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Review

Occurrence and Removal of Pharmaceutical Contaminants in Urine: A Review

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Abstract: With the development of world economies and the continuous improvement of living standards, pharmaceutical and personal care products (PPCPs) have attracted significant attention because of their widespread detection in wastewater and the natural environment. Their biological toxicity, environmental persistence, and other hazardous characteristics might pose a threat to the ecological environment and human health. How to treat source-separated urine as a valuable recyclable resource has become a novel challenge. In this review, we briefly described the sources of pharmaceuticals, explored the various metabolic pathways of pharmaceuticals, and concluded that urinary excretion is the primary metabolic pathway of pharmaceuticals. Next, the current status of pharmaceutical contamination in human urine, sewage plants, and surface water was summarized. It is shown that the concentration of pharmaceuticals in human urine is usually 2–3 orders of magnitude higher than that in sewage plants and surface water. Then, the research progress of various technologies to treat pharmaceutical contaminants in urine was analyzed and compared, indicating the promise of advanced oxidation technologies to treat such wastewater, among which electrochemical oxidation has received widespread attention due to its advantages of cleanness, flexibility, and controllability. Therefore, the research progress of electrode materials and electrochemical technology to treat urine was reviewed, and finally, the future development direction was proposed, namely, coupling membrane treatment technology with intellectual development, which will help realize the scale and industrialization of source-separated urine treatment.

Keywords: pharmaceutical; source-separated urine; metabolic pathways; urine treatment technology; advanced oxidation; electrochemical oxidation

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1. Introduction

Human urine has a complex and diverse composition, containing many contaminants as well as nutrients such as nitrogen and phosphorus, which show both resource and pollution characteristics. Studies have shown that although urine wastewater accounts for only 1% of the volume of domestic wastewater, it contains about 80%, 56%, and 63% of the N, P, and K of domestic wastewater, respectively [1].

Urban sewers were originally used to prevent flooding in urban areas. However, with the development of cities and societies, they were converted to also carry human excreta into wastewater treatment plants (WWTPs) for centralized disposal to ensure the sanitary safety of the city [2]. This treatment method, which has been used up to now, inevitably increases the treatment load and operation cost of the sewage plant and even wastes valuable nitrogen and phosphorus resources to some extent. In 1985, Uno Winblad first

proposed, "Do not mix feces with urine and do not mix feces with water," which was the earliest concept of urine source separation [3]. Since the 1990s, more scholars have started to research source-separated urine and put forward the concepts of sustainable drainage, decentralized drainage, and ecological drainage [4]. The aim is to separate human feces and urine from domestic sewage at the source and reduce the pollution load of sewage plants while recovering energy and resources. Up until now, various recovery technologies have been developed for the resource treatment of urine, including evaporation concentration, guano stone recovery, and membrane separation technology. Tun et al. [5] used direct contact membrane distillation to concentrate nitrogen in source-separated urine and finally obtained a highly concentrated product. Guan et al. [6] studied the recovery of phosphorus from acidified urine by magnetite nanoparticles, and the results show that the recovery rate of phosphorus exceeds 90%.

However, various pharmaceutical contaminants can be detected in human urine. The way these pharmaceutical contaminants enter the environment is greatly influenced by human activities, including oral administration, injection into the human body, and metabolism in the human body, which ultimately discharge into the sewer network in various forms. Previous studies have shown that the body only absorbs a small portion of pharmaceuticals, and the vast majority are excreted in the urine as primitive drugs or metabolites. It has been found that pharmaceutical contaminants such as antibiotics and hormones may affect the safety of the practical application of source-separated urine. Heinonen-Tanski et al. [7] reported that the use of untreated human and animal excreta as nutrient fertilizer in agricultural irrigation may lead to the spread of pathogens. With the introduction of pharmaceutical contaminants into the water environment, the occurrence of hermaphroditic fish in the vicinity of wastewater treatment plants around the world is increasing (Larsson et al. [8]). They can pose a threat to aquatic organisms through the food chain (Sharma et al. [9]). In the long run, these pharmaceutical contaminants are likely to have potential impacts on human health and the ecological environment (Escher et al. [10]). Therefore, their environmental risks need to be addressed in the "collection-treatment-reuse" process of source-separated urine to ensure its safety in the real world. Pharmaceuticals include a variety of prescription and over-the-counter drugs (such as antibiotics, pain, and anti-inflammatory drugs, cardiovascular drugs, hormonal drugs, etc.) [11]. In recent years, with the improvement of detection and analysis technology, pharmaceutical contaminants have been widely detected in sewage, surface water, soil, and human urine, and their biological toxicity, environmental persistence, bioaccumulation, and other characteristics may cause potential risks and hazards to the water environment and human health [12], which has now become a research hotspot in the environmental field. To sum up, it is urgent to develop efficient and economical technologies to remove pharmaceutical contaminants from urine.

The first review on urine treatment was published by Maurer et al. [13], and later, some other reviews and research papers have been published, including a structured overview of urine source separation, further urine treatment, and recovery technology. For example, Yan et al. [14] commented on the application of source-separated urine, suggesting that the sedimentation problem is a massive challenge for practical application. Some other domestic and foreign researchers have summarized the characteristics and research status of existing source-separated urine treatment technologies, including physical, biological, chemical, and ecological treatment methods [15,16]. However, few reviews summarized the degradation of pharmaceutical contaminants in source-separated urine.

This study tried to review the sources, metabolic pathways, and pollution status of pharmaceutical contaminants in the environment and compare the research progress of source-separated urine treatment. It also discussed the possible problems and presented an outlook for future research.

