From Removal to Recovery: An Evaluation of Antibiotics Removal Techniques from Wastewater

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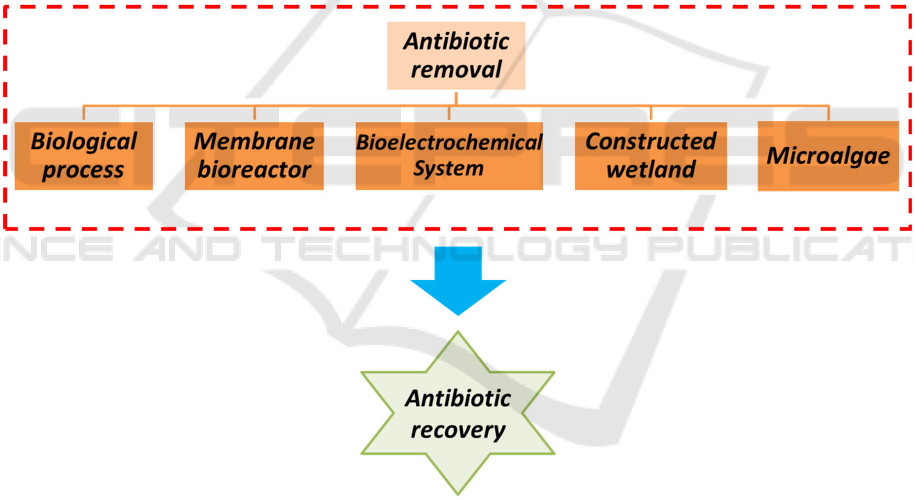
Keywords: Antibiotics Removal, Removal Mechanisms, Biological Wastewater Treatment Technology, Membrane Bioreactor, Bioelectrochemical System, Constructed Wetland, Microalgae.

Abstract: Due to the potential risks to human health and ecosystems, antibiotics contamination is an emerging environmental concern. Currently, numerous technologies have been widely explored for the removal of antibiotics from wastewater, including membrane bioreactor, biological process, bioelectrochemical system, constructed wetland and microalgae. In this review, the fundamental mechanisms and removal efficiency of such technologies were discussed. Besides, current challenges and further direction of the antibiotic removal are present, in which the recovery of antibiotics was highlighted. Thus, a shift of antibiotics from the removal to recovery would be a focus in the future for the sustainable development of society.

Graphical abstract



1 INTRODUCTION

Antibiotics are used in farming and medicine to reduce harmful parasites in crops, and treat diseases by bacterial infection in domesticated animals and humans. Since its discovery in 1928 by Alexander Flemin (K. Gould, 2016), there are now many types of antibiotics such as sulphonamides, macrolides, puinolones, cephalosporins and penicillin. Since the 1950s, people are using antibiotics on a massive scale to treat diseases in poultry and livestock production (E.K. Silbergeld, 2008). They are used to promote the growth of the life stocks by being used as feed additives in reducing the chances of animals getting sick or kills harmful bacteria during fruit or vegetable production (E.K. Silbergeld, 2008). However, the overuse of antibiotics results in the production of harmful chemicals. It was reported that from 2000 to 2010, there was a 36% increase in antibiotics usage in developing countries (Thung, 2016) and this exploitation of antibiotics has caused the appearance of more potent pests that are resistant to the antibiotics, causing more expensive medical treatment, longer treatment period, and increased death rates (Brown, 2016); (Liu, 2016). In a study, it shows that there are about 58% of antibiotics left in

Ma, X.
From Removal to Recovery: An Evaluation of Antibiotics Removal Techniques from Wastewater.
DOI: 10.5220/0011379100003443
In *Proceedings of the 4th International Conference on Biomedical Engineering and Bioinformatics (ICBEB 2022)*, pages 1138-1144
ISBN: 978-989-758-595-1

the feces and urine of those humans and animals that had consumed antibiotics (Zhang, 2015). Therefore, it is crucial to look for a proper way of treating the wastes, so people can prevent the runoff in the livestock industry to pollute water and soil (Ma, 2015); (Lukaszewicz, 2017). Currently, the technologies for removing antibiotics from wastewater include biological process, adsorption, membrane filtration, Fenton, redoxreaciton, photolysis (Martinez, 2013); (Molinos-Senante, 2013). To find out a practical way to remove antibiotics, the review paper takes a deep look into the latest technology available for the antibiotics removal. A comparison among those antibiotics removal techniques was conducted. Besides, we propose a possible solution to increase the feasibility of antibiotics removal system.

