Author(s): Imwene, K.O.; Ngumba, E.; Kairigo, P.K.

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Review

Emerging technologies for enhanced removal of residual antibiotics from source-separated urine and wastewaters: A review

K.O. Imwene[a], E. Ngumba[b], P.K. Kairigo[c,*]

[a] University of Nairobi, Faculty of Science and Technology, Department of Chemistry, PO Box 30197, 00100, Nairobi, Kenya
[b] Jomo Kenyatta University of Agriculture and Technology, Department of Chemistry, P.O. Box 62000-00200, Nairobi, Kenya
[c] University of Jyvaskyla, Department of Biological and Environmental Science, P.O. Box 35, FI-40014, University of Jyvaskyla, Finland

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ABSTRACT

Antibiotic residues are of significant concern in the ecosystem because of their capacity to mediate antibiotic resistance development among environmental microbes. This paper reviews recent technologies for the abatement of antibiotics from human urine and wastewaters. Antibiotics are widely distributed in the aquatic environment as a result of the discharge of municipal sewage. Their existence is a cause for worry due to the potential ecological impact (for instance, antibiotic resistance) on bacteria in the background. Numerous contaminants that enter wastewater treatment facilities and the aquatic environment, as a result, go undetected. Sludge can act as a medium for some chemicals to concentrate while being treated as wastewater. The most sewage sludge that has undergone treatment is spread on agricultural land without being properly checked for pollutants. The fate of antibiotic residues in soils is hence poorly understood. The idea of the Separation of urine at the source has recently been propagated as a measure to control the flow of pharmaceutical residues into centralized wastewater treatment plants (WWTPs). With the ever increasing acceptance of urine source separation practices, visibility and awareness on dedicated treatement technologies is needed. Human urine, as well as conventional WWTPs, are point sources of pharmaceutical micropollutants contributing to the ubiquitous detection of pharmaceutical residues in the receiving water bodies. Focused post-treatment of source-separated urine includes distillation and nitrification, ammonia stripping, and adsorption processes. Other reviewed methods include physical and biological treatment methods, advanced oxidation processes, and a host of combination treatment methods. All these are aimed at ensuring minimized risk products are returned to the environment.

1. Introduction

Pharmaceutical and personal care products (PPCPs) are widely used in a variety of industries and fields, including medicine, business, agriculture, aquaculture, and people's daily lives. This contributes to the ubiquitous nature of PPCPs in the environment (Kümmerer, 2009). The advancements in analytical technology allows for the detection of PPCPs at trace levels in environmental samples (Ngumba et al., 2016). There are direct and indirect ways by which PPCPs enter the environment. (Wang and Wang, 2016). In a nutshell, PPCPs can enter surface water through direct discharge from industries, hospitals, homes, and wastewater treatment facilities. They can also enter through surface runoff when biosolids are spread on agricultural land and eventually reach groundwater through leaching or bank filtration. Since sediment have a range of binding sites, they can adsorb the PPCPs in the surface water compartment (Kastner et al., 2014). Antibiotics are used in various settings, including drugs for humans, animals, and agriculture (Cetecioglu et al., 2016; Kümmerer, 2009). Due to their misuse and weak digestive systems, they are discharged in significant amounts into wastewater (Huang et al., 2015a; Rodriguez-Mozaz et al., 2015). These drugs can then reach waterways through different routes, including treatment plant discharge, fish hatcheries, livestock feeding activity, and surface overflow (Rodriguez-Mozaz et al., 2015; Storteboom et al., 2010). Antibiotics and their metabolites found in naturally occurring ecosystems pose a significant risk to human and environmental health, even in small amounts (Kümmerer, 2009; Wang et al., 2016). Bacteria develop antibiotic resistance genes (ARGs) induced by antibiotics (Fernandes et al., 2015; Yan et al., 2019). According to the (WHO) (O'Neil, 2014), Antibiotic-Resistant Bacteria (ARB) are among the most important public health threats. Wastewater treatment plants have already

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* Corresponding author.
E-mail address: Pius.k.kairigo@jyu.fi (P.K. Kairigo).

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been identified as hotspots for developing ARB and ARGs (Guo et al., 2018a; Neudorf et al., 2017; Bengtsson-Palme et al., 2018). The Horizontal Gene Transfer of the ARGs among bacteria is hypothesized to be facilitated by antibiotic residues in activated sludge (Kim et al., 2014). Due to their lethality in microbial communities (Kummerer et al., 2004; Srinivasan and Sarmah, 2014). In public health and worldwide, the use of antibiotics and pharmaceuticals improved people's lives (Pruden et al., 2012). Despite this, the pharmaceutical sector may produce high-concentration antibiotic effluent released into the environment (Patneedi and Prasadu, 2015). As antibiotic-resistant infections, including superbugs, are becoming more common, worries regarding antibiotics, antimicrobial resistance genes, and antimicrobial resistance bacteria (ARGs and ARBs) in aquatic environments are developing (Heß et al., 2018; Kairigo et al., 2020). According to reports, the percentage of ARB in animals such as pigs, fowl, and cattle has significantly increased, particularly in low- and middle-income nations (Van Boeckel et al., 2019). Modern analytical techniques in chemistry and molecular microbiology have made it possible to identify a wide range of antibiotics and ARGs. Multiresidue antibiotics that are present in waters at trace concentrations of ng to g/L can be found using solid phase extraction (SPE) and liquid chromatography coupled with mass spectrometry (LC-MS), tandem mass spectrometry (LC-MS/MS), or time of flight mass spectrometry (LC -TOF -MS) (Na et al., 2011; Ngumba et al., 2016). It is now possible to determine the prevalence of several ARGs thanks to the invention and use of quantitative PCR (qPCR), droplet digital PCR (ddPCR), epic PCR, DNA microarray, metagenome, and Smart-Chip techniques (Liu et al., 2019). For example, effluent from an Indian wastewater treatment plant, which gathers wastewater from 90 drug processing industries, contained a significant amount of ciprofloxacin (31 mg/L) (Larsson et al., 2007). Antibiotic resistance genes (ARG) were reported to be significantly higher (2.36107 copies/mL) in pharmaceutical company effluent receiving wastewater than in municipal wastewater (9.50105 copies/mL) (Guo et al., 2018b). This shows that existing wastewater treatment plant (WWTP) designs utilized by the pharmaceutical industry, hospitals, and municipal sewage treatment plants fail to eliminate antibiotic residues from wastewater (Hou et al., 2019). Furthermore, depending on the molecule, urinary excretion of ingested antibiotics as parent compound or active metabolite ranges from 5% to over 70% (Jjemba, 2006).

Fig. 1. Antibiotics enter the bio-sphere via multiple routes once excreted by humans and animals.

