traditional processes for them is limited [1]. In addition, carbamazepine, caffeine and triclosan are also frequently detected in surface water and groundwater. Carbamazepine, due to its strong biological inertness, is difficult to remove in both conventional treatment and natural decay, and has been used as a chemical tracer for anthropogenic contamination [1, 3].

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2.3. The migration and transformation of pharmaceutical pollutants in the environment

After drug pollutants enter the water environment, they are jointly regulated by multiple processes such as water-sediment distribution, biodegradation and photolysis.

The water-sediment distribution affects the distribution of pollutants between the aqueous phase and the solid phase. Compounds with strong hydrophobicity (such as some antibiotics and triclosan) are prone to adsorb onto suspended particulate matter and surface sediments and be removed from the aqueous phase. However, they may also form long-term reservoirs in the sediments and be re-released when conditions change [1]. The attenuation of tetracycline antibiotics in rivers is positively correlated with the organic matter content and cation exchange capacity of sediments, indicating that adsorption is their main migration pathway. The adsorption capacity of sediments is affected by pH, ionic strength and the concentration of dissolved organic matter. The distribution coefficients of different chemicals can vary by several orders of magnitude.

The biodegradation efficiency has a high degree of compound specificity. The removal rates of ibuprofen and naproxen significantly increase during the summer when microbial activity is vigorous, while the removal rates of structurally stable drugs such as carbamazepine are generally below 20% [1, 3]. Microbial activity is inhibited in low-temperature environments, and drug contaminants in polar and alpine glacial meltwater may have longer environmental persistence [2]. Barbaro et al. [7] confirmed that PPCPs are enriched in ice and snow through atmospheric condensation, and the melting of glaciers driven by climate warming is promoting the re-release of such pollutants, posing a "secondary source" risk. Biodegradation often cannot be completely mineralized, and the toxicity of intermediate metabolites may be higher than that of the parent compounds.

Photolysis is the most important abiotic transformation pathway in surface water bodies. The half-life of acetaminophen in pure water is approximately 29 hours, but it is shortened to 1.2 hours in estuarine water because nitrates and dissolved organic matter in seawater can act as photosensitizers to accelerate indirect photolysis [1]. The half-life of caffeine varies from 5 hours to over 100 hours in different substrates. The half-life of carbamazepine under simulated daylight is 8 to 38 hours, but it can be extended to over 67 hours when it is shaded by vegetation in actual rivers [1].

In actual environments, the matrix effect often makes it difficult to directly extrapolate the degradation parameters derived in the laboratory, which is a limitation that cannot be ignored in current risk assessment.

3. Conventional and advanced treatment technologies for pharmaceutical contaminants

3.1. Conventional biochemical treatment technology

Conventional biochemical treatment is the main process of municipal sewage treatment plants, including primary sedimentation, activated sludge biological treatment and secondary clarification units, etc. Its design intention is to remove organic matter, suspended solids and nutrients, rather than trace organic pollutants such as PPCPs [1], and it has inherent limitations when dealing with trace drug residues.

In the activated sludge system, PPCPs are mainly removed through biological degradation and sludge adsorption. Under suitable conditions, the removal rates of ibuprofen and naproxen can reach over 80%, while those of carbamazepine, diclofenac, etc. are generally below 30% [1]. The removal efficiency of different processes varies significantly: The sequencing batch reactor can achieve a

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removal rate of up to 85% for 17α -acetylene estradiol, but only 15% for sulfamethoxazole. Conventional processes are difficult to achieve broad-spectrum removal [3]. Membrane bioreactors can extend the sludge age through membrane retention, which can improve the removal efficiency of some PPCPs, but their removal effect on hydrophilic and refractory substances is still very limited [1].

3.2. Advanced oxidation technology

Advanced oxidation technology (AOPs) can non-selectively mineralize or transform organic pollutants by generating highly reactive oxygen species such as hydroxyl radicals (·OH), and it represents a cutting-edge direction in the advanced treatment of PPCPs. Common AOPs include ozone oxidation, Fenton/ photo-Fenton, photocatalytic oxidation and electrochemical oxidation, etc. [1]. It is worth noting that in recent years, advanced oxidation processes based on periodate have attracted extensive attention due to their diverse activation methods and high free radical yield. Relevant reviews have systematically summarized the application potential of this technology in the removal of PPCPs [8]. The prominent advantage of AOPs lies in their broad-spectrum reactivity—hydroxyl radicals can react rapidly with the vast majority of organic pollutants. Research data indicate that under optimized conditions, the removal rate of carbamazepine by ozone oxidation can exceed 95%, and the removal rate of multiple antibiotics by the UV/H_2O_2 process can reach over 90% [3]. However, there are significant efficacy differences among different AOPs: photo-Fenton has a better mineralization efficiency for antibiotics than Fenton alone, while electrochemical oxidation has higher selectivity for certain hormone-like substances [1]. In addition, the background matrix in actual water bodies (dissolved organic matter, bicarbonate, etc.) may quench free radicals, thereby inhibiting the treatment efficiency. This is an unavoidable constraint in the engineering application of AOPs.