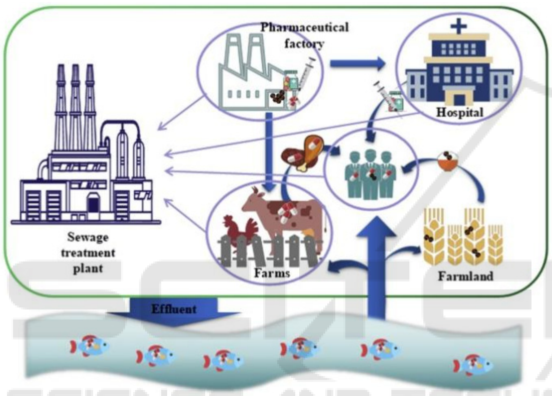


Figure 1: Schematic diagram of antibiotic delivery chain. Adopted from Song et al. (X. Song, 2021).

2 BIOLOGICAL PROCESS

In the biological process, it relies on the oxidation by microbes to effectively decompose antibiotics. Conventional activated sludge (CAS) is the most popular method for the removal of antibiotics through sludge adsorption. However, the method has unstable performance and its removal efficiency ranges from 90% to a negative percentage, which indicates the antibiotics may build up in the reactor. In CAP, the adsorption capacity depends on the antibiotic's hydrophobicity and chemical structure, and the activated sludge's property (Suto, 2017). Compared to the CAS, the sequencing batch reactor (SBR) is more advantageous due to its high flexibility and simple structure and operation. Similar to CAS, the efficiency of SBR depends on property of antibiotics. Besides, hydraulic retention time (HRT), sludge retention time (SRT), condition for redox reaction and temperature can also influence the removal of antibiotics, where HRT and SRT are the most crucial factor (Yang, 2011). Neyestani et al. (Neyestani, 2017) found that the removal efficiency of trimethoprim (TMP) increased from 19% to 71% as the SRT increased from 2 days to 20 days.

Apart from this, anaerobic digestion (AD) is the most efficient way to remove the antibiotics from wastewater due to low energy input and the absence of human intervention. Moreover, the temperature is the most crucial factor affecting the removal efficiency in the anaerobic digestion; for example, Liu et al. (Liu, 2018) reported that the removal efficiencies of tetracyclines (TCs) and fluoroquinolones (FQs) from pig farms were higher in summer than that in winter. It should be noted here that the concentration of antibiotics in the AD system may negatively affect its performance such as hindering the production of CH4 and COD removal. It was reported that the biogas production was reduced by 10% due to the presence of 130mg/L of tylosin, which is attributed to the inhibition of oxidizing bacteria that produce methane by antibiotics (Mitchell, 2013).

3 MEMBRANE BIOREACTOR (MBR)

Membrane bioreactor (MBR) is a more high-quality method for the removal of the antibiotic than biological process due to its long SRT, higher cell concentration, less sludge production and more stable environment for microbial growth, which integrates biological process with micro-filtration (MF) or ultrafiltration (UF) (Cecconet, 2017). The biodegradation of antibiotics can be enhanced by higher microbial concentration and diversity. In MBR, the removal efficiency of antibiotics can be over 90%, including erythromycin, ofloxacin, TC, ciprofloxacin (CIP), and chlortetracycline (Cecconet, 2017); (Tiwari, 2017). For example, Xiao et al. (Xiao, 2017) found that the anaerobic MBR can achieve 94.2 ± 5.5% of TMP and 67.8 ± 13.9% of sulfamethoxazole (SMX) being removed. It should be noted that higher SRT favors the removal of the antibiotic because this facilitates the growth of slow-growing microorganisms and raises the biodiversity of the microbial community, which has positive impacts on the biodegradation of antibiotics.

4 BIOELECTROCHEMICAL SYSTEM

For bioelectrochemical system (BES), it can not only effectively remove antibiotics from wastewater, but also generate electricity through redox reactions performed by anaerobic bacteria at the anode (Logan, 2009) and aerobic bacteria in the cathode by the cellular respiration (Kumar, 2017); (Logan, 2012). The cellular respiration by anaerobic bacteria at the anode chamber produces protons and electrons, in which the proton can diffuse through a proton exchange membrane to the cathode whilst electrons can transfer by the external circuit to react with the electron acceptor at the cathode for the electricity generation. Microbial electrolysis cells (MECs) and microbial fuel cells (MFCs) are the two typical types of BESs which can achieve effective removal of antibiotics with simultaneous energy recovery.

