Analysis of the Current Situation and Treatment Technology of PPCPs in Wastewater

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Abstract: Pharmaceutical and personal care product contaminants (PPCPs) are a representative group of emerging contaminants. The improvement of human living standards and the development of medical technology have led to an increase in the accumulation and release of PPCPs in the environment. This paper aims to investigate the current status of PPCPs pollution and their treatment technology. Currently, there are abundant types of PPCPs, originating from residences, medical facilities, PPCP producers, etc. The main detection techniques for PPCPs are UPLC and HPLC, which can detect efficiently and quantitatively.There are high ecological and environmental risks as well as human health risks associated with the pollution of PPCPs. In addition, this paper further introduces the new technologies and research related to the removal and control of PPCPs at home and abroad, which are mainly divided into physical, chemical and biological methods, and analyzes the scenarios and limitations of each method. Finally, the challenges and problems of PPCPs pollution control are analyzed, and the research outlook of applying the emerging removal technologies to actual wastewater treatment facilities is presented. The study in this paper can provide a reference for further research on PPCPs treatment.

1 INTRODUCTION

Water resources are an indispensable and important resource for human production and life. Both water quantity and quality are crucial indicators of water resources. Currently, the world's population and standard of living are increasing, and so is the demand for water resources. However, the development of modern production technology and economic level may have more impacts and pollution on the environment, thus reducing the quality of water and the amount of available water resources. In addition to reducing water consumption, preventing and combating water pollution has become particularly important.

Water pollution often originates from the discharge of domestic, industrial, and agricultural water into natural water bodies during human activities, and common conventional pollutants include pesticides, feces and urine, heavy metals, oil, and microorganisms. Traditional water treatment methods, regulations, and standards are often set for these pollutants. In recent years, new pollutants that are different from conventional pollutants have appeared on the scene. These include microplastic pollution due to the long-term use of difficult-to-biodegrade plastic products, and persistent organic pollutants (POPs) resulting from the misuse of chemicals such as pesticides.

New pollutants are more diverse than traditional pollutants. However, since emerging contaminants are more difficult to recognize as pollutants due to their insignificant short-term hazards and often low concentrations in the environment (Wang, 2022). At the same time, such pollutants are also often characterized by easy bioaccumulation, are not easily degraded and become enriched with the food chain, posing hazards to the biological chain and human health (Álvarez-Ruiz, 2021).

Pharmaceutical and personal care products (PPCPs) are a representative group of emerging contaminants. PPCPs mainly include painkillers, antibiotics, sunscreen, cosmetics, etc. Due to the improvement of human living standards and the development of medical technology, the number of people using medicines and personal care products

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has increased, increasing the accumulation and emission of PPCPs in the environment. As a result, this type of pollutant has received wider and wider attention in the field of environmental protection. Currently, more than 3,000 types of PPCPs are in use, and this number is increasing with technological advances and product development (Arpin-pont, 2016). The unabsorbed portion of the pharmaceuticals in PPCPs leaves the body through excretion after they are used, and part of the personal care products in PPCPs enters into the water after they have been washed and rinsed. Both go into the drainage system and are treated in sewage plants before being discharged into water bodies. However, as a new pollutant, there is a lack of clear definitions of discharge indicators in wastewater treatment plants and loopholes in traditional wastewater treatment methods. Therefore, most of the PPCPs are still discharged into water bodies, causing water pollution.

This paper aims to discuss the current situation of PPCPs pollution, including types, common sources and related monitoring technologies, and to analyze the environmental and health impacts of PPCPs pollution. It briefly summarizes the domestic and international methods and researches related to the removal and control of PPCPs, which are mainly divided into physical, chemical and biological methods. This will help readers to understand the current status of PPCPs pollution and the progress of treatment technology, and provide reference for further research on PPCPs treatment.

2 BACKGROUNDS OF PPCPS

2.1 Classification of PPCPs

PPCPs can be simply categorized into pharmaceuticals and personal care products. Within pharmaceuticals, they can be further categorized into steroids as well as Nonsteroidal pharmaceuticals (Ebele, 2017).

For steroids, the representative ones are the various types of medicinal hormones, such as Estrone, progesterone, hydrocortisone, phytosterols, etc. Among the steroids, the sex hormones are related to sexual function and secondary sexual characteristics. They are commonly used in the treatment of diseases such as menstrual disorders, habitual abortion, and uterine hypoplasia. On the other hand, adrenocorticotropic hormones have mostly metabolic regulatory effects, often have antiviral, anti-inflammatory, and anti-allergic pharmacological effects, and are used in treating severe toxic infections. These steroids enter water bodies after use, mainly through excretion and improper disposal.