1.1. *Regulations and procedures for hospital wastewater (HWW)*

While states and international organizations have their wastewater treatment recommendations, the WHO's rules for pretreatment of healthcare facility effluents are the most commonly followed (Verlicchi, 2018). Discharged wastewater may contain pharmaceuticals, chemicals, transmissible infections, or radioactive waste (Leal et al., 2010). The WHO recommendations (Chartier et al., 2014; Yan et al., 2020) provide a framework for discussing the harmful properties of these wastewaters and providing a safe HWW management approach. According to European Directive n. 98 of November 19, 2008 (E.U., 2008/98/E.C.), some hospital effluents (i.e., pharmaceuticals and personal care products, or PPCPs) cannot be discharged into sewer systems. They must be treated decentrally before being released into centralized wastewater systems (Carraro et al., 2017).

1.2. *Antibiotic concentrations in urine separated at the source*

The primary excretion route for the majority of consumed medication is through urine as either the parent compound or transformational products. Table 1 shows the percentage excretion rates of selected antibiotics as unchanged compounds in urine. Studies on the determination of antibiotics on source-separated urine are limited. The concentration of antibiotic residues is expected to be much higher in the urine compared to effluent wastewater and surface waters. Sulfamethoxazole (SMX), for instance, is one of the sulfonamide antibiotics that has been widely used to treat bacterial infections in humans, including prostatitis, bronchitis, and urinary tract infections. SMX is frequently used in animal husbandry and the aquaculture industry to treat bacterial infections since it is efficient against both gram-negative and positive bacteria (Wang et al., 2019). By blocking the dihydropteroate synthase, the drug prevents the production of dihydropteroic acid, which further restricts bacterial growth (Bhattacharjee, 2016). A recent study that investigated the concentration of antibiotics and antiretroviral drugs in source-separated urine reported concentration values in mg/L levels with Trimethoprim (12.8 mg/L) and Sulfamethoxazole (7.74 mg/L)

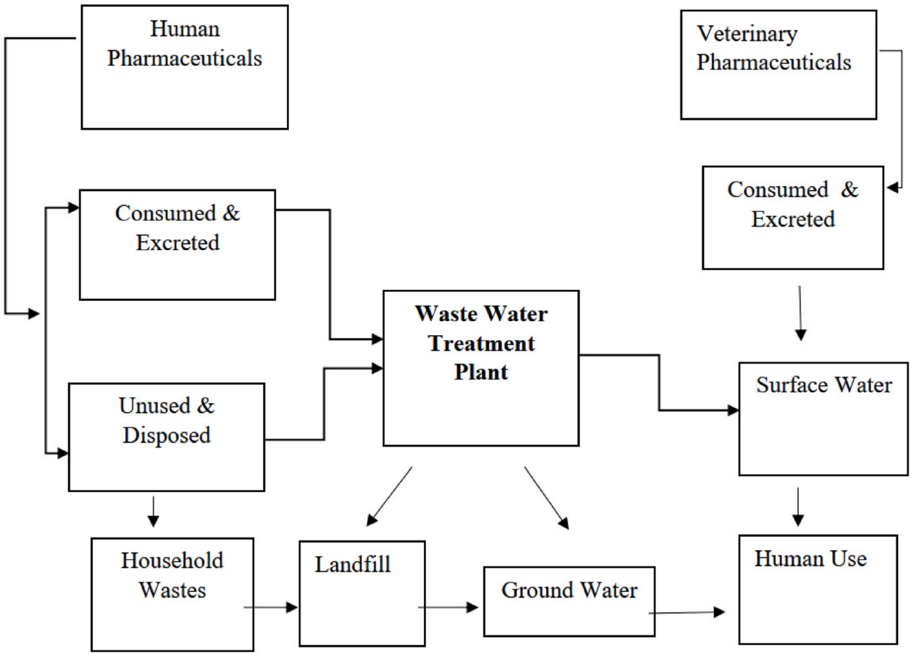

Fig. 1. Sources and distribution of pharmaceuticals in the environment.

Table 1
The following table shows the percentage excretion rates of selected antibiotics as unchanged compounds in urine.

Pharmaceutical compound	Excretion rate as unchanged Compound (%)	Reference
NOR	60	(Ngumba et al., 2020; Bartlett, 2004)
TMP	80–90	(Ngumba et al., 2016, 2020)
AMO	60–80	(Ngumba et al., 2020; Wood et al., 2015)
CIP	80	(Ngumba et al., 2020; K'oreje et al., 2016)
SMX	15–25	(Ngumba et al., 2020; Madikizela et al., 2017; Kimosop et al., 2016)
TET	80–90	Yang et al. (2005)
DOX	70	Nourse et al. (2004)
LEV	85	Yasojima et al. (2006)
AZI	70–80	Pan and Yau (2021)
MET	13–58	Song et al. (2013)

TMP = trimethoprim, SMX = Sulfamethoxazole. NOR = norfloxacin, CIP = ciprofloxacin, CIP = ciprofloxacin, CIP = ciprofloxacin, TET = tetracycline, DOX = Doxycycline, AMO = amoxicillin, LEV = levofloxacin, MET = metronidazole, and AZI = azithromycin.

recording the highest concentrations (Muriuki et al., 2020; Ngumba et al., 2020). These antimicrobials are administered as prophylaxis for HIV/AIDS patients to prevent opportunistic infections.

1.3. *Antibiotic residues in wastewater and surface waterways*

The majority of residual antibiotics in the ecosystem are due to the treated wastewater discharge from point sources such as pharmaceutical industry effluent, hospital effluent, municipal WWTP effluent, industrial farming, and untreated waste discharged directly into water bodies from informal settlements. Secondary processes that are most commonly used and explored include;wastewater stabilization ponds, activated sludge, and trickling filters. On the other hand, these approaches do not eliminate residual antibiotics, releasing them into receiving water bodies (Kairigo et al., 2020; Muriuki et al., 2020). Using a United Kingdom data set as a baseline, the current understanding of residual antibiotic contamination in wastewaters and surface waters for a European country is described. The most studied drugs are nonsteroidal anti-inflammatory drugs (NSAIDs), b-blockers, antidepressants, and antiepileptic carbamazepine. These are widely prescribed (>1000 kg per year) and typically discovered in influent wastewaters. Removal rates range from low (50%) to high (>80%) due to their variable physicochemical properties and vulnerability to biological attacks. Pharmaceuticals effluent in surface waters was reported in ng/L to mg/L levels (Simu et al., 2020). This review aims to provide insights on emerging technologies for enhanced removal of residual antibiotics from source-separated urine and wastewaters. The advantages and disadvantages of selected contemporary antibiotics wastewater cleanup technologies is outlined in Table 2

1.4. *Present and emerging techniques for wastewater treatment and their potential for antibiotic removal*

Wastewater treatment is classified into three broad categories; biological, chemical, and physical. The purpose of wastewater treatment is to make it safe for the intended use. The various treatment processes in a typical WWTP is illustrated in Fig. 2.

