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Antibiotics: environmental impact and degradation techniques

Duygu Takanoğlu Bulut[a,*], Özkur Kuran[a] and Ahmet Koluman[b]

[a]*Aplication of Advanced Technology and Research Center (ILTAM), Pamukkale University, Denizli, 20160, Türkiye.*
[b]*Department of Biomedical Engineering, Pamukkale University, Denizli, 20160, Türkiye.*

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ABSTRACT

Our reliance on antibiotics, life-saving medications that combat bacterial infections, has inadvertently introduced them into the environment. This paper explores the environmental consequences of this unintended release, focusing on the persistence of antibiotics and their disruption of ecological balance. We delve into the rise of antibiotic-resistant bacteria as a major public health concern linked to this environmental contamination. Recognizing the limitations of existing degradation techniques, the paper emphasizes the need for innovative solutions. We explore the potential of novel materials like engineered nanoparticles and biochar alongside investigating unconventional degradation mechanisms found in extreme environments. Ultimately, the paper underscores the importance of collaborative research efforts and the development of sustainable solutions to mitigate the environmental impact of antibiotics and safeguard the future effectiveness of these critical medications.

I. INTRODUCTION

The dawn of the antibiotic era marked a pivotal moment in human history. Antibiotics, those wonder drugs capable of combating bacterial infections that once ravaged humanity, revolutionized healthcare. Their introduction dramatically reduced mortality rates and ushered in an era of improved global health. Antibiotics are among the most widely used drugs worldwide. They are natural, synthetic, or semi-synthetic complex molecules capable of killing microorganisms or inhibiting their metabolic activities. Due to different functional groups in their chemical structures, these biological agents have antibacterial, antiparasitic, and antifungal effects. Antibiotics are drugs specifically designed to treat infections in humans and animals. The first antibiotics were natural compounds produced by microorganisms, such as penicillin, from the culture of *Penicillium* notatum. Antibiotics can be classified in three distinct ways. Based on their chemical structure, antibiotics can be classified as beta-lactams, sulfonamides, aminoglycosides, macrolides, tetracyclines, and fluoroquinolones. According to their spectrum of action, antibiotics can also be categorized into narrow-spectrum, broad-spectrum, and extended-spectrum antibiotics. Antibiotics can also be classified according to their mechanism of action. These mechanisms include inhibition of cell wall synthesis, alteration of cell membranes, inhibition of protein synthesis, inhibition of nucleic acid synthesis, competitive antagonism, and anti-metabolite activity [1-5]. Since Fleming's discovery of penicillin, more than 250 types of antibiotics have been produced worldwide. This variation allows antibiotics to effectively combat different types of bacterial species and treat different infections. Antibiotics are used extensively in various fields, such as medicine, veterinary medicine, agriculture, and aquaculture. Their main purpose is to combat

*Corresponding author. Tel.: +90-258-296 3288; e-mail: dbulut@pau.edu.tr

bacterial infections and protect human, animal, and plant health. However, as with many technological advancements, this triumph comes with unforeseen environmental consequences [6-10].

Unfortunately, our reliance on antibiotics has led to their unintended release into the environment. Improper disposal of unused medications, often due to a lack of public awareness or inadequate infrastructure, allows these life-saving drugs to enter landfills and wastewater streams. Furthermore, even after a course of antibiotics, residual amounts are excreted through human waste. These combined factors contribute to a growing problem – the presence of persistent antibiotic pollutants in soil and water sources [11, 12].

Antibiotics have a multifaceted environmental impact. Unlike many natural compounds, antibiotics often exhibit high persistence, meaning they resist natural degradation processes and linger in the environment for extended periods. This persistence disrupts the delicate balance of microbial communities within soil and water ecosystems. These communities play essential roles in nutrient cycling, decomposition, and maintaining overall ecosystem health. The presence of antibiotics can disrupt these vital functions, leading to unforeseen consequences for entire ecological systems [13, 14].

The rise of antibiotic-resistant bacteria is perhaps the most concerning consequence of environmental antibiotic contamination [14, 15]. The specter of antibiotic resistance presents a chilling possibility – a return to the pre-antibiotic era where even minor infections could prove fatal. Addressing this issue requires a multi-pronged approach. Stricter regulations on antibiotic use in agriculture and medicine are crucial to minimizing environmental release. Research and development of new antibiotics with improved degradability is another crucial step. Furthermore, public awareness campaigns can promote responsible antibiotic use and proper disposal of unused medications [16-18].

By acknowledging the environmental impact of antibiotics and taking proactive steps to mitigate it, we can ensure that these life-saving drugs continue to be effective for generations to come. The fight against infectious disease is a continuous battle, and safeguarding the effectiveness of antibiotics is an essential front in this ongoing war.

This review focuses on the environmental problems caused by antibiotic usage and analyzes the degradation of antibiotic pollution by natural and enhanced methods.

II. ENVIRONMENTAL IMPACT OF ANTIBIOTICS

Antibiotics are one of the most important pollutants, and they have been used increasingly in various sectors since penicillin was discovered in 1929 [19-21]. Global antibiotic consumption is predicted to be between 100,000 and 200,000 tons per year [22, 23].

The World Health Organization (WHO) report on the medical use of antibiotics indicates that total consumption in the 65 countries monitored was 14,000 tons, with the highest consumption observed in Brazil, Turkey, and Iran. Within the European region, the amount of antibiotic usage in 2015, defined by the daily defined dose (DDD) rate, was 19.5 per 1000 inhabitants per day, with the greatest values in Turkey, Greece, and Serbia, with values of 38.2, 33.9, and 31.6, respectively [24]. Conversely, data reported by the European Center for Disease Prevention and Control show that overall antibiotic usage in the European Economic Area in 2021 averaged 16.4 DDD, ranging from 8.3 in the Netherlands to 25.7 in Romania, indicating the likely overuse of antibiotics in several regions of Europe [25, 26].

Antibiotics can treat bacterial infections and kill or inhibit bacterial growth, which promotes animal growth and improves nutritional performance [27, 28]. Unfortunately, due to the low absorption capacity of animal and human intestines, approximately 30-90% of significant antibiotics are excreted in urine or feces [27, 29]. In 2013, humans and animals in China excreted about 54,000 tons of antibiotics, of which over 99% were released into the ecosystem [30]. Non-metabolized antibiotics have been introduced into soil and water via wastewater irrigation and through animal manure and sludge manure [31, 32]. Antibiotics have been extensively found in soil and water systems such as agricultural soils [33, 34], urban green spaces [35], forest lands [36], surface water [37 ,38], groundwater [39], and seawater [40]. Figure 1 shows pathways for antibiotic residues into the environment. Antibiotics in the environment can affect the environment and human health through microbial growth inhibition, poisonous to algae and plants, and the evolution of antibiotic resistance [2, 41]. The presence of low levels of antibiotics in the environment aids in the spread of antibiotic-resistant bacteria and antibiotic-resistance genes and increases the resistance of pathogenic bacteria. The uptake of antibiotics and antibiotic-resistance genes from the environment through food chains poses serious threats to human health, further affecting human gut health and the effectiveness of treatment against bacterial infections. Many derivative compounds (TPs) resulting from the natural degradation of antibiotics, technical water treatment, and metabolism of humans and animals may confer antimicrobial resistance and toxicity due to their structural similarity to the main compounds [42].