The MFC has high antibiotic removal rate due to its high activity of microbes. It was reported that 98% of β-Lactam antibiotics can be removed in single-chambered MFC while two-chambered MFC achieved over 80 % of TCs, quinolones, chloramphenicol, sulfonamides and nitroimidazoles being removed (Zhang, 2017); (Guo, 2016); (Zhou, 2018). In contrast to the MFC, additional power can be applied in MEC, which facilitates the removal of antibiotics. Increasing the applied voltage from 0 to 0.9 V resulted in enhanced removal of sulfadiazine (SDZ) (Yang, 2018), which is attributed to the improved biofilm formation and increased microbial activity (Liu, 2015). Guo et al. (Guo, 2017) argued that the microbial community can be manipulated by additional power to make the system more suitable for a certain kind of antibiotics. Furthermore, biocathode are more effective at removing antibiotics than abiotic cathode since the microbes at the biocathode can gain electrons to boost the electrochemical process (Liang, 2013). Excessive concentration of antibiotics may also detrimentally affect their removal efficiency in BES (Song, 2013); for instance, Wang et al. (Wang, 2015) found that increasing the concentration of SMX from 20 to 200 mg/L, its half-life increased from 1 to 3 d in the MFC reactor.

5 CONSTRUCTED WETLAND

The removal of antibiotics by constructed wetland (CW) relies on plants, microorganisms and substrates which work together through photodegradation absorption, microbial decomposition and plant uptake (García, 2020); (Gorito, 2017). Microbial decomposition plays a crucial role in the removal of antibiotics in CW, which depends on many different microbes, such as the heterotrophic and autotrophic bacteria, protozoan, and funguis (e.g., yeast, basidiomycetes) (Liu, 2019). Biotransformation processes in treating antibiotics heavily depend on their physio-chemical properties, where antibiotics that have more similar properties to naturally occurring organic molecules can be fairly quickly decomposed and then consumed by bacteria (Li, 2014). Besides, antibiotics can be also destroyed directly through photolysis, in which the radiation from light breaks the bond cleavage, making the molecule no longer harmful. Light intensity is an important factor determining the rate of photodegradation while other influencing factors include light frequency and the chemical property of the surrounding water such as pH and water hardness (Batchu, 2014). Photosensitizers such as dissolved organic matter (DOM), nitrate and nitrite can also indirectly influence the photolysis of antibiotics (Jiang, 2010); (Lin, 2005). The presence of DOM may inhibit the removal of CIP via photodegradation, but have positive impacts on the decomposition for chlortetracycline, roxarsone, and SMX (Mangalgiri, 2017). The removal of antibiotics by substrate adsorption is important, in which the antibiotics can be adsorbed to the substrate such as sludge or the surface of biofilm (Liu, 2020). In addition, some antibiotics in CW can be taken up by plants with the help of some specific xenobiotic transporters in the plant cells (Liu, 2013); (Wu, 2015). According to the difference in water flow, there are several types of CW, including free water surface (FWS) flow wetland, verticle subsurface flow (VSSF) wetland, and horizontal subsurface flow (HSSF) wetland (L. Liu, 2013).