2. Methods

This study selects Web of Science as the database to search for relevant peer-reviewed papers. In order to retrieve the research papers related to Chapter III "Classification, Sources and Metabolic Pathways of Pharmaceutical Contaminants", TS = (("classification" OR "source" OR "metabolic pathway" OR "occurrence") AND ("pharmaceutical" OR "drug" OR "antibiotic" OR "medicine") AND ("pollutant" OR "contaminant")) is adopted in the Web of Science search. Similarly, for the fourth chapter "Pharmaceutical contaminants in human urine and other different environmental media", TS = (("pharmaceutical" OR "drug" OR "antibiotic" OR "medicine") AND ("pollutant" OR "contaminant") AND ("urine" OR "urinary") AND ("WWTP" OR "surface water" OR "sewage treatment plant")) is adopted in the Web of Science search. Finally, for the fourth chapter "Research progress of urine treatment", TS = (("urine" OR "urinary") AND ("treatment" OR "processing" OR "electrochemistry" OR "electrocatalysis" OR "electrooxidation") AND ("pharmaceutical" OR "drug" OR "antibiotic" OR "medicine") AND ("pollutant" OR "contaminant")) is adopted in the Web of Science search. Then, we further extracted useful information from the retrieved documents for an in-depth analysis.

3. Classification, Sources, and Metabolic Pathways of Pharmaceutical Contaminants

3.1. Classification of Pharmaceutical Contaminants in the Environment

Pharmaceutical contaminants mainly include eight categories: antibiotics, hormones, antiepileptics, analgesics and anti-inflammatory drugs, blood lipid regulators, β Receptor blockers, and stimulants (Table 1) [17–19]. Among the commonly used drugs, antibiotics have received special attention because of their wide application in human medicine, animal husbandry, and agriculture. Some studies have shown that the most frequently reported substances are antibiotics [20]. There are many types of antibiotics, including sulfonamides β- Lactams, aminoglycosides, fluoroquinolones, macrolides, and tetracyclines. Excessive use and continuous exposure to antibiotics will lead to the emergence of antibiotic-resistant strains, which are easy to cause public health problems [21]. Hormone drugs are another concerned category of drugs, and they are believed to interfere with the human or animal endocrine system [22]. The most deeply studied hormone is the natural steroid estrogen, including estrone (E1), estradiol (E2), and estradiol (E3), which are mainly excreted by humans and animals. Additionally, synthetic steroid estrogen is used as an oral contraceptive, mainly ethinyl estradiol (EE2) [23]. Natural steroid estrogen is not actually a traditional medicine, but it is usually used to study the endocrine-disrupting effect of synthetic hormones in the water. Other drugs, such as blood lipid regulators (clofibric acid, bezafibrate), can inhibit lipolysis in adipose tissue. β Receptor blockers (metoprolol, propranolol) are often used to treat hypertension. Analgesics and anti-inflammatory drugs such as diclofenac and ibuprofen. Common stimulants include caffeine, cocaine, etc., which are used to reduce body fatigue and improve the thinking activity of the mind. Antiepileptic drugs include carbamazepine and primidone, and carbamazepine has become a research focus in recent years due to its refractory nature [24–26].

3.2. Sources of Pharmaceutical Contaminants in the Environment

The way pharmaceutical contaminants enter the environment is usually affected by human activities: some pharmaceuticals enter the human body through oral or injection and finally discharge in various forms into the sewer network. Some topical pharmaceuticals, such as fluoxetine ointment and ofloxacin ointment, usually enter domestic sewage during bathing or swimming. Even expired pharmaceuticals are discarded at will, leading to these pharmaceuticals entering the water, soil, and other environmental media [27]. Moreover, in many low- and middle-income countries, due to inadequate regulatory and legal systems, most pharmaceutical industries choose to discharge wastewater into sewers in violation of regulations (Figure 1). Therefore, wastewater treatment plants are widely considered the primary source of pharmaceutical contaminants entering the environment [28]. The

presence of pharmaceutical contaminants in wastewater treatment plants has been reported in various countries worldwide, usually at a level of several to thousands of ng/L.

Table 1. Classification of pharmaceutical contaminants.

	Subgroups	Representative Compounds
Pharmaceuticals	Antibiotics	Clarithromycin Sulfamethoxazole Sulfadimethoxine Norfloxacin Ciprofloxacin
	Hormones	estrone (E1) estradiol (E2) ethinylestradiol (EE2) estradiol (E3)
	antiepileptics	Carbamazepine Primidone
	Analgesics and anti-inflammatory drugs	Ibuprofen Diclofenac Acetaminophen
	Blood lipid regulators	Gemfibrozil Clofibrate
	β-blockers	Propanolol Metoprolol
	Stimulants	Caffeine Cocaine

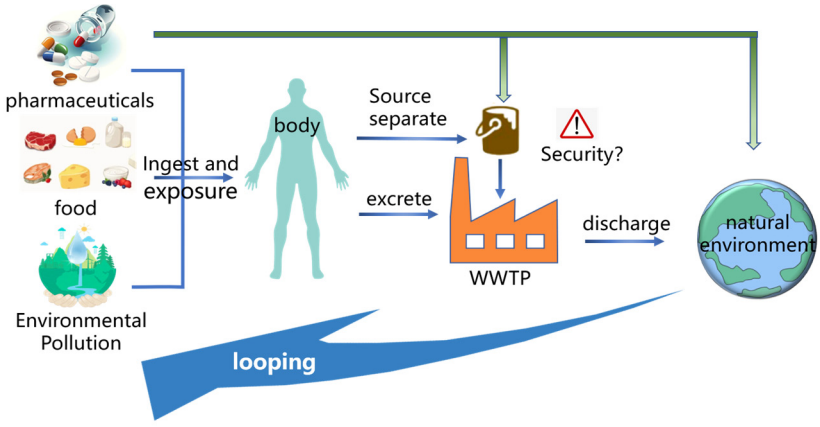

Figure 1. Sources of pharmaceutical contaminants in the environment.

3.3. Metabolic Pathway of Pharmaceuticals in Organisms

It has been shown that pharmaceuticals enter the body either orally or by injection, and the body only absorbs a small percentage. At the same time, the vast majority is excreted in the urine and feces as original medicine or metabolites. For example, Leinert et al. [29] reported that although the situation in the human body varies, an average of 64 (±27)% of different pharmaceuticals are excreted through the urine, while 35 (±26)% are excreted through the feces. The pharmacokinetic correlation analysis presented in Figure 2. Table 2 also indicates that the majority of pharmaceuticals are excreted through the urine.