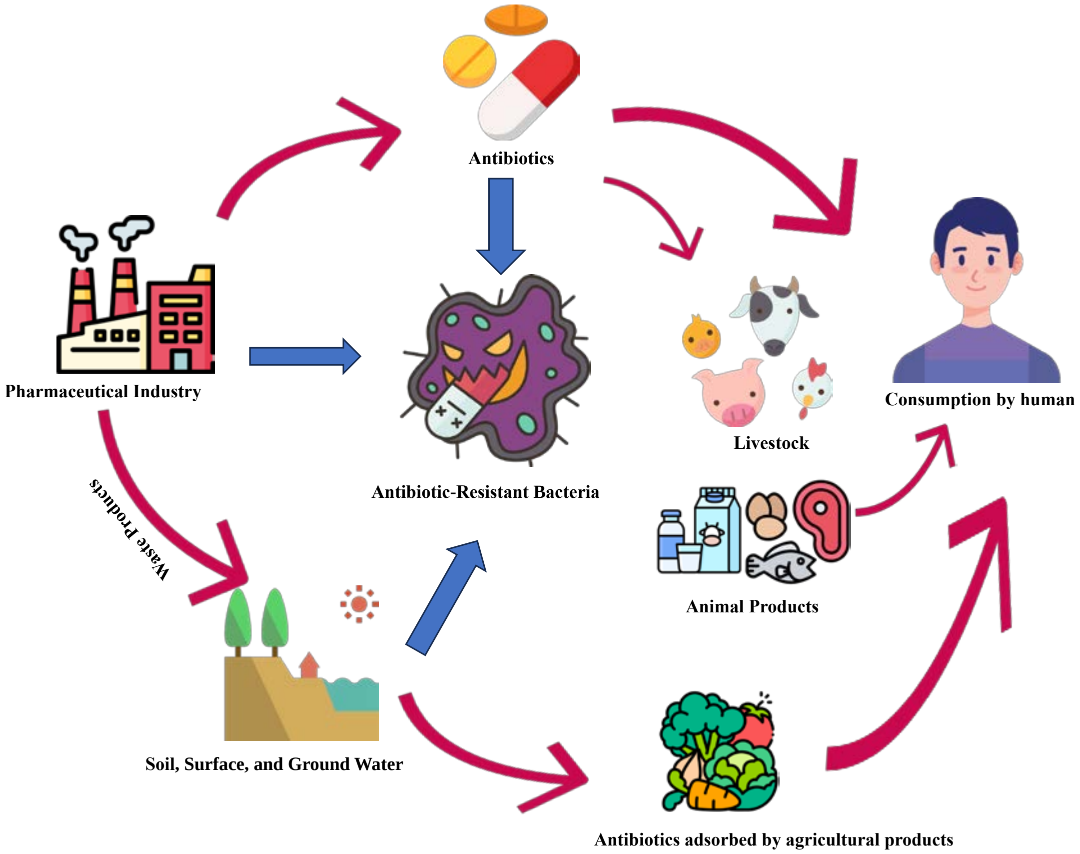

Figure 1. Pathways for antibiotic residues into the environment

Perhaps the most concerning consequence of environmental antibiotic contamination is the rise of antibiotic-resistant bacteria. The constant existence of these drugs in the environment exerts a selective pressure on bacterial populations. Bacteria with mutations or genetic adaptations that allow them to avoid the effects of antibiotics are more likely to survive and reproduce. Over time, this selection pressure leads to the emergence and propagation of antibiotic-resistant bacteria, commonly referred to as "superbugs" [9, 43-45].

These superbugs pose a significant threat to public health as they render previously effective antibiotics useless in combating infections. Once easily treatable, simple infections can become life-threatening when traditional antibiotic therapies fail. This scenario could potentially lead to a return to the pre-antibiotic era, where even minor infections could prove fatal. The potential societal and economic costs associated with widespread antibiotic resistance are staggering, highlighting the urgency of addressing this issue [46-48].

In Organization for Economic Cooperation and Development (OECD) countries, antibiotic resistance rates, including combinations of eight different antibiotics and bacteria, are estimated to have increased from 3%-33% in 2005 to 3%-39% in 2015. If the current trends in resistance and associated factors persist, this range is estimated to increase by 6-40% by 2030. Figure 2 shows the projected rates of antibiotic resistance for some OECD countries by 2030. In addition, between 2005 and 2015, the projected ratio of infections caused by resistant bacteria increased considerably faster than predicted for 2015-2030. However, these averages conceal significant variations across countries and among antibiotic-bacterium combinations [49].

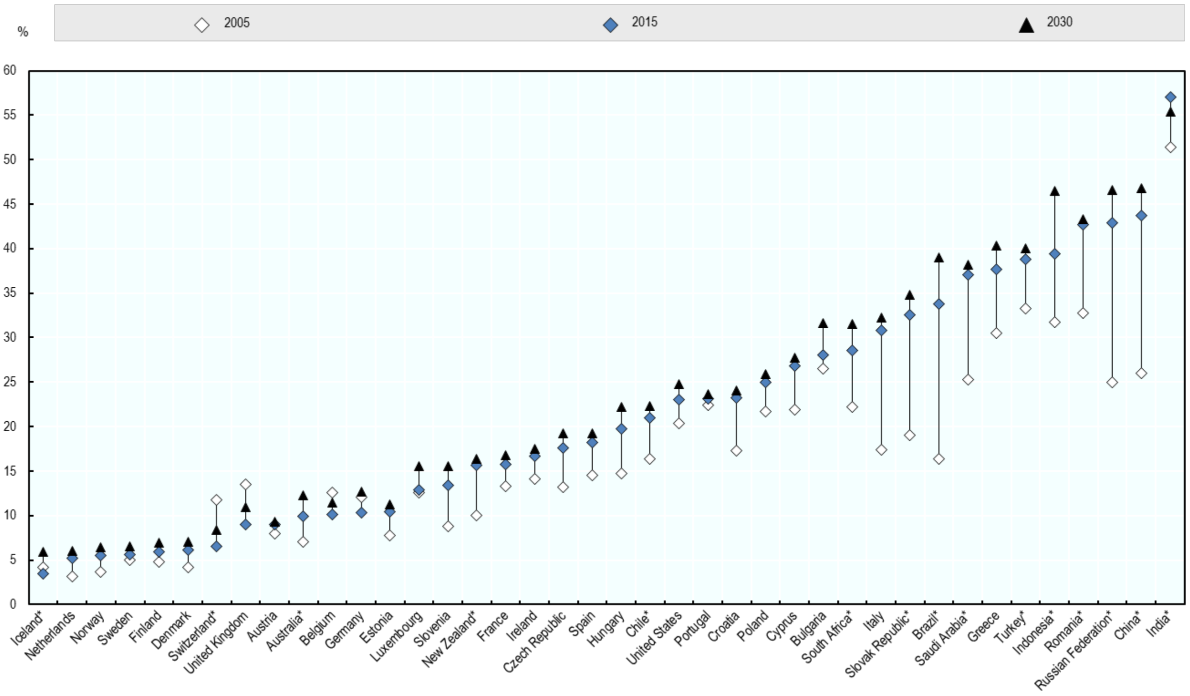

Figure 2. The projected rates of antibiotic resistance for some OECD countries for eight antibiotic-bacterium combinations in 2005, 2015, and 2030 (49)