1.5. *Biological treatment*

In biological treatment, antibiotics are usually eliminated through sludge adsorption and biodegradation (Zheng et al., 2017). When it

Table 2
The benefits and drawbacks of contemporary antibiotics wastewater cleanup technology (Wang and Wang, 2019).

Method	Merit	Demerit
1. Fenton (Using Fe ions and H_2O_2, destructive procedures are used).	• Reactive radicals, such as hydroxide ions, are produced in situ. • There is no sludge generation and no mineralization of organic pollutants. • For recalcitrant chemicals, rapid breakdown and efficiency are essential.	• Unknown by-products are formed, necessitating further inquiry and investigation. • Laboratory scale. • Technical limitations. • A substantial volume of ferrous sludge is generated.
2. Ozonation process	• With this method, antibiotics can be broken down in as little as 240–300 s.	
3. Electrochemical oxidation process	• Method is free of sludge. • Electrochemical Oxidation is not like Fenton technology; depending on the produced oxidant by the electrochemical system, it can be both unselective and selective (Bhuta, 2014).	• This is a capital-intensive method in comparison to other technologies like Fenton. Since different electrodes have differing capacities for producing oxidants, selecting the right electrodes is important for successful electro-oxidation procedures (Bhuta, 2014).
4. Adsorption (A non-destructive method of removing contaminants from an aqueous medium by utilizing a solid substance.)	• A process with great efficiency and rapid kinetics. • Separation of a wide spectrum of contaminants is a strong suit (heavy metal & organic pollutants). • With minimal equipment, it can adapt to various therapy formats. • The treated effluent is of good quality.	• Non-destructive and non-selective techniques. • Regeneration has a high cost and results in loss of material. • The treatment procedure can be altered by changing the pH value. • After wastewater treatment, an additional adsorbent step, such as incineration or regeneration, is required. • Energy requirements are high, and flow rates are limited.
5. Anaerobic treatment	• Reactors are smaller. • As a by-product, methane is produced. • The biomass yield is lower.	• Energy consumption is high. • Operating costs are high. • Low rate of loading • Antibiotics are not well digested by this organism. For greater elimination efficiency, a combination with aerobic treatment is required. • Energy consumption is high.
6. Aerobic treatment	• Increased loading speed. • Antibiotics are broken down more efficiently (Compared to anaerobic).	• Operating costs are extremely high. • Antibiotics are not well digested by this organism. For improved removal efficiency, anaerobic treatment is required
7. Photocatalysis (Using semiconductors and light, destroy organic	• Photocatalysts with good aqueous phase	• Photogenerated electron-hole recombination occurs quickly.

(*continued on next page*)

Table 2 (*continued*)

Method	Merit	Demerit
contaminating chemical structures).	stability, such as TiO2, ZnO, etc. • Non-toxic with high activity. • Photocatalyst recovery efficiency and recyclability. • Low cost, simple to use, and destroys the chemical structure of organic pollutants. • Sunlight, oxygen, and photocatalyst were all needed in large quantities.	• There is a limited response to visible light. • Poor treatment for organic contaminants with high concentrations. • The chemical structure and toxicity of the degraded by-product have not been investigated.
8. Electrocoagulation	• It can handle numerous pollutants in a single run. • Sludge production is minimal. • Low-cost maintenance. • Produces a TDS-lower effluent. • Systems easily handle water quality fluctuations.	• Electrodes have a finite lifespan. • There is a need for active fine-tuning.

Source (Wang and Wang, 2019).

comes to natural medicines, the amount of oxygen they require might be characterized as either aerobic or anaerobic (or both). The most popular aerobic technique is a biological aerated filter system. The most frequent anaerobic processes are Anaerobic Digestion (AD.), Anaerobic Filter (AF), Up-flow Anaerobic Sludge Blanket (UASB) and Anaerobic Baffled Reactor (ABR). The sequencing batch reactor (SBR) and membrane bioreactor (MBR) technologies are frequently used with aerobic and anaerobic approaches. The most popular methods for removing antibiotics from breeding effluent include biological aerated filters (BAF), AD., SBR, and MBR techniques. As existing technology is continously improving in merging many processes, antibiotics have been extracted more efficiently from WWTP effluent. Antibiotics removal from pig wastewater biofilm MBRs (BF-MBRs) has been compared to normal MBRs (Huang et al., 2016). According to experimental data, removing antibiotics can be done selectively using biological technology; nonetheless, the technique and surrounding circumstances limit removal efficiency. As a result, it has limitations in removal of antibiotics residues from breeding wastewater. More research to improve the effectiveness of biological approaches in removing antibiotics is needed (Cheng et al., 2016).

1.5.1. *Aerobic processes*

Aerobic wastewater treatment methods clean water by relying on oxygen-feeding bacteria, protozoa, and other specialized microbes as (opposed to anaerobic systems that do not need oxygen). These systems use the natural microbial decomposition process to break down and eliminate industrial wastewater contaminants. This includes but is not limited to Wastewater stabilization ponds, aerobic Bioreactors, activated Sludge systems, Rotating Biological Contactors, and Phytoremediation of wastewater (Constructed (built) Wetlands) (Sahota and Sharma, 2020).

1.5.1.1. Wastewater stabilization ponds. A secondary wastewater treatment system, such as a water stabilization pond, an oxidation pond, or a lagoon, treats waste or sewage from industries, residential areas, and other sources. Ponds are built to improve natural still water ponds (Gruchlik et al., 2018). They can be anaerobic (influences microorganisms' growth that needs oxygen, like algae and bacteria). Aerobic exercise encourages the development of microorganisms that have or need oxygen, like algae and bacteria). Historically, the primary aim of ponds was to provide holding time for wastewater so that natural processes could settle it (Ensink et al., 2007).

1.5.1.2. Aerobic bioreactor. In terms of antimicrobial wastewater treatment, bioreactor systems (B.Rs) are thought to outperform typical activated sludge treatments (CAS) (Sahar et al., 2011; Nguyen et al., 2012). In B.R.s, sludge retention time (SRT) and flocculation size might increase biodegradation potential and sorption capacity (Fernandez-Fontaina et al., 2012; Helbling et al., 2012). Two of the most prevalent methods for removing antibiotics from wastewater during biological treatment are sorption and biodegradation (Cheng et al., 2018).