Nonsteroidal pharmaceuticals also come in a wide variety of types, including antibiotics such as penicillin, tetracycline, and amoxicillin, analgesics such as paracetamol, anti-inflammatories such as ibuprofen, diclofenac, and naproxen, as well as allergy medications and antidepressants. They are widely used in daily treatment and enter the water column through pathways similar to steroids.

Among personal care products, the most common nowadays are UV screens, which include benzophenone-3, homosalate, octyl-methoxycinnamate, etc. There are also fragrances, which may include musk xylol, galaxolide, tonalide, and so on, as well as disinfectants and conservation agents.

The main sources of PPCPs in water bodies are homes, medical facilities, PPCP producers, livestock farms, and landfills (Adeleye, 2022). In homes, it enters water bodies with the excretion and washing of every individual who uses pharmaceuticals and personal care products. Healthcare facilities are primarily source of pharmaceuticals, which enter the water column with the water used to clean medical devices and with the excretions and washings of patients. Producers of PPCPs have a variety of routes of leakage into the water column from their production lines, resulting in effluents that contain high levels of specificity and concentration of PPCPs. In livestock farms, there may be misuse of antibiotics and other substances, and excretions from the livestock enter the environment. On the other hand, landfills may discharge PPCPs from leachate into water bodies, depending on the waste landfilled and the temperature at the time of landfilling. On the other hand, wastewater treatment plants often aggregate wastewater containing PPCPs transmitted from various sources. Due to the current capacity limitations of wastewater treatment plants, which are not set up to treat PPCPs, most of the PPCPs will not be treated by conventional means and will continue to flow into natural water bodies.

2.2 Detection Methods of PPCPs

PPCPs have non-negligible hazards, and in addition to bioaccumulation and toxicity, they are characterized by a high degree of invisibility. This is due to the fact that PPCPs, although not easily degradable, have a relatively low concentration in water bodies compared to traditional pollutants. Currently, HPLC and UPLC are the main techniques used for the detection of pharmaceuticals. This detection technique uses a liquid as the mobile phase and separates the components within a column for the purpose of detection. Hong et al. established the use of UPLC in order to rapidly detect more than 60 pharmaceuticals in water and to quantify the drugs (Hong, 2015). UPLC-APPI-MS is also one of the techniques used for the detection of personal care products, Lung et al. developed a method for the detection of six synthetic musks using this technique with high sensitivity and chromatographic separation was achieved in a relatively short period of time (Lung, 2011). High performance liquid chromatography-mass spectrometry is also one of the main techniques used for the detection of personal care products, Lu Jing et al. developed a method for the determination of 19 PPCPs in water using this technique (Lu, 2019). HPLC-MS and UPLC-MS have shown high efficiency and accuracy in determining the concentration of PPCPs, but as more and more PPCPs are being used and entering into the water column, there are still many compounds that need detection methods with high efficiency and high sensitivity.

3 ENVIRONMENTAL AND HEALTH IMPACTS OF PPCPS

3.1 Ecological Impact

PPCPs that have not been removed by wastewater treatment enter natural water bodies, where the large volume of natural water has a dilution effect, and where the concentration of PPCPs will be lower than that measured at the wastewater treatment plant. Despite of the low concentration, the characteristic of PPCPs that deserves our attention is that it is detected in a very wide range. In addition to natural water bodies, PPCPs have been detected in algae, shellfish, shrimps, crabs, fish, and even birds, and they can be transported and distributed along the food chain in living organisms. In addition, PPCPs exhibit inhibitory effects on the growth of algae and adverse effects on the ability of some organisms to reproduce. Due to its bio-accumulative and hazardous nature and the pseudo persistence caused by its continuous release into the environment, PPCPs are susceptible to ecological impacts.

He et al. found that sulfadiazine (SDZ) and triclocarban (TCC) adversely affected the reproduction of microorganisms such as Daphnia magna in water bodies, and that TCC was easily enriched in Daphnia magna (He, 2023). The concentration of PPCPs in water bodies has not caused extreme effects on the growth and reproduction of microorganisms in water bodies. However, it is foreseeable that if PPCPs continue to be discharged uncontrolled into the natural environment, when their concentration reaches a certain level, the inhibitory effect on the reproduction of microorganisms in the water bodies is too strong, and it may cause destructive impacts on the microbial population structure. Some studies have evaluated the environmental risk of PPCPs. Nie et al. used the RQ model to evaluate the environmental risk of estrogen in the Yangtze River estuary, which expressed a moderate, or even partly high, environmental risk in different seasons (Nie, 2015). PPCPs not only affect the ecosystems, but also pose a threat to human health through the ecosystems.

3.2 Human health risks

While PPCPs enter the food chain in the water environment and enter the bodies of animals, it may also migrate into the soil environment and accumulate in crops. The PPCPs that humans release into the natural environment may end up back on human's own tables or even in human bodies, posing health risks. On the one hand, a large portion of PPCPs are often disease-specific or designed for topical use, and they have the property of disrupting the normal healthy human endocrine system and cannot accumulate in the body over time. For example, medicines such as steroids may interfere with the body's normal hormone metabolism. Personal care products such as synthetic fragrances have been clinically observed to affect the central nervous system at certain concentrations (Yang, 2010).