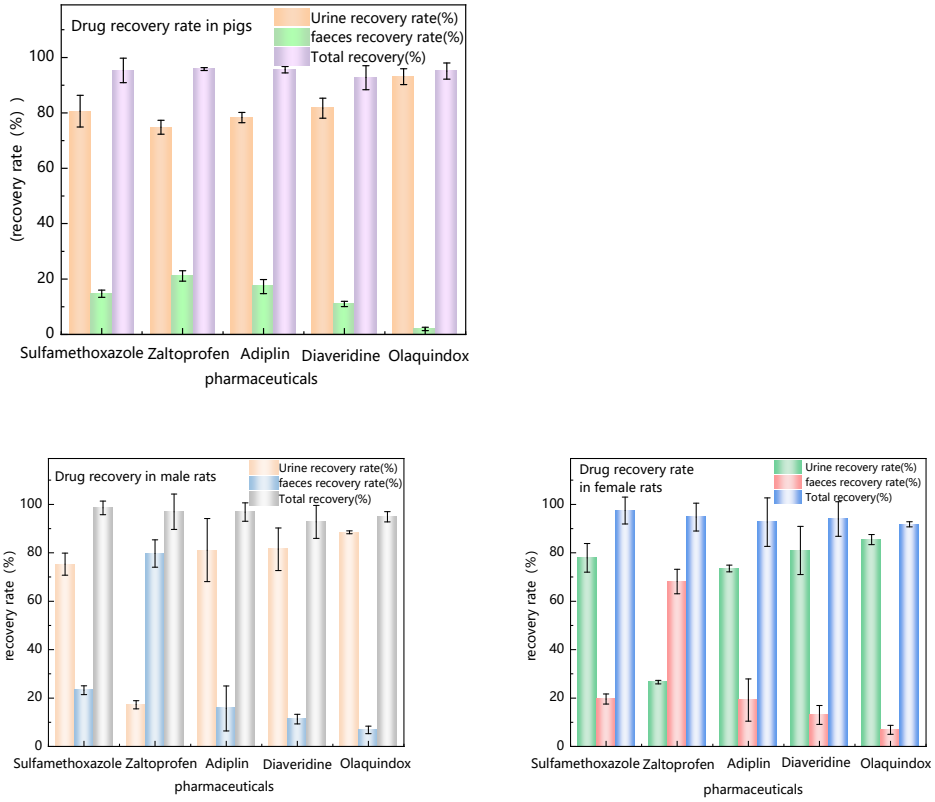

Figure 2. Drug recovery after intramuscular drug injection in different animals (data from [30–34]).

Table 2. Drug recovery after intramuscular drug injection in different animals.

	Drug Name	Recovery Rate in Urine (%)	Recovery Rate in Feces (%)	Total Recovery Rate (%)	Reference
Drug recovery after a single intramuscular drug injection in pigs	Sulfamethoxazole	80.59 ± 5.72	14.72 ± 1.31	95.31 ± 4.41	[30]
	Zaltoprofen	74.80 ± 2.52	21.13 ± 1.90	95.82 ± 0.51	[31]
	Adiprin	78.28 ± 1.86	17.29 ± 2.54	95.57 ± 1.16	[32]
	Diaveridine	81.7 ± 3.61	11.00 ± 0.97	92.70 ± 4.23	[33]
	Olaquindox	93.08 ± 2.87	1.98 ± 0.61	95.07 ± 2.93	[34]
Drug recovery in male rats after a single intramuscular drug injection	Sulfamethoxazole	75.32 ± 4.54	23.24 ± 1.79	98.56 ± 2.82	[30]
	Zaltoprofen	17.23 ± 1.70	79.73 ± 5.65	96.97 ± 7.28	[31]
	Adiprin	81.12 ± 13.03	15.7 ± 9.27	96.82 ± 3.81	[32]
	Diaveridine	81.50 ± 8.81	11.30 ± 2.01	92.80 ± 6.81	[33]
	Olaquindox	88.48 ± 0.56	6.82 ± 1.57	94.89 ± 2.09	[34]
Drug recovery in female rats after a single intramuscular drug injection	Sulfamethoxazole	77.9 ± 5.93	19.58 ± 2.09	97.48 ± 5.56	[30]
	Zaltoprofen	26.61 ± 0.73	68.16 ± 5.06	94.77 ± 5.76	[31]
	Adiprin	73.53 ± 1.40	19.18 ± 8.73	92.7 ± 10.01	[32]
	Diaveridine	80.98 ± 9.92	13.00 ± 3.88	93.98 ± 7.14	[33]
	Olaquindox	85.45 ± 2.08	6.87 ± 1.86	91.79 ± 1.03	[34]

Radiolabeling technology has the advantages of high sensitivity, high accuracy, and high specificity. It is internationally recognized as the most recommended method for studying drug absorption, metabolism, excretion, and elimination. Some scholars studied the excretion of tritium-labeled drugs in pigs and rats after a single administration. The five drugs were recovered to essentially more than 90% of the total radioactivity over the cumulative recovery period in different animals, indicating no significant accumulation of these five drugs in the studied animals [30–34]. Comparison of drug excretion after a single intramuscular injection of different drugs in the same species of animals. In pigs, the urinary recovery of all five drugs after a single intramuscular injection of the drugs was

above 70%, of which the urine recovery rate of olaquindox was 93.08% and only 1.98% in feces, indicating that the five drugs were mainly excreted through urine and the kidney was the primary excretory organ [34]. In rats, after a single intramuscular injection of the drugs, the urinary recovery of most of the drugs was higher in males or females, exceeding 70%, but the fecal recovery of zaltoprofen in male and female rats was 79.73% and 68.16% [31]. Respectively, indicating that although most of the drugs were excreted through urine, there existed a small proportion of drugs that were excreted through feces, which confirmed the view of Leinert et al. [29].

The major metabolic pathways of these drugs include acetylation, hydroxylation, glycolipidation, and carboxylation. For example, acetylation is the main metabolic pathway of sulfamethoxazole in organisms [35], and the metabolites produced by metabolic transformation may be as toxic or active as the parent compound or even much more toxic than the drug itself [36]. On the other hand, most of the pharmaceuticals and their metabolites have good solubility in water but still have difficulty in degradation and transformation, showing a general persistent or pseudo-sustained state due to their continuous discharge of sewage [37], thus threatening the growth of aquatic organisms and bioaccumulating through the food chain, ultimately affecting human health and safety.