6 MICROALGAE

Microalgae can effectively remove antibiotics by bioadsorption, bioaccumulation, biodegradation, volatilization and photodegradation (SHena, 2021); (Leng, 2020); (Sutherland, 2019). Microalgae integrated systems generally include closed photobioreactors (PBRs) and open ponds (Hena, 2021), where suspension-based microalgae cultivation is always involved. The classes and concentrations of antibiotics highly affect the antibiotic removal by microalgae integrated system. When the concentrations of antibiotics are higher or close to tolerable level of microalgae, the microalgae growth may be inhibited by antibiotics, thus negatively affecting the antibiotic removal (Sun, 2017). However, increasing the antibiotic concentration within the tolerable level of microalgae may have insignificant impacts on the antibiotic removal (Leng, 2020). Li et al. (Li, 2015) argued that the tolerable concentration of amoxicillin for C. pyrenoidosa ranges from 300 to 400 mg/L. Besides, microalgae C. pyrenoidosa can achieve 83–100% of amoxicillin and 7–23% of cefradine being removed while the two antibiotics are at the same level (Li, 2015). This indicates that microalgae may have different removal efficiency for the different antibiotic kinds even they are at the same level (Leng, 2020). Apart from this, the microalgae growth is influenced by microalgae growth inhibitors, dissolved oxygen, design of the photobioreactor, salinity, CO2 concentration, temperature, illumination, solution pH and nutrient concentration, which in turn affects the antibiotic removal by microalgae (Norvill, 2016). The increase in the nutrient concentration is beneficial for the microalgae growth metabolisms, which also has positive impacts on the antibiotic removal; for instance, the removal efficiency of CIP can be increased by over 3 times while adding 4 g/L sodium acetate to Chlamydomonas Mexicana cultures (Xiong, 2017). Moreover, the solution pH is a crucial factor affecting the antibiotic removal in the microalgae integrated system since it may determine the hydrolysis of some ionic antibiotics. Norvill et al. (Norvill, 2017) found that alkaline pH favors the removal of TC by microalgae while the pH ranging from 6.3-8.0 may have negligible effects on the removal of 7-ACA (Guo, 2016). The removal of antibiotics can be also enhanced by strong light irradiation, where reactive oxygen species can be generated (Norvill, 2017).

7 FUTURE PERSPECTIVE

Currently, the risks of antibiotics have been widely explored and many technologies have been developed for their effective removal. Even though bio-based processes could achieve effective antibiotic removal, the stability and reliability of bacteria are still a big challenge for their full-scale applications. Therefore, the selection of appropriate bacteria for biodegrading certain kinds of antibiotics may be a possible solution to increasing the removal performance of bio-based processes. For the MBR and BES, membrane fouling is a big barrier to maintain their performance. This is despite the fact that anaerobic MBR and MFC can recover energy through biogas and electricity production. Thus, further development should focus on improving the properties of membranes with low synthesis costs. The antibiotic removal by CW and microalgae is achieved by multiple mechanisms, so it is essential to identify the specific removal mechanism for certain kinds of antibiotics. Overall, the antibiotic removal requires substantial input of energy and chemicals. Compared to the antibiotic removal from wastewater, the antibiotic recovery is more promising and can create a circular economy because the recovered antibiotics can be reused in animal feed. As shown in Table 1, Pan et al. (Pan, 2015) utilized forward osmosis (FO) process with thin film composite membrane for the recovery of TC. In this scenario, more than 97% TC can be rejected in the feed side of the FO process, which produces the TC-rich streams for further use. Similarly, Li et al. (Li, 2017) employed eggshell membrane (ESM)-derived MgFe2O4 to adsorb doxycycline (DC) with the maximal adsorption capacity of 308 mg/g, after which acid treatment and magnetic separation were conducted for the adsorbent regeneration and DC concentration. However, the technologies mentioned above for the antibiotic recovery may not achieve high-quality stream with rich antibiotics due to the presence of other foreign substances, which may negatively affect the further application of recovered antibiotics. Thus, future research should aim to develop technologies for recovering antibiotics with high-quality products.

Table 1: Summary of technologies for antibiotic recovery from wastewater t-SNPs: thiol functionalized silica nanoparticles.

Technology	Wastewater	Antibiotic	Efficiency	Reference
Foam separation with t-snps	Pharmaceutical wastewater	streptomycin sulfate	recovery rate at 90.1 ± 4.5%	Shu et al. (T. Shu, 2018)
Forward osmosis process	Synthetic wastewater	TC	99% of TC being concentrated	Pan et al. (S.-F. Pan, 2015)
Membrane (RO-UF) filtration	Oxytetracycline waste liquor	oxytetracycline	recovery rate over 60%	Li et al. (S.-z. Li, 2004)
gradient elution preparative chromatography	Crystallization mother-liquor streams	ertapenem	0.6·kg·ertapenem/d·kg·s tationary phase	Sajonz et al. (P. Sajonz, 2006)
Electrodialysis with ultrafiltration membrane	Synthetic wastewater	penicillin G−	20.3% of penicillin G− concentrated	Lu et al. (H. Lu, 2016)

8 CONCLUSION

The review paper demonstrated the effectiveness of several technologies for the antibiotic removal, in which bio-based processes have been widely used for the antibiotic removal. However, there are still some challenges that need to be solved to further improve the removal efficiency, such as how to maintain the microbial community in the removal processes. Besides, the antibiotic recovery is preferred compared to the antibiotic removal, but more research are needed to make it feasibly viable.

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