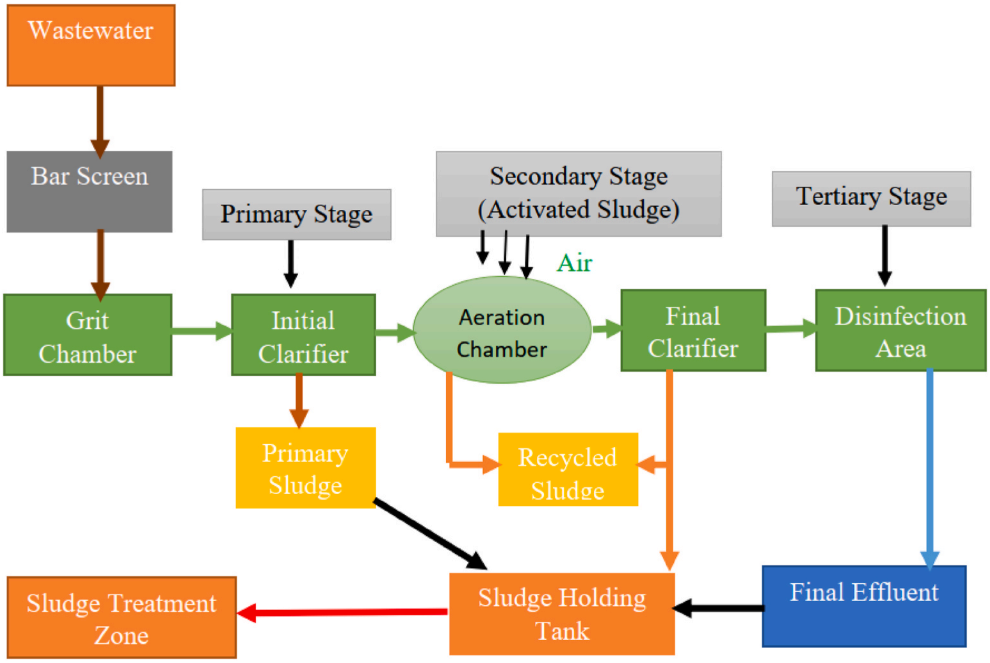

Fig. 2. Various treatment processes in a typical WWTP (Khan et al., 2021).

Furthermore, previous research found that bio carrier's attached-growth processes favored the enrichment of slow-growing bacteria (Leyva-Díaz et al., 2016), boosting the biodegrading antibiotic potential of MBRs.

1.5.1.3. Activated sludge. The activated sludge approach has been widely used in industrial and domestic wastewater treatment because of its wide microbial variety and activity, removing most organic pollutants and nutrients (Wagner and Loy, 2002). According to studies, the composition and variety of the microbial population indicated the greatest impact on the stability and performance of wastewater treatment systems (Miura et al., 2007). The biological community of activated sludge comprises viruses, bacteria, protozoa, fungus, algae, and metazoans, with a wide range of natural variability. Bacteria make up 95 percent of all microorganisms in this complex ecosystem, and they play a critical role in wastewater treatment (Jenkins, 1993).

1.5.1.4. Rotating Biological Contactors. Rotating biological contactors (RBC) use disc, surface, media, and biofilm reactors to provide an alternative to the activated sludge (AS) process. A solid medium in the RBC favors microbial development in a static biofilm (Singh and Mittal, 2012). RBCs are wastewater treatment system that offers quality effluent at acceptable levels and excellent removal efficiency of organics at a reasonable cost. It is appealing because of its ease of use, low sludge generation, small footprint, low maintenance, and low operating costs. Variable disk submergence levels and operation under anaerobic and anoxic conditions in a single unit can result in complete nitrification/denitrification (Cortez et al., 2008).

1.5.1.5. Phytoremediation of wastewater (constructed (built) wetlands). An artificial wetland-based wastewater treatment system mixes soil, plants, and bacteria to purify the swamp's wastewater. For instance, microbial breakdown and plant adsorption are water purification processes (Kumar et al., 2015). Built wetlands are easy to maintain and manage and cost-effective (Kumar et al., 2018). In a previous study, Sulfonamide (SMX.), tetracycline (TET), and quinolone (Q.N.) are all removed at high quantities from breeding wastewater in constructed wetlands (Logan and Rabaey, 2012; Logan, 2009; Hu et al., 2008a; Vymazal, 2005), with removal rates ranging from 49.43% to 85%, 69 percent to 100%, and 82 percent to 100%, respectively. In a different study, Enrofloxacin, oxytetracycline, and arsenic trioxide have all been found to be removed from marine aquaculture effluent using artificial wetlands (Wu et al., 2015). Free water surface flow constructed wetlands (FWS-CWs), vertical subsurface flow constructed wetlands (VSF-CWs), and horizontal subsurface flow constructed wetlands (HSF-CWs); are three forms of manufactured wetlands classified by water flow direction (HSF-CWs). Anaerobic and aerobic bacteria break down antibiotics in vertical subsurface flow wetlands with more extended hydraulic retention periods (HRT) (Choi et al., 2016). As a result, the VSF-CW is frequently used for sewage treatment. An artificial wetland's hydraulic loading rate (HLR) refers to how much wastewater it can treat per unit volume or area per day. The hydraulic retention time (HRT) reduces as the HLR rises. Antibiotic consumption can be calculated daily in a built wetland by multiplying the HLR by the initial antibiotic concentration. As a result, antibiotic clearance rates are affected by the HLR (Rahman et al., 2020; Huang et al., 2015b). Several antibiotics have different clearance efficiencies depending on HRT levels. According to (Liu et al., 2013; Huang et al., 2017), increasing the HRT from one to three days can improve the sulfamethoxazole removal rate in artificial wetlands with various antibiotic removal designs. The SMX elimination rate increased from 4% to 55, and 59% in integrated horizontal and vertical flow constructed wetlands, respectively. On the other hand, HRT affected ENR or florfenicol elimination. Due to structural changes, these three antibiotics require different elimination techniques. Bacteria are the principal decomposers of Sulfamethoxazole (SMX). Although florfenicol is stubborn and hard to remove from a traditionally built wetland, the fill material's adsorption is critical for its removal. Furthermore, pH influences antibiotic action, temperature influences microbial proliferation, and light affect plant development, indirectly influencing antibiotic removal effectiveness (Boto et al., 2016). The removal rates of TC., SM., and QN. in celery aquatic-based system in summer after 60 days of operation (72, 47, and 22%, respectively) were significantly higher than in a spinach aquatic-based system (33, 20, and 7%, respectively) (Zhang et al., 2018). However, antibiotic removal effectiveness in the winter did not differ significantly between the two submerged plant filter beds (Liu et al., 2016a).

1.5.2. *Anaerobic processes*

Anaerobic wastewater treatment processes transform organic pollutants into biogas, thereby eliminating them from the wastewater by functioning without oxygen.