Addressing the environmental impact of antibiotics and the subsequent rise of antibiotic resistance requires a multifaceted approach. On the regulatory front, stricter control of antibiotic use in agriculture and medicine is crucial to minimize environmental release. The agricultural sector, in particular, relies heavily on antibiotics for prophylactic use in livestock, contributing significantly to environmental contamination. Developing and implementing stricter regulations on the use of antibiotics in agriculture, coupled with the exploration of alternative treatments and preventative measures, is essential [50-52].

In medicine, promoting responsible antibiotic use through targeted education campaigns for healthcare professionals and the public alike is crucial. Overprescription and misuse of antibiotics in human healthcare contribute to resistance development. Educating healthcare professionals on appropriate antibiotic prescribing practices and encouraging patients to complete prescribed antibiotic courses are essential steps in tackling this issue [53-56].

The scientific community plays a vital role in developing new strategies to combat antibiotic resistance. Research and development efforts should focus on novel antibiotics with improved degradability, minimizing their persistence in the environment. Additionally, exploring alternative treatment options such as bacteriophages (viruses that specifically target bacteria) and immunomodulatory therapies holds promise for future generations of antibacterial treatments [57-59].

Finally, fostering international collaboration is essential to effectively address the problem of antibiotic resistance. The spread of resistant bacteria transcends national borders, highlighting the need for coordinated efforts across the globe. Sharing best practices on antibiotic stewardship, promoting research collaboration, and facilitating knowledge transfer are all crucial steps toward mitigating this global threat. The story of antibiotics serves as a poignant reminder that even the most revolutionary medical advancements can have unforeseen consequences. Our collective reliance on these life-saving drugs has inadvertently led to their release into the environment, disrupting ecosystems and fueling the emergence of antibiotic-resistant bacteria. Addressing this complex issue requires a multi-pronged approach that encompasses stricter regulations, responsible antibiotic use practices, and ongoing research and development. By acknowledging the environmental impact of antibiotics and taking proactive steps to mitigate it, we can ensure that these wonder drugs continue to be effective for generations to come. The fight against infectious disease is a continuous battle, and safeguarding the effectiveness of antibiotics is an essential front in this ongoing war [58, 60].

In the battle against antibiotic resistance, the 'One Health' policy of the WHO is of critical importance. One Health is an integrated approach aimed at sustainably balancing and optimizing the health of people, animals, and ecosystems. This policy recognizes the direct link between people, animals, and the environment. It aims to address various health issues, such as the spread of infectious diseases, antimicrobial resistance, food and water security. The policy also elaborates on the need for monitoring and surveillance systems to prevent and control health threats. Additionally, WHO, in collaboration with the Group of 20 (G20) countries, will promote international cooperation and knowledge transfer to guide the implementation of the One Health policy. Global efforts to use antibiotics responsibly and reduce their environmental impact will thus set the course for the future of infectious disease management. Antibiotics will remain the cornerstone of global health [61].

III. DEGRADATION TECHNIQUES OF ANTIBIOTICS

Antibiotics are ionizable compounds and can exist in nature as neutral or charged (positive or negative). Because of their different chemical properties, antibiotics have different absorption and degradation mechanisms in soil. There are many biotic and abiotic factors that influence the rate of degradation of these compounds in nature. Therefore, the half-life and degradation mechanism of each antibiotic compound in the soil is different. For example, β-lactams are more sensitive to abiotic degradation than macrolides or sulfonamides, while quinolones and tetracyclines are more sensitive to photodegradation [2, 62-64].

Unfortunately, traditional wastewater treatment methods are not adequate for the removal of antibiotics with high polarity. Hence, many different techniques have been or are being tried by researchers to degrade the environmental impact of antibiotics. These include physical, chemical and biological processes, each of which has its own advantages and disadvantages. In this review article, some of the widely used and high-potential degradation techniques for the degradation of antibiotics are discussed. Table 1 summarizes of the advantages and disadvantages of the antibiotic degradation methods discussed in this review.

Microbial augmentation offers a promising strategy for remediating contaminated sites. This technique involves the deliberate introduction of specifically chosen microbial strains known to possess potent antibiotic degradation capabilities. These strains are equipped with specialized enzymes that can efficiently break down targeted antibiotics. By introducing these "super degraders" into contaminated soil or water, the overall degradation rate can be significantly accelerated. However, biodegradation of antibiotics with the help of microbial strains depends on the type of antibiotic, the microbial species used, temperature, wastewater components, carbon and nitrogen sources present in the environment [65]. In recent years, many microbial species capable of antibiotic degradation have been isolated. For example, in a laboratory setting, the antibiotic 10 mg/L sulfamethoxazole was completely inhibited within three days using the bacterium Shewanella sp. MR-4 [66]. In another study using hospital wastewater, 5 mg/L Ciprofloxacin antibiotic was inhibited by 74% in fourteen days using Bacillus sp. (KM504129) bacteria [67]. However, the implementation of microbial augmentation requires careful consideration and planning. The introduction of foreign microbial strains into an existing ecosystem can disrupt the delicate balance of native microbial communities. Furthermore, the potential for horizontal gene transfer exists, whereby antibiotic resistance genes harbored by the introduced strains could be transferred to resident bacteria, potentially exacerbating the problem of resistance. To make sure this method works and lasts, strict criteria must be used to choose the strains that are introduced and close attention must be paid to how they affect the ecosystem [68-70].

Another area of exploration involves the use of enzymes for targeted antibiotic degradation. Enzymes, nature's biological catalysts, can be harnessed to break down specific antibiotics with remarkable efficiency. One such enzyme receiving significant attention is laccase, which exhibits the ability to degrade a broad spectrum of antibiotics. By introducing laccase to contaminated environments, the breakdown of targeted antibiotics can be significantly accelerated. In a study investigating the degradation mechanisms of thirty-eight different antibiotics, the laccase enzyme produced by T. Versicolor was used. In the study, 96.6% degradation rate was obtained for amoxicillin and 88.6% degradation rate for ampicillin, which are included in the penicillin group, while this rate was reported to vary between 50.1% and 59.4% for ofloxacin, ciprofloxacin, enrofloxacin, danofloxacin and marbofloxacin antibiotics included in the fluoroquinolone group. Finally, it was found that this rate varied between

26.0% and 48.4% for antibiotics included in the tetracycline group, such as oxytetracycline, chlortetracycline, doxycycline, and tetracycline. These differences between the removal rates are thought to be due to the molecular structure of antibiotics [71, 72]. However, the use of enzyme-based techniques also presents limitations. The breakdown products generated from antibiotic degradation by enzymes like laccase might possess novel properties, including residual antimicrobial activity. These breakdown products could contribute to the overall pool of environmental contamination, negating the intended benefits of remediation. Therefore, a thorough understanding of the breakdown products and their potential environmental impact is crucial before the widespread application of this technique. Furthermore, the cost-effectiveness and scalability of enzyme-based remediation for large-scale environmental applications require further investigation [73-75].