4. Pharmaceutical Contaminants in Human Urine and Other Different Environmental Media

It has been shown that traditional wastewater treatment processes such as flocculation, sedimentation, and activated sludge treatment have limited effects on the removal of pharmaceutical contaminants, and the removal efficiency is usually about 20~30% [38,39]. Therefore, most of the inadequately degraded pharmaceuticals are discharged back into the natural water bodies. The presence of several high concentrations of pharmaceutical contaminants and metabolites has been detected in wastewater plants in several regions of the world [40–42]. Studies on human exposure characteristics and the health risks of emerging contaminants have also summarized the concentration of ECs in human urine [43–46]. Table 3 and Figure 3 list the concentrations of some pharmaceuticals in humans, wastewater plants, and surface water, respectively. The results show that pharmaceutical concentrations in urine are usually 2–3 orders of magnitude higher than those in municipal wastewater treatment plants, and pharmaceutical concentrations in wastewater treatment plants and surface water are generally at the ng/L level.

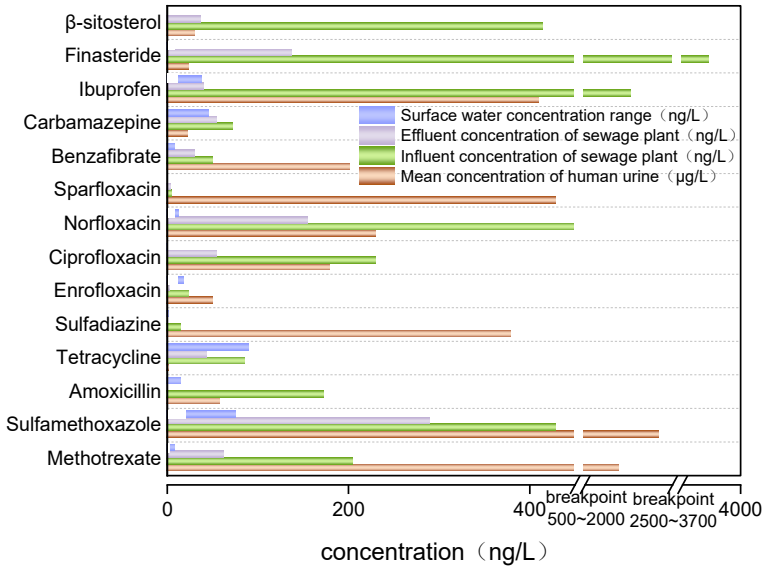

Figure 3. Concentration distribution of pharmaceutical contaminants in human urine, sewage plants, and surface water (data from [43,47–69]).

Table 3. Concentration distribution of pharmaceutical contaminants in human urine, WWTPs, and surface water.

Pharmaceutical	Human Urine (μg/L)	Reference	Influent in WWTP (ng/L)	Effluent in WWTP (ng/L)	Reference	Surface Water (ng/L)	Reference
Methotrexate	2199 (0.7–12800) [a]	[43]	205	63	[47]	6–8	[48]
Sulfamethoxazole	2430 (ND–7740) [a]	[43]	430	290	[49]	19.25–75.48	[50]
Amoxicillin	58.1 (ND–310) [a]	[43]	172.6	ND	[51]	0–15.1	[52]
Tetracycline	1.4 (ND–2.8) [a]	[43]	85.4	43.1	[53]	ND–90.7	[54]
Sulfadiazine	380 [b]	[55]	15	ND	[56]	ND–1.898	[57]
Enrofloxacin	50 [b]	[55]	23.93	2.47	[58]	10.5–18.7	[59]
Ciprofloxacin	180 [b]	[55]	231	55	[56]	0.12–0.63	[60]
Norfloxacin	230 [b]	[55]	468	155	[56]	7.0–12.9	[59]
Sparfloxacin	430 [b]	[55]	4.7	4.1	[61]	-	-
Benzafibrate	202 [b]	[62]	50	30	[63]	8	[63]
Carbamazepine	22.7 [b]	[62]	72	55	[48]	46	[64]
Ibuprofen	411 [b]	[62]	2265	40	[48]	11–38	[65]
Finasteride	23.3 [b]	[62]	3840	138	[66]	7.7–8.6	[67]
β-sitosterol	30.8 [b]	[62]	415.56	37.22	[68]	ND	[69]

Note: ND: not detected; [a]: average concentration (concentration range); [b]: average concentration.

As the most concerning pharmaceutical, antibiotics are detected in more than 67% of urine samples, as shown in Figure 4. Two subgroups of the highly detected antibiotics, fluoroquinolones and sulfonamides, are widely used in animal husbandry and aquaculture, and they may continue to produce resistant strains that are released into the environment and enter the food chain to be absorbed by humans, consistent with the study by Li et al. [70]. In addition to antibiotics, painkillers (such as ibuprofen), antiepileptics, and lipid regulators are also commonly detected in human urine. Ngumba et al. [43] found that sulfamethoxazole, meperidine, and lamivudine were frequently detected in source-separated dry toilets in residential areas in Zambia, especially sulfamethoxazole with a maximum detected concentration of 2430 μg/L, and pointed out that the reason was that local residents took a lot of related pharmaceuticals to prevent HIV infection. Since 2010, 28 million Zambians have been able to receive antiretroviral therapy (ARV), indicating that high-frequency use of specific pharmaceuticals may also be responsible for elevated concentrations of pharmaceutical contaminants in urine. Zhong et al. [55] investigated the urine of 1170 adult residents in Shenzhen in 2017 and detected a variety of antibiotics in urine samples, such as sulfadiazine and sulfamethoxine, at concentrations of 380 and 260 μg/L. Zhong also investigated and analyzed the sources of antibiotic exposure, and the results indicated that antibiotics in meat might be an important source. Kyriakides et al. [71] reported that 45 antibiotic residues were detected in pork sold in Cyprus (a European country). It is worth noting that norfloxacin was banned by the Chinese Ministry of Agriculture as early as 2015 [72], which indicates that some banned antibiotics have already penetrated daily life.