1.6. *Microbial electrolysis cells (MECs) and Microbial Fuel Cells (MFCs)*

These are examples of bio-electrochemical systems (BES), emerging anaerobic biotechnologies that can speed up pollutant removal (Richardson et al., 2005; Larsson et al., 2007; Guo et al., 2018b). Microorganisms consume organic compounds and transfer electrons to the anode in Microbial Fuel Cells (Hou et al., 2019). MECs, unlike MFCs, use an electric-driven hydrogen evolution mechanism during the wastewater treatment process to produce a large output of hydrogen (Hu et al., 2008b). In recent years, MFC has been used in conjunction with traditional wastewater treatment methods like constructed wetland (C. W.) and membrane bioreactor (MBR) to achieve the best of both worlds. An innovative wastewater treatment method is biofuel cells combined with a constructed wetland system (Fang et al., 2015). Due to the benefits of simultaneous filtration and biological function, power production using an MFC included within a C.W. membrane bioreactor technology has been recognized as a viable wastewater treatment option (Li et al., 2017a). Combining MFC with MBR systems has several advantages, including improved discharge quality, fouling reduction, and significant energy savings (Su et al., 2013; Ijanu et al., 2020).

1.7. *Chemical treatment*

The chemical process of wastewater treatment are classified as primary, secondary or tertiary treatment technologies. Water treatment technologies include but not limited to; distillation, crystallization, evaporation, neutralization, solvent extraction, oxidation, coagulation, centrifugation, sedimentation, precipitation, electrolysis, electrodialysis, ion exchange, reverse osmosis and adsorption. Some of these methods are described below. (Gupta et al., 2012). The concepts, range of applications, speed, and economics of the many water treatment systems covered in this article are different from one another.

1.7.1. *Advanced oxidation processes (AOPs)*

AOPs are oxidation techniques that use intense oxidizing hydroxyl radicals (*OH) produced during reactions as the primary oxidant to break down and mineralize organic contaminants in water. The *OH decomposes organic pollutants into small organic molecules, that are easier to break down(CO_2 and H_2O) by breaking chemical bonds or using electron transfer, addition, and substitution mechanisms. Ozonation, electrochemical Oxidation, and Fenton reaction are techniques primarily used to degrade antibiotics in waste streams (Huang et al., 2021). These techniques have been effectively employed to remove or degrade harmful pollutants, or they have been applied as a pretreatment to change resistant pollutants into biodegradable molecules that can then be treated by traditional biological techniques. The production of reactive free radicals, of which the hydroxyl radical (*OH) is the most significant, determines the effectiveness of AOPs. It should be emphasized that all of the advanced oxidation processes are chain-reaction sequences, and once a radical is produced, they typically include the propagation of radical cycles (Wang and Xu, 2012).

1.7.1.1. Electrochemical oxidation. In water treatment, electrochemistry is a relatively new techniquebased on the theory that the reactant loses electrons oxidized in the anode during the electrochemical reaction (Hakizimana et al., 2017). On the other hand, the reactant in the cathode loses electrons and becomes reduced. Refractory organic compounds are generally eliminated when the anode is oxidized. Electrolytic recovery, electrochemical Oxidation, electrolytic air flotation, Electrodialysis, and micro-electrolysis are the most often used electrochemical wastewater treatment techniques (Anglada et al., 2009). The electrochemical method, often known as the "Environmentally Friendly" method, provides a significant benefit compared to other ways. For example, the electrochemical process is usually performed at room temperature and pressure and is extremely efficient. These approaches can be used on their own or in combination with others. It has a small environmental impact, produces no secondary emissions, and is relatively mechanized. In the future, anode and electrochemical reactor research will be crucial (Hui and Jian Long, 1999).

1.7.1.2. Ozonation or catalytic ozonation. Ozonation or catalytic ozonation is an environmentally-friendly technology for wastewater treatment. Ozone has long been considered a powerful disinfectant and oxidant. Ozone is predominantly an oxidant in acidic environments. However, it mainly relies on free radical reactions in neutral and alkaline environments. Ozone has a much higher oxidation capacity than other typical oxidants but with several setbacks: high capital cost, high electricity consumption, more complex procedure, dangerous reactivity, and toxicity (Yasar and Tabinda, 2010). This process, which includes homogeneous and heterogeneous catalytic ozonation, can be used to improve the degradation efficiency of organic pollutants. In the homogeneous catalytic ozonation process, liquid catalysts, particularly transition metal ions such as Fe^{2+}, Mn^{2+}, Ni^{2+}, Co^{2+}, Cd^{2+}, Cu^{2+}, Ag^{+}, Cr^{3+}, and Zn^{2+} are used in the reaction solution. These catalysts can excite ozone, resulting in the formation of hydroxyl radicals (·OH) and improving degradation efficiency. Solid catalysts such as metal oxide, activated carbon, porous materials, and their composite materials are used in the heterogeneous catalytic ozonation process (Wang and Chen, 2019).

1.7.1.3. Fenton oxidation. In traditional Fenton/Fenton-like processes, H_2O_2 is typically added via bulk feeding, which may be the cause of H_2O_2's low utilization efficiency. By using Fenton/Fenton-like catalysts, the in-situ produced H_2O_2 from the activation of O_2 might be broken down to form a •OH radical in this unique Fenton/Fenton-like process. Emerging pollutants are degraded in part through the catalytic activation of O_2 and the breakdown of in-situ produced H_2O_2 (Liu et al., 2020). First utilized the Fenton system in organic synthesis but progressively used it for industrial wastewater treatment as people's knowledge grew. The Fenton reaction can be carried out at normal temperature and pressure, although it has significant environmental consequences, e.g., removal of essential heavy metals and the production of $Fe(OH)_3$ sludge which requires further separation and proper disposal technique making it expensive (Joseph et al., 2006). It's a sophisticated oxidation method that's easy to use and has mild reaction conditions, which cannot overlook the Fenton reaction's flaws. However, one cannot ignore Fenton's reaction flaws which include oxidant loss as a result of free radical scavenging and H_2O_2 breakdown. The other is the formation of iron mud in neutral conditions, which is difficult to treat (Bokare and Choi, 2012).

1.7.1.4. Photocatalytic oxidation. Ultraviolet photocatalytic Oxidation is a type of Oxidation that occurs when a substance is exposed to UV light. When oxidants undergo oxidative decomposition in the presence of ultraviolet light, free radicals develop a more robust oxidative capacity, allowing them to oxidize more difficult-to-decompose organic pollutants using only oxidants. Depending on the oxidants utilized, photocatalytic Oxidation can be classified into various categories, which for example include UV/O_3, UV/H_2O, $UV/H_2O_2/O_3$ and others. This review does not give description of the reaction steps. On the other hand, the impact of this approach on the treatment of refractory organic matter is undeniable (Pathak et al., 2017).