Table 1. Advantages and disadvantages of antibiotic degradation techniques

Degradation Method	Advantages	Disadvantages
Microbial Augmentation	• Natural Process • Wide range of activity in degrading different pollutants • Cost-effectiveness	• Disruption of the delicate balance of local microbial communities. • Antibiotic-resistant bacteria may occur owing to horizontal gene transfer
Enzyme Degradation	• Mild operation conditions • Environmentally friendly • Biodegradability	• Stability and standardization issues • Antibiotic-resistant bacteria may occur owing to horizontal gene transfer
Membrane Filtration	• High separation efficiency • Small space requirements • Environmental friendly	• High maintenance cost • Not degraded, only filtered • Membrane fouling
Electrocoagulation	• Simple installation and operation • No need for chemical processing • Can be fully automated • Large treatment capacity • Relatively short time	• The formation of passive films on the electrode surface leads to increased operation time and energy consumption • High operating cost of the reactor
Photocatalytic Degradation	• Fast reaction rate • No secondary pollution • Natural energy source	• Special reactor design • Catalysts usually include rare metals • Low quantum efficiency

Advanced treatment techniques are being explored for large-scale antibiotic removal from water sources. Constructed wetlands and bioreactors utilize naturally occurring microbial communities housed within a controlled environment. Contaminated water is channeled through these systems, allowing the resident microbial communities to degrade the antibiotics present. This approach leverages the power of natural degradation processes within a managed setting, offering a potential solution for wastewater treatment. However, the effectiveness and scalability of constructed wetlands and bioreactors for broader environmental remediation efforts require further investigation [76-78].

Membrane technology is a new approach to separation method that includes selectively permeable membranes with a wide range of applications for removing antibiotics from water sources. Membrane technology is classified into three groups depending on their driving forces: low-pressure (microfiltration, ultrafiltration, and distillation), high-pressure (nanofiltration, and reverse osmosis), and osmotic pressure (forward osmosis, electrodialysis, and liquid membrane). Membrane filtration is a physical technique that does not require any chemical additives and provides a high degree of separation. Also, this method is environmentally friendly, offers good selective filtration, and has small space requirements. For example, in one study, a tetracycline hydrochloride antibiotic was found to be filtered by 89% in a treatment process with a sulfated cellulose nanocrystal composite membrane [79, 80]. In another study, a two-stage reverse osmosis membrane filtration system was examined for pig farm wastewater treatment. The presence of sixteen different target antibiotic resistance genes, which accounted for 72.64% of the total antibiotic resistance genes in pig farm wastewater, was monitored at each stage of the membrane filtration process. After the integrated membrane filtration process, it was reported that more than 99.02% of total contaminants and 99.79% of antibiotic-resistant genes were filtered out of the tested effluents [81]. However, this technology does not remove or degrade the pollutant. The process only transfers the contaminants to a new phase (membrane), where they accumulate in a denser form within the membrane filter. As a result of the small size of the antibiotic molecules and the tendency of other contaminants to clog the pores, the membrane module needs to be changed frequently, incurring an extra maintenance cost [65, 79, 82].

The emergence of enhanced degradation techniques represents a significant step forward in combating antibiotic contamination in the environment. Techniques such as microbial augmentation, enzyme-based degradation, and advanced treatment systems offer promising solutions for accelerating antibiotic removal and mitigating their long-term environmental impact. However, each technique presents its own limitations and requires careful consideration. Ultimately, a multifaceted approach combining responsible antibiotic use, enhanced degradation techniques, and ongoing research and development is crucial to ensure the continued effectiveness of antibiotics and protect the health of our planet [83-85].

In recent years, the electrocoagulation process has emerged as a promising technology. With low sludge production and low treatment costs, it effectively removes a wide range of contaminants from water and wastewater [86, 87, 88]. Electrocoagulation is a new approach for the removal of pharmaceutical compounds, including antibiotics, from water and wastewater. Electrocoagulation offers the advantages of both traditional chemical coagulation and electrochemical methods [89-91]. This process involves immersing iron or aluminum electrodes into a wastewater-containing electrolytic solution and applying direct current. The coagulant is generated through the oxidation of the anode and simultaneously produces hydroxyl ions and hydrogen gas at the cathode. The gases generated at the electrodes cause a flotation effect, which separates contaminants from the floc-foam layer on the water surface. The generation of metal hydroxide from the dissolved metal cations combines with the hydroxyl ions to form metal hydroxide. Ultimately, it removes pollutants in the wastewater through complexation or electrostatic attraction. The generation of Fe(OH)n(s) can be divalent or trivalent. It is strongly related to the water's pH and electrolyte concentration, so the optimal pH of the water must be maintained. Electrocoagulation has been found to be efficient in removing pharmaceutical compounds, including antibiotics, from water and wastewater [92]. For instance, a study reported that electrocoagulation using an iron electrode was effective in degrading 100% of tetracycline antibiotic from water in 15 minutes, with an initial pH of 4.3, a current density of 4.17 A/m^2, and a conductivity of 1000 µS/cm [93]. Another study reported that, in comparison to an iron anode, an aluminum electrode was capable of removing 99.8% of 10 mg/L TC within the first 20 minutes under optimized parameters with a 9 V voltage and 2 cm electrode spacing. This was in contrast to the 83.8% removal rate observed for the iron electrode in the same period [91]. However, energy consumption plays a vital role in the electrocoagulation method. The energy consumption is related to the electric current and applied voltage, and the mass of iron electrodes dissolved in the solution is described by Faraday's law. Therefore, optimizing the operating conditions, such as the electrode material, current density, pH, and electrolyte concentration, is crucial to achieving efficient and cost-effective removal of pharmaceutical compounds from water and wastewater [94-97].