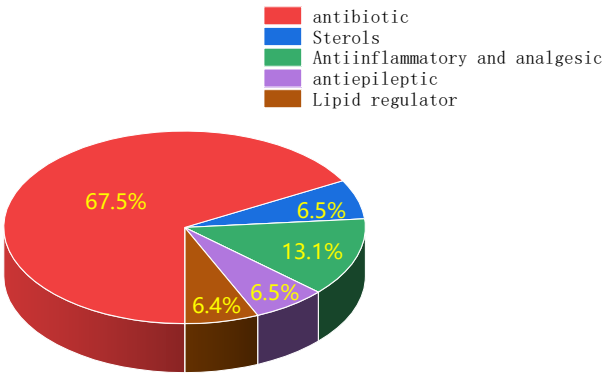

Figure 4. The main categories of detected pharmaceutical contaminants in urine (data from [43,47–69]).

The concentrations of pharmaceutical contaminants in sewage plants and surface waters selected for this study do not represent universal levels. Among them, sulfonamide antibiotics were frequently detected in sewage plants. Anke et al. [49] detected high concentrations of sulfamethoxazole in sewage plants near Zurich International Airport in

Switzerland and in a living area in the canton of Garsaint due to the high consumption of this pharmaceutical in human medicine. However, there was a huge difference in the detection of the pharmaceuticals between the sites selected for the study, leading to high concentrations of some of the pharmaceuticals detected, which may be related to the industrial structure of each region. For example, Sim et al. [73] studied that the concentration of paracetamol in the influent of a municipal sewage treatment plant in South Korea was 6.80 ± 2.41 μg/L, but the average concentration of paracetamol detected in the sewage plant of a nearby hospital is 45 μg/L, 5~12 times the concentration of traditional Chinese medicine in urban sewage and even higher than the level detected in human urine. The reasons that affect the difference in pharmaceutical concentration in the influent and effluent of the sewage treatment plant may be the specific nature of the pharmaceutical and the factors related to the sewage treatment plant, such as the type of treatment process, sludge age, sludge concentration, etc., or the quality of the influent water, COD concentration, and other factors [49,51,53]. The removal effect of the same process on mixed sewage with different pharmaceuticals may also be different [47,56]. After treatment at the plant, the pharmaceutical concentration in the surface water is generally another order of magnitude lower.

5. Research Progress in Urine Treatment

According to the above survey, pharmaceutical contaminants are detected in different environmental media, including human urine, and the treatment efficiency of traditional WWTPs is low, which might pose potential environmental risks, so there is an urgent need to develop efficient treatment methods for emerging contaminants.

5.1. Source Separation of Urine

Urine source separation refers to the collection and treatment of urine at the source to remove pollutants and achieve resource recovery. The overall process consists of three steps: the first step is the "front end" of urine separation and collection; the second step is the "middle end" of urine transportation and storage; and the third step is the "back end" of urine treatment [74]. Due to the special nature of urine source separation, the traditional "collection-transportation-storage–treatment plant" mode suffers from clogged pipes, increased transportation costs, nutrient loss, odor irritation, etc. [75,76]. Therefore, the treatment in the sewage treatment plant can only achieve the purification of wastewater but cannot effectively recover nitrogen and phosphorus nutrients in the wastewater. It is urgent to develop a new mode of source-separating urine treatment. Based on this, on-site source separation treatment of urine wastewater is recognized as one of the most promising models.

5.2. Research Progress in Urine Treatment Technology

5.2.1. Physical Treatment

In the last few decades, researchers around the world have adopted several methods to remove pharmaceutical contaminants from wastewater. Common methods include physical, chemical, and biological treatment. Physical methods of wastewater treatment are the most common and basic treatment methods, mainly using electrical attraction, van der Waals forces, gravity, and other effects to separate and remove pollutants [77]. Physical methods include coagulation, sedimentation, membrane treatment, and adsorption. There is a lot of research on the physical treatment of pharmaceutical contaminants. For example, Hassan et al. [78] used synthetic ZnO nanoparticles to treat ibuprofen, ephedrine, and propranolol in urine wastewater, and the experimental results showed that the removal rate of pharmaceuticals was above 99%, while the removal rate of TP was 59.9%, and the treated urine could be used as nutrients for agricultural production. Antonini et al. [79] used guano stones and air blowing techniques to recover nutrients from urine wastewater; total phosphorus (TP) removal was about 98% and total nitrogen (TN) removal was about 90%. Marcela et al. [80] examined the adsorption performance of rice (RH) and coffee (CH)

husk wastes as adsorbents for norfloxacin in simulated urine and found that CH husk wastes were more effective with a removal rate of 83.54%. Other researchers have also used biochar, nanofiltration membranes, resins, and other methods to remove pollutants [81–83]. However, the high production cost of commercial adsorbents and the difficulties in the treatment of waste adsorbents and membranes limit their application. And conventional physical methods only separate pollutants from wastewater and do not completely degrade them, so there are still potential environmental risks.

5.2.2. Biological Treatment

In biological treatment, pharmaceuticals are usually removed by biodegradation. The most widely used methods for the removal of pollutants from wastewater are aeration biofilters (BAF), anaerobic digestion (AD), sequencing batch reactor activated sludge processes (SBR), and membrane bioreactor (MBR) technologies. With the development and coupling of technologies, the efficiency of treating pollutants has steadily increased. Udert et al. [84] used anaerobic biological treatment technology to remove pollutants in urine wastewater. Köpping et al. [85] examined the removal of emerging contaminants from urine after nitrification treatment, which required two orders of magnitude less activated carbon than that required for treatment in central wastewater treatment plants. However, according to current studies in WWTPs, the biological treatment technology has been proven to be inefficient for many pharmaceuticals, such as the antibiotic sulfadiazine, the antihypertensive metoprolol, and the antiepileptic carbamazepine [46]. Moreover, these systems are not clearly designed to eliminate stubborn pharmaceutical contaminants. Furthermore, the domestication and survival of bacteria in highly concentrated, source-separated urine is also a problem. Therefore, if a biological method is to be used to treat pharmaceutical pollutants in urine, it is necessary to better study the removal mechanisms of contaminants, coexisting ions, and organic substances and constantly optimize the bacterial culture and treatment process.