1.8. *Physical treatment*

The physical approach of wastewater treatment involves the removal of contaminants utilizing natural forces such as electrical attraction, forces of van der Waals, gravity, and physical barriers (Matilainen et al., 2010). Physical treatment of pollutant chemicals does not usually result in a change in their chemical structure. There are notable exceptions, such as when the physical condition of scattered compounds is altered, leading them to agglomerate, as seen in the filtering and vaporization phases. Physical methods include sedimentation, coagulation, membrane treatment, adsorption, distillation, and filtering (Kamaraj and Vasudevan, 2015).

1.8.1. *Electrocoagulation process*

Electrocoagulation (E.C.) technology is a wastewater treatment technique that uses electric current as the primary power source. In most cases, the power supply used to create an electric current in E.C. is either an alternating current (A.C.) power supply or a direct current (D.C.) power supply. Most research (Nasrullah et al., 2019; Balla et al., 2010) employ D. C, in the E.C. approach, they use electricity as their primary power source to generate an electric current, according to most specialists. Several trials have previously been conducted on this technology, demonstrating that E.C. is the best way for treating wastewater gram of E.C. reactor set up and treatment process (Hashim et al., 2020).

To create chemical aggregates and coagulate contaminants, coagulating agents like Fe^{3+} or Al^{3+} salts are applied to wastewater treatment. Following that, several coagulation technologies (Phoon et al., 2020) enable their removal from effluent. Charge shielding for contaminants diminishes electrostatic repulsion when energy blockade is required to build the aggregate fast (Homem and Santos, 2011). Electric current is used in dissolving Aluminium (Al) or iron (Fe) electrodes that have become absorbed in the effluent during the E.C. operation. Electronic suspension aids in the rise of Al^{3+} or Fe^{3+} ions in solution or OH^{-} Ions depend on pH, and the coagulants aid the separation of pollutants from wastewater (Ozyonar and Karagozoglu, 2014).

1.8.2. *Adsorption on biochars (treatment technologies for human urine)*

A carbon-rich substance known as biochar can be made from a variety of organic waste feedstock, including municipal sewage sludge and agricultural waste. The typical processes for producing biochar are pyrolysis, gasification, and hydrothermal carbonization. Due to its distinctive characteristics, including high carbon content and cation exchange capacity, wide specific surface area, and stable structure, biochar has drawn increased interest (Wang and Wang, 2019). Biochar has been utilized to recover nutrients from various waste streams, including human urine that has been isolated at the source. The rising use of medications has raised concerns since certain drugs are not entirely digested in human urine. Drugs may adsorb when biochar is used for resource recovery, posing a risk of pollutants entering the environment (Ling, 2019); therefore, the disposal mechanism of the used biochar is paramount. Incineration is touted as the most efficient way of disposal. This review study also explores the sorption of nutrients and medicines by biochar. It shows that various properties of biochars can be used to recover nitrogen and phosphate and remove drugs from source-separated urine (Ling, 2019). Many mechanisms for contaminant adsorption on biochar include pore-filling tools, electrostatic interactions, hydrophobicity, partitioning, and hydrogen bonding (Inyang and Dickenson, 2015). Biochar is a flexible adsorbent because of these many processes. It has a high capacity for eliminating radioactive elements (Jang et al., 2018) and specific organic contaminants from water and wastewater, including antibiotics (Solanki and Boyer, 2017), volatile organic compounds (Kumar et al., 2020), polychlorinated biphenyls (Xu et al., 2012), polycyclic aromatic hydrocarbons (Beesley et al., 2010), agrochemicals, and aromatic dyes (Qiu et al., 2009). Despite extensive research and reviews on biochar, there remains a knowledge gap on the use of biochar to separate drugs and nutrients found in urine. Biochar can eliminate up to 100% of pharmaceutical groups from human urine (Ahmed et al., 2015). If the biochar is solely selective for medications, the rectified urine can be delivered to the soil as a pharmaceutical-free nutritional product. A significant disadvantage of this technique is the disposal or handling of pharmaceutical-laden biochar. Pharmaceutical-contaminated biochar must be recycled or burned. The main driving forces for medicines' adsorption to biochar are intermolecular forces (van der Waals force and hydrogen bonding), all dominant at low solution pH (Solanki and Boyer, 2017; Zhang et al., 2019). On the other hand, nutrient adsorption is more common at higher solution pH.

1.8.3. *Membrane technology*

Membrane technology absorbs contaminants when wastewater passes through small membrane perforations. The most often utilized methods are microfiltration, ultrafiltration, nanofiltration, and reverse osmosis (Yu et al., 2011). The membrane technique has a high yield, is simple to use, and is inexpensive. Despite the shortage of research into using membrane technology to recover antibiotics in treatment facilities, antibiotics may have been extracted from various forms of wastewater—eliminated antibiotics at an 87 percent rate. Hence reducing the dissolved organic carbon by 40%, biodegradability rose 4.6-fold, and ecotoxicity was lowered by 58 percent when U.V. and nanofiltration were employed to treat wastewater in sewage treatment plants (Song et al., 2017). Additional toxins in the wastewater could be filtered using membrane technology. Using nanofiltration and reverse osmosis, ARGs, nitrogen, and phosphorus can successfully remove other pollutants from swine wastewater (Glaze et al., 1987). More studies are needed to investigate membrane technology to remove antibiotics from wastewater and source-separated urine.

1.9. *Sedimentation and coagulation*

Coagulation introduces chemical agents into wastewater, rapidly mixed and dispersed, transforming stable contaminants into dangerous and precipitable substances (Peydayesh et al., 2021). Coagulating is a complicated process. Squeezing and removing bound water around hydrophilic colloids is crucial for enhanced pharmaceutical wastewater treatment (Guo et al., 2017). As a result, the flocculant's nature is essential and linked to coagulation's effect. Flocculants are commonly made up of inorganic metal salts and polymers (Zhao et al., 2021). Suspended solids, chromaticity, and hazardous organic waste can all be removed with this technique (Yiping and Yu, 2010). It can also aid in faster biodegradation of pharmaceutical wastewater. After coagulation, the most frequent procedure is sedimentation. Pollutants, which have a higher density than wastewater, can be separated by gravity. Coagulation and sedimentation have advantages such as ease of use and proven technology; however, removing dissolved organic materials is difficult (Rubi et al., 2009).