Due to recent nanotechnology and green chemistry developments, photocatalysis is a potential approach for degrading pharmaceutical contaminants. Photocatalysis, a novel method in wastewater treatment, has shown significant potential for reducing antibiotic residues. Hence, advanced oxidation processes such as photocatalysis, which provide the possibility of total mineralization of organic compounds via the in situ production of hydroxyl radicals, which are strong oxidants that unselectively oxidize organic molecules, have been adopted as an alternative to water remediation [98, 99]. In the photocatalysis method, antibiotic molecules are first adsorbed to the surface of the material used as photocatalysts. Antibiotic molecules adsorbed on the surface of the photocatalyst degrade under the influence of light absorbed by the photocatalyst. This allows antibiotic molecules to be converted into simpler and harmless components. Photocatalytic materials are generally active at specific wavelengths, such as ultraviolet (UV) or visible light. Consequently, semiconductor compounds or metal-organic frameworks (MOFs) are generally preferred as photocatalysts. In recent years, visible light active semiconductors such as Cu_2O [100], WO_3 [101], $BiVO_4$ [102], Fe_2O_3 [103], g-C_3N_4 [104, 105], have been widely used as photocatalysts. Metal-organic frameworks (MOFs) are highly porous structures with customizable functions. Due to these properties, they can be used to adsorb and degrade specific antibiotics [106]. TiO_2 semiconductor, which is often used as a photocatalyst by researchers in recent years, has been reported to degrade 0.5 mg/L oxytetracycline antibiotic 100% in eighty minutes under natural sunlight [107, 108]. Another widely used semiconductor, Bi_2WO_4, was used to reduce the antibiotic levofloxacin. This study varied the catalyst dose and levofloxacin concentration to determine the optimum degradation conditions. As a result of the experimental studies, 0.4 g/L Bi2WO4 was used for 20 mg/L levofloxacin concentration, and the degradation efficiency was 100%. In the further study, the levofloxacin dose varied in the 10-40 mg/L range for 0.4 g/L Bi_2WO_4 concentration. With increasing levofloxacin dose, the degradation rate was found to decrease from 98% to 49% [109]. The effectiveness of photocatalytic methods in reducing antibiotics depends on the properties of the photocatalyst used, the processing conditions (e.g., light intensity and wavelength, pH value of the solution), and the chemical structures of the antibiotic molecules. Therefore, the process must be optimized and studied carefully to achieve the best results [98].

IV. CONCLUSIONS

The environmental impact of antibiotics presents a complex challenge demanding a multifaceted response. While existing degradation techniques play a vital role, the limitations they present necessitate a focus on innovation. The development of novel materials like engineered nanoparticles and biochar, coupled with exploration of unconventional degradation mechanisms, holds tremendous promise for the future of antibiotic remediation. By fostering collaborative research efforts and prioritizing these innovative approaches, we can safeguard our environment from the deleterious effects of antibiotic contamination. Ultimately, our collective well-being hinges on securing a sustainable future where antibiotics remain effective tools in the fight against infectious diseases while ensuring the health of the ecosystems upon which we all depend.

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REFERENCES

1. Gothwal R, Shashidhar T (2015) Antibiotic Pollution in the Environment: A Review. Clean Soil Air Water 43: 479-489. https://doi.org/10.1002/clen.201300989
2. Cycoń M, Mrozik A, Piotrowska-Seget Z (2019) Antibiotics in the Soil Environment—Degradation and Their Impact on Microbial Activity and Diversity. Frontiers in Microbiology 10:338 https://doi.org/10.3389/fmicb.2019.00338
3. Zhang X, Xu Y, Liu Y, Wei Y, Lan F, Wang R, Yang Y, Chen J (2024) Research progress and trend of antibiotics degradation by electroactive biofilm: A review. Journal of Water Process Engineering 58:10846. https://doi.org/10.1016/j.jwpe.2024.104846
4. Yang Q, Gao Y, Ke J, Show P L, Ge Y, Liu Y, Guo R, Chen J (2021) Antibiotics: An overview on the environmental occurrence, toxicity, degradation, and removal methods. *Bioengineered 12*(1), 7376–7416. https://doi.org/10.1080/21655979.2021.1974657
5. Kümmerer K (2009) Antibiotics in the aquatic environment–a review–Part, I. Chemosphere 75:417–434. https://doi.org/10.1016/j.chemosphere.2008.11.086
6. Mwangi J, Hao X, Lai R, Zhang Z (2019) Antimicrobial peptides: new hope in the war against multidrug resistance. Zoological Research 40:488-505. https://doi.org/10.24272/j.issn.2095-8137.2019.062
7. Zhao Y, Chen Z, Chen Y, Xu J, Li J, Jiang X (2013) Synergy of non-antibiotic drugs and pyrimidinethiol on gold nanoparticles against superbugs. Journal of the American Chemical Society 135(35):12940-12943. https://doi.org/10.1021/ja4058635
8. Schneider E, Reyes-Ortega F, Velkov T, Li J (2017) Antibiotic-non-antibiotic combinations for combating extremely drug-resistant Gram-negative 'superbugs'. Essays in biochemistry 61(1):115-125. https://doi.org/10.1042/EBC20160058
9. Ramalingam A (2015) History of Antibiotics and Evolution of Resistance. Research Journal of Pharmacy and Technology 8:1719-1724. https://doi.org/10.5958/0974-360X.2015.00309.1
10. Koluman A, Dikici A (2013) Antimicrobial resistance of emerging foodborne pathogens: Status quo and global trends. Critical Reviews in Microbiology 39(1):57–69. https://doi.org/10.3109/1040841X.2012.691458
11. Pramanik A, Jones S, Pedraza F, Vangara A, Sweet C, Williams M, Ruppa-Kasani V, Risher S, Sardar D, Ray P (2017) Fluorescent, Magnetic Multifunctional Carbon Dots for Selective Separation, Identification, and Eradication of Drug-Resistant Superbugs. ACS Omega 2:554-562. https://doi.org/10.1021/acsomega.6b00518
12. Gao Y, Pramanik A, Patibandla S, Gates K, Hill G, Ignatius A, Ray P (2020) Development of Human Host Defense Antimicrobial Peptide-Conjugated Biochar Nanocomposites for Combating Broad-Spectrum Superbugs. ACS applied bio materials 3(11):7696-7705. https://doi.org/10.1021/acsabm.0c00880.s001
13. Helfand M (2008) β-lactams against emerging ‘superbugs’: progress and pitfalls. Expert Review of Clinical Pharmacology 1:559-571. https://doi.org/10.1586/17512433.1.4.559
14. Dominguez D, Meza-Rodriguez S (2019) Development of antimicrobial resistance: future challenges. Pharmaceuticals and Personal Care Products:Waste Management and Treatment Technology, 1st edn. Butterworth-Heinemann, ss 383-408. https://doi.org/10.1016/B978-0-12-816189-0.00016-0
15. Jones L, Howe R (2014) Microbial Resistance and Superbugs. In: Steven LP, David WW, Randle J, Tracey C. (eds) Biofilms in infection prevention and control, 1st edn. Academic Press, pp. 257-285. https://doi.org/10.1016/B978-0-12-397043-5.00015-3
16. Bonatelli M, Oliveira L, Pinto T (2020) Superbugs Among Us: Who They Are and What Can You Do to Help Win the Fight?. Front. Young Minds. 8:5. https://doi.org/10.3389/frym.2020.00005
17. Rashid M, Tariq P, Rashid H, Ali Z, Andleeb S, Gul A, Ozturk M, Altay V (2020) Superbugs, silver bullets, and new battlefields. Biodiversity and Biomedicine 81-106. https://doi.org/10.1016/b978-0-12-819541-3.00006-2
18. Velkov T, Zhu C, Haddleton D, Li J (2017) Novel Antimicrobial Peptides: Targeting Wound Infections Caused by ‘Superbugs’ Resistant to All Current Antibiotics. In: Shiffman, M., Low, M. (eds) Burns, Infections and Wound Management. Recent Clinical Techniques, Results, and Research in Wounds, vol 2. Springer, Cham. pp. 203-211. https://doi.org/10.1007/15695_2017_34