On the other hand, the misuse of PPCPs such as antibiotics has led to the persistence of low concentrations of pharmaceuticals in the environment, which is largely responsible for the emergence of drug-resistant genes in bacteria, which are also passed on as bacteria proliferate (Storteboom, 2010). The occurrence of drug-resistant bacteria is a test for the human healthcare system, which means that even colds may be more difficult to cure, and doctors need to be more careful in terms of the number of types of drugs they prescribe. The threat that PPCPs pollution can pose to human health can no longer be ignored and deserves more attention.

4 TREATMENT TECHNOLOGY OF PPCPS

PPCPs pollution control technology can be categorized into physical, chemical and biological treatment technologies through the methods used. Physical treatment methods do not change the chemical properties of PPCPs substances, such as adsorption method. They are usually simple, flexible, and have few by-products, and are suitable for wastewater with relatively high concentrations of pollutants. Chemical treatment methods change the chemical properties of PPCPs substances. They have high removal efficiency and complete degradation. Biological treatment methods apply microorganisms, plants and animals for the degradation of pollutants. They have been widely used in the degradation of organic pollutants, and in recent years there has been greater progress in the treatment of PPCPs technology. They can effectively reduce the toxicity of pollutants and the quality of the effluent is relatively stable.

4.1 Physical Treatment Methods

Physical treatment methods refer to the removal of pollutants in the environment by physical means and do not change the chemical nature of the pollutants. Commonly used physical treatment methods include sedimentation, filtration and flotation. The principle of the precipitation method is that the relative density is different, thus separating the suspended substances in the liquid. The principle of the filtration method is to use porous filter media, thereby retaining the suspended particles in the liquid. The principle of the flotation method is to pass air as a carrier in order to remove pollutants in water that have a density similar to that of water. However, the concentration of PPCPs in water is usually small and dissolved in water, so physical adsorbents need to be introduced in order to achieve removal using physical treatment methods.

Common physical adsorbents are activated carbon, biochar, graphene-based adsorbents, carbon nanotubes, etc. (Ayati, 2023). Javier et al. investigated the adsorption performance of powdered activated carbon (PAC) as an adsorbent for four antibiotics in water bodies, and the adsorption performance for different antibiotics expressed different adsorption capacities, with the removal of enrofloxacin at 28% and sulfadiazine at 67% at the adsorbent concentration of 100 mg/L. For antibiotics present in trace quantities in the water column, PAC showed an effective removal capacity (Berges, 2021). Shin et al. investigated the adsorption properties of NaOH-activated biochar (BC) from spent coffee wastes as an adsorbent for pharmaceuticals in water such as naproxen, diclofenac, and ibuprofen, and the chemically activated BC had a stronger adsorption capacity compared to normal BC (Shin, 2021).

The physical adsorption treatment method is simple to apply and has demonstrated effective removal of PPCPs substances with fewer by-products. However, it should be noted that the adsorbent needs subsequent regeneration treatment. From the study, it can be seen that the removal effect is closely related to the type of the adsorbent and pollutants, and the removal rate is relatively limited, which is more suitable as a pre-treatment of a wastewater treatment plant, or for the case of a single type of pollution.

4.2 Chemical Treatment Methods

Chemical treatment methods require changing the chemical properties of the contaminant to achieve the purpose of removing the contamination. Commonly used chemical methods include chemical precipitation, which is similar to physical precipitation, with the difference being that the pollutants are recovered and removed through chemical means by making them easier to precipitate. In addition to this, there are electrolysis, oxidation, photocatalysis and other chemical treatment methods to degrade the pollutants in the water body, so that the pollutants are harmless.

Peng et al. prepared CuZnAl-layered double hydroxide and examined its performance for photocatalytic degradation of Naproxen under UV irradiation, and the removal efficiency of Naproxen was up to 98.25% under the optimum conditions they tested, i.e., pH 9.0 and catalyst dosage of 0.25 g/L (Peng, 2022). Luo et al. used the UV365-LED/chlorine process for the degradation and removal of metronidazole with degradation efficiency up to 90.6%, and it was also found that increasing the intensity of UV light, the amount of chlorine and decreasing the pH could help in the degradation efficiency of metronidazole (Luo, 2023). Li et al. established a new electrochemical peroxone process, which can realize electrogenerated H2O2/O3 and can efficiently electrochemically decompose ibuprofen (Li, 2024).