2. A summary of a combined system for the elimination of antibiotics

Previous wastewater treatment techniques, i.e., stand-alone C.W. and MBR systems, are insufficient to repair the devastated environment. An MFC-linked electrochemical system, i.e., a Microbial Fuel Cell (MBR-MFC, CW-MFC-MEC, and CW-MFC), has been developed as a connected BES system for antibiotic removal (Rozas et al., 2010). For T.C. removal, a one-of-a-kind continuous flow MFC-sorption system was built. They discovered that a high T.C. concentration, a high electrolyte concentration, and a low pH value might all aid in increasing T.C. adsorption capability (Liu et al., 2014). Ideally, increasing the number of MFCs in series, which increases the current, could improve T.C. removal efficiency (Lan et al., 2019). However, rather than being removed, it was adsorbed, indicating additional therapeutic options should be explored. An MFC-PEC and MFC-EC systems were developed in another study by (Doherty et al., 2015) to degrade T.C. in the cathode chamber. Additionally, a new linked MBR-MFC system utilizing doped ($FeOOH/\ TiO_2$) filled activated carbon ($FeOOH/\ TiO_2$-GAC) was discovered to reduce membrane fouling while still removing over 90% of T.C. hydrochloride from the process. Reduction processes involving oxygen via the 2e pathway promoted hydrogen peroxide production (Cecconet et al., 2017).

2.1. *Antibiotic elimination using CW-MFCs*

As a unique technology, the ability of CW-MFC to remove antibiotics has received a lot of interest. T.C. and SMX were efficiently eliminated in CW-MFCs (Yang et al., 2015; Jiang et al., 2016), with T.C. outperforming SMX. Furthermore, Sulfadiazine (SDZ) concentration was lower than in the open approach (Li et al., 2017b). Several pharmacological and physical processes (Zhang et al., 2016; Li et al., 2018) that could be involved in antibiotic clearance in CW-MFCs were shown to be responsible for this decrease. Even though built wetland, the two microbial cells, that is, (electric cells and fuel cells) can promptly remove antibiotics, the risk of antibiotic resistance gene release has received little attention when these techniques are used (Zhang et al., 2017; Song et al., 2018). The MEC is an excellent antimicrobial pretreatment, and the effluent can be released into CW-MFCs for further biodegradation (Dordio and Carvalho, 2013). The connected system degrades significant amounts of SMX. For instance, MECs may be supported by coupled CW-MFCs for proper pretreatment of SMX (>85.73 percent), followed by CW-MFCs for additional SMX degradation (Huang et al., 2015c). With the stacked CW-MFCs providing a relatively constant power supply. These paired systems for removing antibiotics without using a separate power source are practical and energy-efficient.

2.2. *Present and emerging treatment technologies for human urine*

Separation at source and treatment of human urine is a field that is a research area gaining popularity because of the merits of management of relatively lower volumes of highly concentrated samples as compared to the volumes and concentration ranges encountered in centralized treatment systems. Current processes for urine treatment technologies are tabulated in Table 3**.**

2.3. *A combination of electrodialysis, microfiltration, and ozonation*

Electrodialysis is a method that utilizes anion and cation exchange membranes and electric potential as a driving mechanism to desalinate industrial wastewater (Liu et al., 2016b). Electrodialysis, when used

Table 3
A summary of urine treatment technologies (Larsen et al., 2013).

Technology		References
1.	A combination of Electrodialysis, Microfiltration and Ozonation	Pronk et al. (2007)
2.	Struvite Precipitation. (Phosphorus Recovery)	(Etter et al., 2011; Winkler et al., 2013)
3.	Ammonia Stripping	(Winkler et al., 2013; Zhang et al., 2012)
4.	Distillation and Nitrification	Udert et al. (2003b)
5.	Urine Electrolysis	Joseph et al. (2006)
6.	Adsorption	Singh and Gupta (2016)

immediately after electrocatalysis, is expected to perform a variety of tasks, which includes and are not limited to (1) Effective elimination of small molecule acids and intermediate intermediates generated during electrocatalysis, (2) removing tiny organic compounds that are resistant to electrocatalysis directly, and (3) The fragmentation of big molecules during electrocatalysis causes less fouling of the ion exchange membrane (Gurreri et al., 2014).

Furthermore, macromolecules such as cellulose and humic substances (H. Ss) are resistant to these reactions. Fortunately, the electrofiltration process can resolve the issues mentioned above. Electro-microfiltration is a separation technique that employs both physical and electrochemical methods. Compared to typical membrane filtration, it opens up possibilities for efficient filtration in which charged solutes are pulled away from the membrane's surface by applying an electric field, resulting in high flux and rejection (Wei et al., 2015). Any charged species, including ions and colloids, might theoretically be removed by electrofiltration if a high electric field is applied. There have been various applications for turbid liquids, lubricants, and drinking water purification (Hakimhashemi et al., 2012; Tsai et al., 2010). Ozone has long been thought to be a highly powerful oxidant and disinfectant. Ozone is predominantly an oxidant in acidic environments. However, it mostly relies on free radical reactions in neutral and alkaline environments (Miklos et al., 2018).

2.4. Struvite formation

The phosphorus recovery from wastewater streams is becoming increasingly important (Baimer, 2004). Studies on phosphorus recovery from urine redirected from feces have been conducted since human urine is one of the most important sources of phosphorus (Larsen and Gujer, 1996). With the addition of magnesium salt and alkaline conditions, phosphorus, the majority of which is in the form of phosphate, could be retrieved from urine as struvite ($MgNH_4PO_4\ 6H_2O$) (i.e., struvite-recovery conditions) (Ban and Dave, 2004). Although this reaction suggests that it can recover phosphorus from urine, chemical reactions relating to the generation of struvite in urine need to be better understood to optimize and control the response for practical application. Equilibrium models are effective instruments for deciphering chemical phenomena. Although several models for struvite precipitation in urine have been developed (Udert et al., 2003a; Masrura et al., 2020), they were not designed for struvite recovery but rather to prevent struvite precipitation from clogging pipes. As a result, those models don't consider residues like magnesium hydroxide ($Mg\ (OH)_2$), which can form during struvite recovery, and nothing is known about the best circumstances for struvite formation. This research explores an equilibrium model which predicts struvite formation in urine under struvite-recovery conditions while accounting for any residues that may arise. In this method, urine is separated from feces using a urine-diversion toilet constructed in the laboratory and stored in a storage tank for a few months (Kutzer et al., 1995).

2.5. Ammonia stripping

Gas stripping is a wastewater treatment method that is widely utilized. It's used to remove VOCs from wastewater (Gonzalez Benito and Garcia Cubero, 1996), ammonia from industrial effluent (Bonmati and Floats, 2003), and swine dung (Basakcilardan-Kabakci et al., 2007). Human urine has only been used in a few studies: have recovered ammonia from human urine by stripping and absorption at a laboratory scale (Pradhan et al., 2017).