19. Ghosh S, et al (2023) Recent progress on the remediation of metronidazole antibiotic as emerging contaminant from water environments using sustainable adsorbents: a review. Journal of Water Process Engineering 51:103405. https://doi.org/10.1016/j.jwpe.2022.103405
20. Bodus B, O'Malley K, Dieter G, Gunawardana C, McDonald W (2024) Review of emerging contaminants in green stormwater infrastructure: Antibiotic resistance genes, microplastics, tire wear particles, PFAS, and temperature. Science of The Total Environment 906:167195. https://doi.org/10.1016/j.scitotenv.2023.167195
21. Haider R (2023) Penicillin and the antibiotics revolution global history. Asian Journal of Pharmaceutical Research 13(1):55–62. https://doi.org/10.52711/2231-5691.2023.00011
22. Mahmud F, et al (2024) Antibiotic-contaminated wastewater treatment and remediation by electrochemical advanced oxidation processes (EAOPs). Groundwater for Sustainable Development 25:101181. https://doi.org/10.1016/j.gsd.2024.101181
23. Fallahzadeha RA, et al (2020) Investigating the effect of photo-electro oxidation process modified with activated carbon bed as a porous electrode on amoxicillin removal from aqueous solutions. Desalination and Water Treatment 185:185–195. https://doi.org/10.5004/dwt.2020.25400
24. WHO, 2019b (2019) WHO report on surveillance of antibiotic consumption [www Document]. URL. https://www.who.int/publications/i/item/who-report-on-surveillance-of-antibiotic-consumption Accessed 25 April.
25. Machowska A, Stålsby Lundborg C (2019) Drivers of Irrational Use of Antibiotics in Europe. International Journal of Environmental Research and Public Health 16(1):27. https://doi.org/10.3390/ijerph16010027
26. Rzymski P, Gwenzi W, Poniedziałek B, Mangul S, Fal A (2024) Climate warming, environmental degradation and pollution as drivers of antibiotic resistance. Environmental Pollution 346:123649 https://doi.org/10.1016/j.envpol.2024.123649
27. Conde-Cid M, Núñez-Delgado A, Fernández-Sanjurjo MJ, Álvarez-Rodríguez E, Fernández-Calviño D, Arias-Estévez M (2020) Tetracycline and Sulfonamide Antibiotics in Soils: Presence, Fate and Environmental Risks. Processes 8 (11):1479. https://doi.org/10.3390/pr8111479
28. Yang Q, Gao Y, Ke J, Show PL, Ge Y, Liu Y, et al (2021) Antibiotics: An overview on the environmental occurrence, toxicity, degradation, and removal methods. Bioengineered 12(1):7376–7416. https://doi.org/10.1080/21655979.2021.1974657
29. Lyu J, Yang L, Zhang L, Ye B, Wang L (2020) Antibiotics in soil and water in China–a systematic review and source analysis. Environmental Pollution 266:115147. https://doi.org/10.1016/j.envpol.2020.115147
30. Zhang QQ, Ying GG, Pan CG, Liu YS, Zhao JL (2015) Comprehensive evaluation of antibiotics emission and fate in the river basins of China: source analysis, multimedia modeling, and linkage to bacterial resistance. Environ Sci Technol 49(11):6772–6782. https://doi.org/10.1021/acs.est.5b00729
31. Wu J, Wang J, Li Z, Guo S, Li K, Xu P, Ok YS, Jones DL, Zou J (2022) Antibiotics and antibiotic resistance genes in agricultural soils: a systematic analysis. Critical Reviews in Environmental Science and Technology 53(7):847-864. https://doi.org/10.1080/10643389.2022.2094693
32. Zhao Y, Yang QE, Zhou X, Wang FH, Muurinen J, Virta MP, et al (2020) Antibiotic resistome in the livestock and aquaculture industries: Status and solutions. Critical Reviews in Environmental Science and Technology 51(19):2159–2196. https://doi.org/10.1080/10643389.2020.1777815
33. Jin C, Wei S, Sun R, et al (2020) The Forms, Distribution, and Risk Assessment of Sulfonamide Antibiotics in the Manure–Soil–Vegetable System of Feedlot Livestock. Bulletin of Environmental Contamination and Toxicology 105:790–797. https://doi.org/10.1007/s00128-020-03010-9
34. Gu J, Chen C, Huang X, Mo J, Xie Q, Zeng Q (2021) Occurrence and risk assessment of tetracycline antibiotics in soils and vegetables from vegetable fields in Pearl River Delta, South China. Science of The Total Environment 776:145959. https://doi.org/10.1016/j.scitotenv.2021.145959
35. Li M, Yang L, Yen H, Zhao F, Wang X, Zhou T, Feng Q, Chen L (2023) Occurrence, spatial distribution and ecological risks of antibiotics in soil in urban agglomeration. Journal of Environmental Sciences 125:678–690. https://doi.org/10.1016/j.jes.2022.03.029
36. Zhao F, Chen L, Yang L, Sun L, Li S, Li M, Feng Q (2020) Effects of land use and rainfall on sequestration of veterinary antibiotics in soils at the hillslope scale. Environmental Pollution 260:114112. https://doi.org/10.1016/j.envpol.2020.114112
37. Moles S, Gozzo S, Ormad MP, Mosteo R, Gómez J, Laborda F, Szpunar J (2022) Long-term study of antibiotic presence in Ebro river basin (Spain): identification of the emission sources. Water 14(7):1033. https://doi.org/10.3390/w14071033
38. Bilal M, Mehmood S, Rasheed T, Iqbal HMN (2020) Antibiotics traces in the aquatic environment: persistence and adverse environmental impact. Current Opinion in Environmental Science &Health 13:68–74. https://doi.org/10.1016/j.coesh.2019.11.005