5.2.3. Chemical Treatment

The stagnant inefficiency of conventional methods and the increasing water pollution have continuously propelled the development of new technologies. Nowadays, advanced oxidation processes such as Fenton oxidation, ozone oxidation, and electrochemical oxidation in chemical treatment have become the most promising wastewater treatment methods [86,87]. Advanced oxidation technology can generate a large number of strong oxidizing groups, such as hydroxyl radicals or sulfate, in the treatment of organic wastewater, which can efficiently degrade the persistent contaminants in the wastewater and improve the biochemical properties of the wastewater. Cotillas et al. [88] investigated the degradation performance of chloramphenicol using electrolytic and photoelectrolytic ultrasonic electrolysis, and the results showed that pollutants such as chloramphenicol could be completely mineralized, achieving a significant reduction in pollution diffusion. Diana and Clozaril [89,90] investigated the degradation of pharmaceuticals in urine by the sonochemical advanced oxidation process, which was also able to reduce the bacterial activity, and the removal rates are above 90%. Sebuso et al. [91] prepared multilayered graphene (MLG) from biomass waste, synthesized MLG nano-sheets from corn husks through multiple processes, and studied the photocatalytic degradation performance of MLG/ZnO nano-composites on doxycycline (DOX). The results showed that the degradation rate of DOX reached 95% under ultraviolet light. This scheme of preparing high-performance materials from biomass waste provides a sustainable way for solving environmental problems. These show that advanced oxidation processes have a wide range of prospects for the treatment of pharmaceuticals. [88,90–92]. The model diagram of common treatment methods corresponding to urine treatment technology is shown in Figure 5.

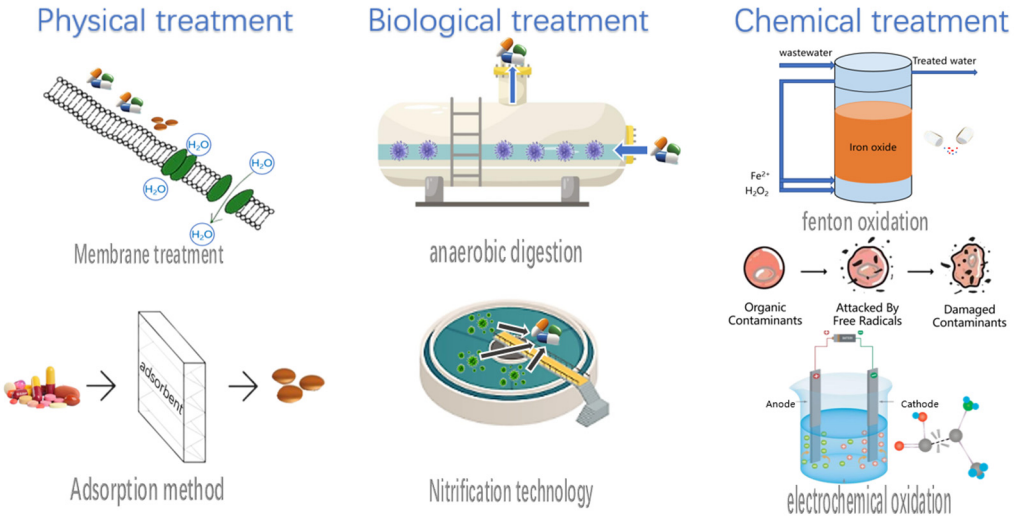

Figure 5. Model diagram of common urine treatment technology.

In the process of waste degradation, there may be by-products, some of which will reduce the degradation efficiency and some of which are more effective than primitive molecules. During the electrochemical treatment of urine, chloride ions (Cl^-) may be produced. Although chloride ions can play a great role in ammonia nitrogen removal and the oxidation of organic matter, they are prone to produce highly toxic organochlorine disinfection by-products such as chlorate and perchlorate in the actual oxidation process [93]. Therefore, Radjenovic et al. [94] suggest that there is a critical value of activated chlorine in wastewater treatment that produces the minimum amount of toxic substances with the best treatment effect. At present, Wang et al. [95] have achieved the regulation of the generation and removal of DBPs (disinfectant byproducts) during electrochemical oxidation of urine treatment, providing technical support for the safe and efficient application of electrochemical treatment of urine. In some cases, the by-products produced during the treatment of urine can improve its degradation efficiency. For example, in the study of sonochemical treatment of urine, Liu et al. [92] proposed that the inorganic ions generated by the hydrolysis of urine push the target pollutants towards the boundary layer of the cavitation bubbles, which strengthens the degradation of pollutants by the hydroxyl radicals generated by the acoustic wave.

5.2.4. Electrochemical Advanced Oxidation

Some advanced oxidation processes (AOPs), which are considered promising technologies for treating emerging contaminants, have been widely studied for their pharmaceutical removal capacities and limitations. For example, the Fenton advanced oxidation process and the photocatalytic degradation process can effectively degrade ibuprofen [96,97]. However, slow kinetics and the presence of organic matter lead to slow performance degradation; thus, more stringent operating conditions (e.g., oxygen supply and pH) need to be designed, and toxic intermediates may also be produced [98,99]. Tang et al. [100] studied the photocatalytic degradation of norfloxacin and found that, in practical application, further filtration and treatment of the catalyst were needed, which caused the difficulty and uncertainty of the experiment. In addition, due to the ability of anions to capture hydroxyl radicals and their inevitable existence in wastewater, they surely pose a negative impact on the role of AOPs [101]. In the past few decades, photocatalytic oxidation has been one of the more focused AOP technologies and has been successfully applied to the degradation of pharmaceutical contaminants in urine. There is a lack of research on the development of catalysts working in the visible light region, which leads to a lack of definite understanding of the degradation mechanism, reaction efficiency, and operability of practical application processes under different light sources [102]. Electrochemical water treatment technology is a green wastewater treatment technology. Compared with traditional water treatment

technology, it has the following three advantages: (1) Clean. Exogenous chemicals are usually rarely demanded. Electrochemical technology has a good treatment effect on drug pollutants and will not or rarely produce secondary pollution, so it is called a clean treatment method. [103] (2) Flexible. Electrode shape and size can be accurately controlled. The types of reactors are flexible, and there are no rigid requirements for water treatment sites. It can be used alone or in combination with other technologies. [104] (3) Controllable. If the sensor matches the performance of the monitoring system, it is easy to realize automatic control. [105] Further, the application of electrochemical technology in water treatment is still at the forefront. [105–107] However, electrochemical technology also has drawbacks, the biggest being its high cost. For wastewater with poor conductivity, it is necessary to add conductive salts. Therefore, in recent decades, researchers have been committed to the development of low-cost electrode materials.