2.6. Nitrification and distillation

Biological nitrification to stabilize nutrients in urine is an alternative to struvite synthesis for urine nutrient recovery (Udert et al., 2003). Because volatile ammonia is converted to nitrate during nitrification, a recent study of the inactivation of bacterial and viral surrogates in urine found that nitrification is insufficient as a stand-alone method for pathogen inactivation (Bischel et al., 2015). The nitrified urine can be distilled to produce a concentrated fertilizer with pathogen-killing properties. Nitrified urine is filtered for many hours at 80 °C in a pilot reactor (Udert et al., 2015). As a result, urine nitrification does not provide a major advantage in treating pathogens or drugs found in urine. Combining biological nutrient stabilization with distillation's post-nitrification phases and advanced treatment, on the other hand, can increase the liquid fertilizer's quality by inactivating pathogens and removing pharmaceuticals.

2.7. Electrolysis of urine

Pollutants can be oxidized electrochemically by one of two mechanisms: (1) anodic Oxidation with a direct current source. At the anode surface, contaminants from the bulk solution are removed. (2) Pollution oxidation at the surface of the anode. As a result, the electrochemical process's efficiency will be determined by correlating substrate mass transfer and electron transport at the electrode surface. (3) Indirect Oxidation where a mediator ($HClO$, $H_2S_2O_8$, and others) is electrochemically generated to carry out the Oxidation. Both oxidation mechanisms may coexist, especially in electro-oxidation processes of aqueous effluents, (Salek, 2019).

2.8. Adsorption

Adsorption is a surface phenomenon in which an adsorbate-containing solution binds to the surface of an adsorbent. There are two forms of adsorption: physisorption and adhesion. van der Waals forces bind the adsorbate to the adsorbent during chemical interactions between the adsorbate and the adsorbent cause chemisorption. Chemisorption is irreversible, selective, and exothermic. Physisorption is reversible, weak, and usually endothermic (Lashaki et al., 2016). For instance, activated carbon is recognized as one of the first and most widely used adsorbents for urine and wastewater treatment for removing organic and inorganic pollutants. The application in adsorption process mainly depends on the surface chemistry and pore structure of porous carbons (Bhatnagar et al., 2013). Biochar is a carbon-rich solid derived from pyrolysis of biomass under oxygen-free or sometimes low oxygen content conditions and at temperatures above 250 °C. This adsorption technique is used to remove organic and inorganic environmental contaminants, where it is used as an adsorbent for the immobilization of toxic elements such as heavy metals (Aviso et al., 2019).

Electro-oxidation of pollutants in wastewater is fulfilled through two different approaches, as shown in Fig. 3.

2.9. Urine source separation, treatment, and nutrient recovery

In the circular economy, urine source separation is crucial. On the other hand, pharmaceutical micropollutants in urine restrict it from being used as a fertilizer, particularly in the food industry. Because uptake of pharmaceutical and personal goods by food crops has been observed (Christou et al., 2019; de Boer et al., 2018a), urine must be rendered safe (free of micropollutants) before it can be utilized to fertilize plants. Adsorption of compounds into modified carbonaceous material is an established method for micropollutants removal in wastewater streams (Manjunath and Kumar, 2021). Researchers tested the removal of pharmaceuticals from synthetic urine using various biochars and found that each compound tested was released at a rate greater than 90%. They also found that there was still enough nitrogen and phosphorus in the soil following the biochar treatment. Utilization of Pineapple peel boichar and lateritic soil was shown to be useful in the recovery of $NH4^+$-N from source separated urine (Otieno et al., 2021). Apart from struvite recovery, additional research should focus on other phosphorus recovery technologies to reduce chemical consumption and

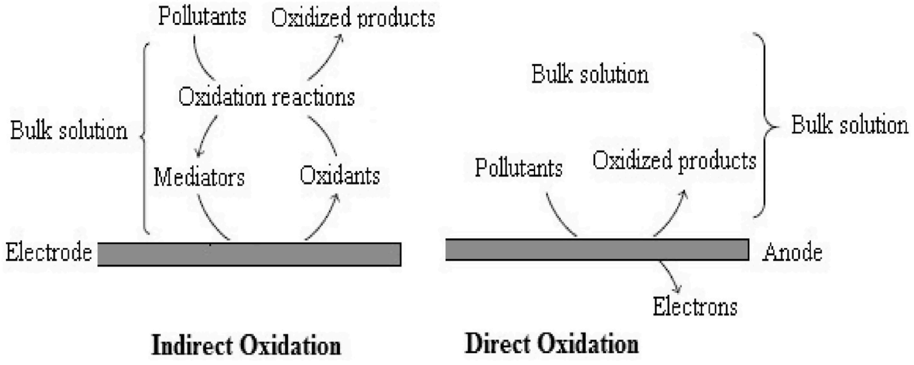

Fig. 3. Pollutant removal pathways in electrochemical Oxidation (Deng and Englehardt, 2007).

create other phosphorus products, such as calcium phosphate, that are more suited to today's fertilizer businesses (De Boer et al., 2018b).

In conclusion, therefore, sustainable and efficient nutrient recycling requires successful management of the whole chain from collection, transportation, and storage to processing and end-use to achieve cost-effective and acceptable end products. There must be a market for the end products; otherwise, they will lose the benefits of nutrient recovery. In addition, to develop technologies, logistics, and regulations, there is a need for an attitude change to make alternative technologies competitive compared to current ones.

2.10. Outlook

Bioelectrochemical systems (BES) have been connected to electrochemical reaction and microbial metabolism and are being studied as a potential technique for emerging pollutants treatment, particularly antibiotics. Moving forward, the development of technologies for the removal of residual antimicrobials from source-separated urine is a whole area of focus. Further research is paramount on the association between microbial diversity, antimicrobials biodegradation, and ARG abundance. Consequently, there is a need for further research into the co-metabolism process in the BES, which contributes to antimicrobials degradation.

3. Conclusion and forecast

This review highlighted the destiny and methods of several treatment methodologies for antibiotics removal from wastewater streams and source-separated urine, including advanced oxidation processes, biological and physical processes, and hyphenated systems. Some of the conclusions include:

(1) Built wetlands are low-cost to run and offer good decontamination, high efficiency, ease of maintenance and management, alongside antibiotic selectivity.
(2) The antibiotics biodegradation treatments impact process conditions, water quality, and environmental parameters.
(3) In combination with AOP therapies, antibiotic elimination is exceptionally successful. They offer a lot of promise for development and application in wastewater treatment.
(4) Antibiotics can be removed using membrane technology and is still a viable option for antibiotic removal from wastewater plants.
(5) separation at the source and treatment of human urine is a major step in controlling the flow of pharmaceuticals into receiving water bodies, especially in informal settlements.

Moving forward, significant efforts should be dedicated towards development of sustainable, low-cost, efficient, and easy-to-manage antibiotic removal systems. Additionally, continuous improvement of existing infrastructure should be prioritized to increase removal efficiency for micropollutants of pharmaceutical nature.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

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