3.3. The challenges faced by the existing processing technologies

The processing cost is the primary factor restricting the promotion of advanced processing technology. The energy consumption and chemical agent costs of advanced oxidation technology are significantly higher than those of conventional processes [1, 3], and the operating costs can be several times higher than those of the activated sludge process. Membrane filtration is confronted with the problems of frequent component replacement and high cleaning costs [9]. The catalytic membrane developed by Varghese et al. [10] can simultaneously achieve membrane separation and photocatalytic degradation. In areas with weak infrastructure, high costs make it difficult to popularize deep technologies [1].

The toxicity of by-products is a significant hidden danger for AOPs. Oxidation may generate intermediate products that are more toxic than the parent products [1]. The photoconversion products of ketoprofen have higher toxicity. The inhibitory effect of diclofenac conversion products on Vibrio Fischeri persists [3]. It is one-sided to only take the maternal removal rate as the evaluation index and it must be included in the toxicity assessment of the transformation products.

The selective pressure of antibiotics on bacterial communities can promote the spread of ARGs [3]. Traditional biological treatment may instead accelerate its horizontal transfer. Although ozone and ultraviolet light can reduce ARGs, their efficiency is affected by water quality [3]. The residual sub-inhibitory concentration still maintains selective pressure, and it is necessary to pursue near-complete removal.

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The AOPs parameters optimized in the laboratory are difficult to achieve the same effect in real sewage, and the quenching effect of background substances on free radicals cannot be ignored [1]. Most of the existing technologies operate in batches, and there are adaptation issues when coupled with the continuous flow mode [3]. The stability of some materials in large-scale applications remains to be verified [9]. The ARGs co-removal process lacks systematic engineering guidance standards [4].

4. Conclusion

This paper reviews the pollution sources, occurrence characteristics, migration and transformation, and treatment technologies of PPCPs, and draws the following conclusions. First, the input of PPCPs is diverse and continuous. Municipal sewage, hospital wastewater, industrial wastewater and aquaculture wastewater are the four major pollution sources. The removal rate of carbamazepine by the conventional process is less than 40%, and the tail water still constitutes a continuous input. The concentration of antibiotics in aquaculture wastewater can be several orders of magnitude higher than that in municipal sewage, making it an important driver for the spread of resistance genes. The detection frequency of antibiotics in some areas of the Yellow River Basin exceeds 80%. Second, the fate of PPCPs is jointly regulated by water-sediment distribution, biodegradation and photolysis. The photolysis rate is highly dependent on the water matrix. The half-life of acetaminophen in pure water is 29 hours, but it is shortened to 1.2 hours in estuarine water. Biodegradation is a compound specification. The removal rate of ibuprofen can reach over 80%, while that of carbamazepine is less than 20%. Laboratory parameters are difficult to extrapolate directly.

Third, there are significant differences in the efficiency of processing technologies. Conventional processes are inadequate for refractory substances. Although advanced oxidation technology can achieve a removal rate of over 90%, it faces the bottlenecks of cost and toxicity of by-products. Hybrid technology is the development direction. Current evaluations mostly focus on parent body removal, with insufficient assessment of the toxicity of transformation products, which may underestimate ecological risks. Fourth, the coordinated removal of resistance genes is a unique challenge. Traditional biological treatment may accelerate the horizontal transfer of ARGs, and strategies such as combined ultraviolet/chlorine disinfection need to be developed. In terms of limitations, there is a lack of a unified benchmark for performance comparison. The discussion on the low-temperature trend is weak, and glacial meltwater is becoming a "secondary source". The synergistic/antagonistic effects have not been fully explored. In the future, a standardized evaluation system should be established, the long-term operation tracking of AOPs should be strengthened, the toxicological identification of transformation products should be deepened, the ARGs synergistic removal process should be explored, and a resilient system to deal with pulse pollution should be developed.

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