Chemical treatment technology can reach a high removal rate and complete degradation of pollutants. Photocatalytic degradation due to its environmentally friendly characteristics, has gained widespread attention in recent years, but it is difficult to apply to larger-scale wastewater treatment. Ozone oxidation treatment is also efficient but relatively costly.

Chemical treatment technologies usually cost high and are complicated to apply.

4.3 Biological Treatment Methods

Biological treatment methods, on the other hand, introduce biological means in the process of removing pollutants, mainly utilizing the metabolism of microorganisms to absorb or degrade pollutants in water. Microbial treatment methods can be simply divided into aerobic biological treatment and anaerobic biological treatment, but also often used in conjunction with the two. Biochemical tanks are currently one of the main treatment units in the sewage treatment plant. In addition to microorganisms, a part of plants and animals can also be used as pollutant absorption treatment, often used in artificial wetlands.

Mokhtariazar et al. compared the ability of two technologies, Membrane bioreactor (MBR) and MBR with fixed-bed packing media (FBMBR), to remove Naproxen, and demonstrated that FBMBR has a higher efficiency for the removal of PPCPs represented by Naproxen (Mokhtariazar, 2024). Iliopoulou et al. developed a system that combined strictly anaerobic MBBR and aerobic MBR, which was effective for the removal of metronidazole (MTZ), trimethoprim (TMP), sulfamethoxazole (SMX) , and valsartan (VAL), can achieve removal efficiencies of more than 65% and biotransformation is the main principle for the removal of these PPCPs, while Bacteroidetes are the dominant microorganisms in this system (Iliopoulou, 2023).

The biological treatment methods have a stable system, the effluent water quality is also stable, and it can effectively reduce the toxicity of pollutants. Compared with the traditional activated sludge method, MBR technology has obvious removal effect on PPCPs pollutants, but the cost is also greatly increased. Artificial wetlands, phytoremediation and other methods are less costly and environmentally friendly but are susceptible to environmental climate and other factors, and are relatively less efficient.

5 CHALLENGES AND PERSPECTIVES OF POLLUTION CONTROL OF PPCPs

Since the concept of PPCPs has come into the limelight, more attention has now been paid to its pollution control, and a large number of studies have been conducted on the detection and removal of various types of PPCPs pollutants. In terms of pollutant detection, the main technical challenge faced by the research initially is that PPCPs usually exist in trace amounts in the aqueous environment and are relatively difficult to detect. With the introduction of UPLC, HPLC and other detection methods, the difficulty of trace detection has been reduced. The new challenge is to find and identify the pollutants with high ecological risk in the water body. In the face of the rich variety of PPCPs pollutants, there is a need to identify which ones require our immediate attention and control.

Current studies have focused on several common types of PPCPs contaminants, mainly pharmaceuticals, such as Naproxen and ibuprofen, and relatively little research has been conducted on personal care products. Different pollution control methods have different removal effects for different PPCPs contaminants, and the research on removal methods is still far from adequate in the case of increasing types of PPCPs. In addition, most of the pollution control methods are highly targeted, and it is difficult to find a generalized control method. In the face of the complex pollution of the water environment, how to apply these methods to achieve the best PPCPs pollution control efficiency at a lower cost is also a difficult problem.

Wastewater treatment plants, as the core of urban drainage systems, are responsible for the removal of most PPCPs. Therefore, future research should focus on how to apply emerging removal technologies to actual wastewater treatment facilities, as well as upgrading existing treatment processes to enable them to effectively respond to the challenges of novel pollutants. Through interdisciplinary cooperation and technological innovation, pollution control of PPCPs will be able to achieve better results.

6 CONCLUSIONS

PPCPs are potentially hazardous emerging contaminants that deserve attention and in-depth study. In this paper, the current status of PPCPs contamination is first introduced, and the types of PPCPs are discussed from pharmaceuticals and personal care products, as well as the common sources. The concentration of PPCPs in water is usually trace, and the commonly used detection methods are HPLC and UPLC, which can achieve high efficiency and high sensitivity. The pollution of PPCPs has high ecological and environmental risks as well as human health risks. In terms of PPCPs removal technology, the adsorption method, which is commonly used in physical treatment methods, is simple and has fewer by-products, but the removal effect is related to the type of application, which is suitable for pre-treatment or simple pollutants. Chemical treatment methods can completely degrade pollutants, but the application is complex and costly, while photocatalytic degradation is suitable for small-scale wastewater treatment. Biological treatment methods have a stable system, can reduce toxicity, and the effluent quality is stable, but the MBR method is costly, and the artificial wetland method is susceptible to climate, so they need to be adapted to local conditions. Understanding the current status of PPCPs pollution and treatment technology can provide a reference for further research on PPCPs removal. Future research on PPCPs pollution control should aims to discover and identify PPCPs pollutants with high ecological risks and apply emerging treatment technologies to actual wastewater treatment facilities to face the challenges of emerging contaminants.

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