39. Ma N, Tong L, Li Y, Yang C, Tan Q, He J (2022) Distribution of antibiotics in lake water-groundwater - sediment system in Chenhu Lake area. Environmental Research 204(C):112343. https://doi.org/10.1016/j.envres.2021.112343
40. Li F, Chen L, Chen W, Bao Y, Zheng Y, Huang B, Mu Q, Wen D, Feng C (2020) Antibiotics in coastal water and sediments of the East China Sea: distribution, ecological risk assessment and indicators screening. Marine Pollution Bulletin 151:110810. https://doi.org/10.1016/j.marpolbul.2019.110810
41. Xu L, Zhang H, Xiong P, Zhu Q, Liao C, Jiang G (2021) Occurrence, fate, and risk assessment of typical tetracycline antibiotics in the aquatic environment: a review. Science of The Total Environment 753:141975. https://doi.org/10.1016/j.scitotenv.2020.141975
42. Jia WL, Song C, He LY, Wang B, Gao FZ, Zhang M, Ying GG (2023) Antibiotics in soil and water: Occurrence, fate, and risk. Current Opinion in Environmental Science & Health 32:100437. https://doi.org/10.1016/j.coesh.2022.100437
43. Sutandhio S, Alimsardjono L, Wasito E (2018) Antimikroba: Magic Bullet Versus Superbugs. Jurnal Widya Medika 4(1):38-43.
44. Jadhav P, Dabhade M, Girish K (2013) Superbugs: Challenge to Medicinal Chemistry. International journal of pharma and bio sciences 4(4):230-236.
45. Baral B, Mozafari M (2020) Strategic Moves of "Superbugs" Against Available Chemical Scaffolds: Signaling, Regulation, and Challenges. ACS pharmacology & translational science 3(3):373-400. https://doi.org/10.1021/acsptsci.0c00005
46. McKendry R (2012) Nanomechanics of superbugs and superdrugs: new frontiers in nanomedicine. Biochemical Society transactions 40(4):603-608. https://doi.org/10.1042/BST20120082
47. Alpert P (2017) Superbugs: Antibiotic Resistance Is Becoming a Major Public Health Concern. Home Health Care Management Practice 29:130-133. https://doi.org/10.1177/1084822316659285
48. Bassegoda A, Ivanova K, Ramon E, Tzanov T (2018) Strategies to prevent the occurrence of resistance against antibiotics by using advanced materials. Applied Microbiology and Biotechnology 102:2075-2089. https://doi.org/10.1007/s00253-018-8776-0
49. OECD (2018) Stemming the Superbug Tide: Just A Few Dollars More. OECD Health Policy Studies, OECD Publishing https://doi.org/10.1787/9789264307599-en Accessed 25 May 2024.
50. Martínez J (2009) Environmental pollution by antibiotics and by antibiotic resistance determinants. Environmental pollution 157(11):2893-902. https://doi.org/10.1016/j.envpol.2009.05.051
51. Baquero F, Martínez J, Cantón R (2008) Antibiotics and antibiotic resistance in water environments. Current opinion in biotechnology 19(3):260-265. https://doi.org/10.1016/j.copbio.2008.05.006
52. Serwecińska L (2020) Antimicrobials and Antibiotic-Resistant Bacteria: A Risk to the Environment and to Public Health. Water 12(12):3313. https://doi.org/10.3390/w12123313
53. Keen P, Patrick D (2013) Tracking Change: A Look at the Ecological Footprint of Antibiotics and Antimicrobial Resistance. Antibiotics 2:191-205. https://doi.org/10.3390/antibiotics2020191
54. Kümmerer K (2004) Resistance in the environment. The Journal of antimicrobial chemotherapy 54(2):311-320. https://doi.org/10.1093/JAC/DKH325
55. Sanderson H, Fricker C, Brown R, Majury A, Liss S (2016) Antibiotic resistance genes as an emerging environmental contaminant. Environmental Reviews 24:205-218. https://doi.org/10.1139/ER-2015-0069
56. Pruden A, Larsson DGJ, Amézquita A, Collignon P, Brandt KK, Graham WD, Lazorchak JM, Suzuki S, Silley P, Snape JR, Topp E, Zhang T, Zhu YG (2013) Management Options for Reducing the Release of Antibiotics and Antibiotic Resistance Genes to the Environment. Environmental Health Perspectives 121:878-885. https://doi.org/10.1289/ehp.1206446
57. Bungau S, Tit DM, Behl T, Aleya L, Zaha DC (2021) Aspects of excessive antibiotic consumption and environmental influences correlated with the occurrence of resistance to antimicrobial agents. Current Opinion in Environmental Science & Health 19:100224. https://doi.org/10.1016/j.coesh.2020.10.012
58. Larson E (2007) Community factors in the development of antibiotic resistance. Annual review of public health 28:435-447. https://doi.org/10.1146/ANNUREV.PUBLHEALTH.28.021406.144020
59. Kim S. Aga D (2007) Potential Ecological and Human Health Impacts of Antibiotics and Antibiotic-Resistant Bacteria from Wastewater Treatment Plants. Journal of Toxicology and Environmental Health Part B 10:559 - 573. https://doi.org/10.1080/15287390600975137
60. Yu X, Sharma V, Li H (2019) Environmental Antibiotics and Antibiotic Resistance: From Problems to Solutions. Frontiers of Environmental Science & Engineering 13(3):47. https://doi.org/10.1007/s11783-019-1137-0
61. WHO (2017) 'One Health'. https://www.who.int/health-topics/one-health#tab=tab_1 Accessed 28 May 2024.
62. Mora-Gamboa MPC, Rincón-Gamboa SM, Ardila-Leal LD, Poutou-Piñales RA, Pedroza-Rodríguez AM, Quevedo-Hidalgo BE (2022) Impact of Antibiotics as Waste, Physical, Chemical, and Enzymatical Degradation: Use of Laccases. Molecules 27:4436. https://doi.org/10.3390/molecules27144436