Research Progress of Electrode Materials

In electrochemical systems, the choice of electrode materials and electrode preparation are important factors in determining the treatment efficiency and cost, where the choice of anode materials directly or indirectly affects the efficiency of organic removal [108]. The BDD (boron-doped diamond) electrode has become the most widely used electrochemical oxidation anode due to its excellent performance. Özcan et al. emphasized the use of BDD electrodes in the study of the electrochemical degradation of norfloxacin. However, the high cost also limits its large-scale application in wastewater treatment [109]. How to reduce the cost is crucial for the practical application of BDD electrodes.

Mixed metal oxide electrodes, also known as DSA (boron-doped diamond) electrodes, are electrode materials with electrochemical stability that not only produce large amounts of active oxidants but also help to reduce costs [110]. DSA electrodes have been reported in many studies on the degradation of pharmaceuticals in urine due to their stability (Table 4). Isabelle et al. [111] used an MMO/Ti/RuO_2IrO_2 electrode to degrade mixed pharmaceuticals in urine wastewater, which showed that the photo-electrolysis was more effective in removing the pharmaceutical compared to single electrolysis, further verifying that electrochemistry can be coupled with other techniques to enhance the degradation ability. Sindy et al. [112] investigated the electrochemical degradation of norfloxacin (NOR) in urine on a Ti/IrO_2 anode, during which it was found that fresh urine containing large amounts of urea took more time to degrade norfloxacin.

Table 4. Treatment of different pharmaceutical contaminants in urine.

Pharmaceuticals	Processing Technology	Treatment Effect	Urine Type	Reference
Ibuprofen, ephedrine and propranolol	ZnO nanoparticles for chemical coagulation	Removal rates all over 99%	Real urine	[78]
Norfloxacin	RH adsorption CH adsorption	Removal rates were 30.6% and 83.54%, respectively	Synthetic urine	[80]
Sulfonamides	Biochar/H_2O_2	Removal rates all over 80%	Hydrolysis of urine	[81]
Propranolol, ethinyl estradiol, ibuprofen, diclofenac, and carbamazepine	Nanofiltration Membrane	Fresh urine: drug retention > 92% Synthetic urine: drug retention > 73%	Fresh urine/ synthetic urine	[82]
Diclofenac	Ion exchange resin	Removal rate over 90%	Synthetic urine	[83]
11 pharmaceuticals including carbamazepine and metoprolol	Nitrification + Adsorption	Removal rate of 90%	Synthetic urine	[85]
Chloramphenicol	Photodissolution	Chloramphenicol fully mineralized	Synthetic urine	[88]

Table 4. *Cont.*

Pharmaceuticals	Processing Technology	Treatment Effect	Urine Type	Reference
Clozaril	Acoustic Chemistry/ UVC/H_2O_2	Removal rate of 90%	Synthetic urine	[89]
Ampicillin	Acoustic Chemistry	Removal rate of 92%	Synthetic urine	[90]
Penicillin G, Meropenem and Chloramphenicol	Electrolysis/ Light-Electrolysis	Removal rate of > 70/82–100%	Synthetic urine	[106]
Norfloxacin	Electrolysis	Removal rate up to 100%	Synthetic urine	[107]
Ibuprofen	Electrolysis	Fully mineralized	Synthetic urine	[110]

Removal rate (%): $\frac{c_0-c}{c_0} \times 100\%$, C_0: initial concentration, C: post-reaction concentration.

PbO_2 is another type of non-reactive anode material that has been often studied because of its good electrical conductivity, low cost, and high oxygen precipitation capacity. However, its lifetime is short, and Pb^{2+} will be released into solution [113]. It is usually doped with some rare-earth elements to improve its stability and catalytic activity [114]. Wang et al. [115] used Ti/SnO_2-Sb/Ce-PbO_2 to degrade ibuprofen, formed carboxylic acid through a series of hydroxylation, decarboxylation, and benzene ring cleavage reactions, and then oxidized the carboxylic acid to H_2O and CO_2 through successive hydroxylation to achieve complete mineralization of IBP. Zhou et al. [116] used a rare-earth-doped Ti/SnO_2eSb/PbO_2 anode to degrade triclosan in human urine for the first time. The results showed that the removal rate of triclosan reached 90%, and the quantitative structure-activity relationship model also verified its potential risk to aquatic organisms.

The integration of cathodic hydrogen production into electrochemical purification cells is very promising in order to further reduce the cost of electrooxidation. In addition, the combination of electrochemical and photochemical processes is also a research hotspot in the simultaneous degradation of organic pollutants and power generation [117]. At present, self-powered photochemical cells have been developed, which can effectively reduce the supply of external power sources and reduce treatment costs and maintenance. Another option to compensate for the cost is to transform the biochemical properties of refractory organic compounds by partial electro-oxidation and then feed the wastewater to MFCs for power generation [118].

Table 5 shows the research examples of electrochemical oxidation (EO) in urine. The researchers focus on the selection of electrode materials, the preparation process, optimal operating conditions, and economic costs to improve the wastewater treatment capacity of electrochemical oxidation technology.

Table 5. Electrochemical treatment of urine wastewater containing pharmaceutical contaminants.

Anode Type	Processing Objects	Operating Conditions	Main Results	Energy Consumption Analysis	Reference
Ti/SnO_2eSb/PbO_2	Simulated urine wastewater containing 5 mg/L triclosan	Electrode spacing: 10 mm Current density: 10 mA/cm^2	Triclosan removal rate: 90%	Ton of water power consumption: 4.5~47.8 kWh	[116]
Ti/Ru0.3Ti0.7O_2	Simulated urine wastewater containing 200 mg/L tetracycline	Electrode spacing: 6 mm Current density: 10–40 mA/cm^2 Electrolysis time: 3 h	Tetracycline removal rate: 50%	Electricity consumption per ton of water: 2.85–4.1 kWh	[119]
Ti/RuO_2-IrO_2	Simulated urine wastewater containing cephalexin	Current density: 6 mA/cm^2 Electrolysis time: 2 h	Degradation rate of ciprofloxacin: 80%	Electricity consumption per ton of water: 0.088 kWh	[120]
Nanocrystalline Diamond (NCD)	simulated urine wastewater containing 15 mg/L ciprofloxacin	Current density: 40 mA/cm^2 Electrolysis time: 60 min Temperature: 25 °C	Degradation rate of ciprofloxacin: 90.4%	Electricity consumption per ton of water: 22.9 kWh	[121]

Table 5. *Cont.*

Anode Type	Processing Objects	Operating Conditions	Main Results	Energy Consumption Analysis	Reference
Ag/AgCl/KCl	Simulated urine wastewater containing 10 μM-1 mM cefazolin	Current density: 0.5–150 mA/cm^2 Electrolysis time: 0–500 min	Current density: 150 mA/cm^2, electrolysis: 20 min, cefazolin residue < 0.5‰	Maximum power consumption of 3.7 kWh per ton of water	[122]
MMO-RuO_2-IrO_2	Simulated urine wastewater containing 50 mg/L penicillin G	Current density: 30 mA/cm^2 Electrolysis time: 2 h	Penicillin G removal rate ≥ 99%	SEC: 0.5 kWh (% inhibition)-1	[123]
Sb-Sn-Ta-Ir/Ti	Simulated urine wastewater containing 50 mg/L uric acid	Electrode spacing: about 20 mm Current density: 7.46 mA/cm^2 Electrolysis time: 42.79 min	COD removal rate: 92% TOC removal rate: 89%	Electricity consumption per ton of water: 2.479 kWh	[124]
100–8000 ppm BDD anode	Simulated urine wastewater containing 50 mg/L penicillin G	Current density: 30 mA/cm^2 Electrolysis time: 8 h Charge through: 6.4 Ah dm^{-3}	200 ppm-BDD has the best effect, a 100% removal rate of penicillin G, and a 90% reduction of toxicity	SEC: 0.15 kWh (% inhibition)-1	[125]

Parra et al. [119] reported the degradation of 200 mg/L of the antibiotic tetracycline in the urine matrix using a Ti/Ru0.3Ti0.7O_2 anode. The EO process with DSA was considered the most efficient treatment, obtaining a 50% removal rate after 3 h of electrolysis. Power consumption per ton of water is 2.85–4.1 kWh, and the consumption increased along with the hydrolysis of urine. Similar results were reported by Fabrizio et al. [122] during electrolysis of urine solutions containing cefazolin; the antibiotic is basically removed after 20 min of treatment at 150 mA/cm^2, and the max power consumption per ton of water is 2.85–4.1 kWh. Different electrodes, different operating conditions, and processing objects lead to significant differences in degradation efficiency and energy consumption. It can be seen from Table 5 that the energy consumption of electrochemical oxidation with a titanium-based electrode is generally low, while that with a diamond electrode is high. Therefore, the future application of DSA electrodes is promising.

Prospects for the Development of Electrochemical Technology

In the future, the combination of electrochemistry and other processes will be a promising trend in urine treatment. Membrane treatment technology has been proven to be able to recover nutrients in urine, but membrane fouling has become an important factor limiting its large-scale application [126]. Combining electrochemical technology with membrane treatment processes mitigates membrane fouling through electrostatic repulsion, electrochemical degradation reactions of contaminants, and electrophoretic movement of ionic components. Sun et al. [104] reported that electrochemically coupled membrane treatment processes to recover nutrients, water, and other resources from urine have wide promise. In addition, artificial intelligence technology has now rapidly developed, indirectly promoting the development of electrochemical technology. Using the latest advances in intelligent technologies such as sensors and communication technologies, researchers are committed to developing intelligent control models and promoting their application for the on-site treatment of source-separated urine [127]. The goal of precisely controlling the electrochemical reaction process while achieving maximum treatment efficiency will be achieved.

With the continuous progress of electrochemical technology, the industrialization process of its application in urine treatment has also accelerated. Yixing Eco-sanitary Manufacture Co., Ltd., China, introduced electro-catalytic oxidation treatment technology for source-separated urine developed by Professor Hoffmann's team at Caltech to develop an eco-toilet. The clean water was generated to flush the toilet. The solar power was used in the eco-toilet. At present, the continuous circulation and self-sufficiency of power and water have been realized in the eco-toilet, which has been applied in Yixing, Hong Kong, South Africa, etc.

6. Conclusions

Urine, as a special source of pollution, can also be a certain resource. How to properly treat it has become a hot research topic in recent years. This paper overviewed the sources, metabolic pathways, and pollution status of pharmaceutical contaminants and the research progress of urine treatment, then summarized the following conclusions:

(1) The vast majority of pharmaceuticals are excreted in the urine, and these pharmaceuticals enter the wastewater treatment plant with the domestic wastewater, so the wastewater treatment plant is the main source of pharmaceutical contaminants in the environment;
(2) The results of the research show that pharmaceutical concentrations in urine are typically 2–3 orders of magnitude higher than those in municipal wastewater treatment plants and that pharmaceutical concentrations in wastewater treatment plants and surface water are generally at ng/L levels, posing potential risks to humans and the ecological environment;
(3) Compared to physical and biological methods, the advanced electrochemical oxidation method is more effective and promising in treating pharmaceutical contaminants in urine. This technology is now maturing, but the cost is still too high, and in the future, it needs to be considered for coupling with other technologies to further reduce costs.

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