63. Pan M, Chu M (2016) Adsorption and degradation of five selected antibiotics in agricultural soil. Science of the Total Environment 545-546:48-56. https://doi.org/10.1016/j.scitotenv.2015.12.040
64. Ezzariai A, Hafidi M, Khadra A, Aemig Q, El Fels L, Barret M, Merlina G, Patureau D, Pinelli E (2018) Human and veterinary antibiotics during composting of sludge or manure: Global perspectives on persistence, degradation, and resistance genes. Journal of Hazardous Materials 359:465-481. https://doi.org/10.1016/j.jhazmat.2018.07.092
65. Can L, Li T, Liming Z, Weiqian T, Lanqing M (2021) A Review of the Distribution of Antibiotics in Water in Different Regions of China and Current Antibiotic Degradation Pathways. Frontiers in Environmental Science 9:692298. https://doi.org/10.3389/fenvs.2021.692298
66. Mao F, Liu X, Wu K, et al (2018) Biodegradation of sulfonamides by Shewanella oneidensis MR-1 and Shewanella sp. strain MR-4. Biodegradation 29:129–140. https://doi.org/10.1007/s10532-017-9818-5
67. Liyanage G Y, Manage P M (2018) Removal of Ciprofloxacin (CIP) by bacteria isolated from hospital effluent water and identification of degradation pathways. International Journal Of Medical, Pharmacy And Drug Research 2(3):37-47. https://dx.doi.org/10.22161/ijmpd.2.3.1
68. Sun M, Luo Y, Teng Y, Jia Z, Li Z, Deng S (2013) Remediation of polycyclic aromatic hydrocarbon and metal-contaminated soil by successive methyl-β-cyclodextrin-enhanced soil washing–microbial augmentation: a laboratory evaluation. Environmental Science and Pollution Research 20:976-986. https://doi.org/10.1007/s11356-012-1064-0
69. Ye M, Sun M, Wan J, Fang G, Li H, Hu F, Jiang X, Kengara F (2015) Enhanced soil washing process for the remediation of PBDEs/Pb/Cd-contaminated electronic waste site with carboxymethyl chitosan in a sunflower oil–water solvent system and microbial augmentation. Environmental Science and Pollution Research, 22:2687-2698. https://doi.org/10.1007/s11356-014-3518-z
70. Haslmayr H, Meißner S, Langella F, Baumgarten A, Geletneky J (2014) Establishing best practice for microbially aided phytoremediation. Environmental Science and Pollution Research 21:6765-6774. https://doi.org/10.1007/s11356-013-2195-7
71. Chmelová D, Ondrejovič M, Miertuš S (2024) Laccases as Effective Tools in the Removal of Pharmaceutical Products from Aquatic Systems. Life 14(2):230. https://doi.org/10.3390/life14020230
72. Becker D, Giustina S V D, Rodriguez-Mozaz S, et al (2016) Removal of antibiotics in wastewater by enzymatic treatment with fungal laccase – Degradation of compounds does not always eliminate toxicity. Bioresource Technology 219:500-509. https://doi.org/10.1016/j.biortech.2016.08.004
73. Bilal M, Ashraf S, Barceló D, Iqbal H (2019) Biocatalytic degradation/redefining "removal" fate of pharmaceutically active compounds and antibiotics in the aquatic environment. The Science of the total environment 691:1190-1211. https://doi.org/10.1016/J.SCITOTENV.2019.07.224
74. Yang, L, Hu D, Liu H, Wang X, Liu Y, Xia Q, Deng S, Hao Y, Jin Y, Xie M (2021) Biodegradation pathway of penicillins by β-lactamase encapsulated in metal-organic frameworks. Journal of hazardous materials 414:125549. https://doi.org/10.1016/j.jhazmat.2021.125549
75. Chen Z, Liu X, Chen L, Han Y, Shen Y, Chen B, Wang M (2023) Deglycosylation Inactivation Initiated by a Novel Periplasmic Dehydrogenase Complex Provides a Novel Strategy for Eliminating the Recalcitrant Antibiotic Kanamycin. Environmental science & technology 57(10):4298-4307. https://doi.org/10.1021/acs.est.2c09565
76. Chen J, Wei X, Liu Y, Ying G, Liu S, He L, Su H, Hu L, Chen F, Yang Y (2016) Removal of antibiotics and antibiotic resistance genes from domestic sewage by constructed wetlands: Optimization of wetland substrates and hydraulic loading. The Science of the total environment 565:240-248. https://doi.org/10.1016/j.scitotenv.2016.04.176
77. Chen J, Ying G, Wei X, Liu Y, Liu S, Hu L, He L, Chen Z, Chen F, Yang Y (2016) Removal of antibiotics and antibiotic resistance genes from domestic sewage by constructed wetlands: Effect of flow configuration and plant species. The Science of the total environment 571:974-82. https://doi.org/10.1016/j.scitotenv.2016.07.085
78. Liu X, Guo X, Liu Y, Lu S, Xi B, Zhang J, Wang Z, Bi B (2019) A review on removing antibiotics and antibiotic resistance genes from wastewater by constructed wetlands: Performance and microbial response. Environmental pollution 254 Pt A, 112996. https://doi.org/10.1016/j.envpol.2019.112996
79. Zhang X, Cai T, Zhang S, Hou J, Cheng L, Chen W, Zhang Q (2024) Contamination distribution and non-biological removal pathways of typical tetracycline antibiotics in the environment: A review. Journal of Hazardous Materials 463:132862. https://doi.org/10.1016/j.jhazmat.2023.132862
80. Palacio DA, Becerra Y, Urbano BF, Rivas BL (2020) Antibiotics removal using a chitosan-based polyelectrolyte in conjunction with ultrafiltration membranes. Chemosphere 258:127416. https://doi.org/10.1016/j.chemosphere.2020.127416
81. Liang C, Wei D, Zhang A, Ren Q, Shi J, Liu L (2021) Removal of antibiotic resistance genes from swine wastewater by membrane filtration treatment. Ecotoxicology and Environmental Safety 210:111885. https://doi.org/10.1016/j.ecoenv.2020.111885

82. Homem V, Santos L (2011) Degradation and removal methods of antibiotics from aqueous matrices-A review. Journal of Environmental Management 92(10):2304-2347. https://doi.org/10.1016/j.jenvman.2011.05.023
83. Ganiyu S, Hullebusch E, Cretin M, Esposito G, Oturan M (2015) Coupling of membrane filtration and advanced oxidation processes for removal of pharmaceutical residues: A critical review. Separation and Purification Technology, 156:891-914. https://doi.org/10.1016/J.SEPPUR.2015.09.059
84. Wang J, Zhuan R (2019) Degradation of antibiotics by advanced oxidation processes: An overview. The Science of the total environment 701:135023. https://doi.org/10.1016/j.scitotenv.2019.135023
85. Li J, Ren S, Qiu X, Zhao S, Wang R, Wang Y (2022) Electroactive Ultrafiltration Membrane for Simultaneous Removal of Antibiotic, Antibiotic Resistant Bacteria, and Antibiotic Resistance Genes from Wastewater Effluent. Environmental science & technology 56(21):15120-15129. https://doi.org/10.1021/acs.est.2c00268
86. Attour A, Touati M, Tlili M, Ben Amor M, Lapicquec F, Leclerc JP (2014) Influence of operating parameters on phosphate removal from water by electrocoagulation using aluminum electrodes. Separation and Purification Technology 123:124–129. https://doi.org/10.1016/j.seppur.2013.12.030
87. Griche NB, Attour A, Mostefa MP, Guesmi S, Tlili M, Lapicque F (2019) Fluoride removal from water by electrocoagulation: effect of the type of water and the experimental parameters. Electrochimica Acta 316:257–265. https://doi.org/10.1016/j.electacta.2019.05.130
88. Mokni S, Tlili M, Jedidi N, Hassen A (2022) Applicability of electrocoagulation process to the treatment of Ofloxacin and Chloramphenicol in aqueous media: Removal mechanism and antibacterial activity. Journal of Water Process Engineering 49:103080. https://doi.org/10.1016/j.jwpe.2022.103080
89. Shahedi A, Darban AK, Taghipour F, Jamshidi-Zanjani A (2020) A review on industrial wastewater treatment via electrocoagulation processes. Current Opinion in Electrochemistry 22:154–169. https://doi.org/10.1016/j.coelec.2020.05.009
90. Al-Raad AA, Hanafiah MM (2021) Removal of inorganic pollutants using electrocoagulation technology: a review of emerging applications and mechanisms. Journal of Environmental Management 300:113696. https://doi.org/10.1016/j.jenvman.2021.113696
91. Oladipo AA, Mustafa FS, Ezugwu ON, Gazi M (2022) Efficient removal of antibiotic in single and binary mixture of nickel by electrocoagulation process: Hydrogen generation and cost analysis. Chemosphere 300:134532. https://doi.org/10.1016/j.chemosphere.2022.134532
92. Saad MS, Balasubramaniam L, Wirzal MDH, Abd Halim NS, Bilad MR, Md Nordin NAH, Adi Putra Z, Ramli FN (2020) Integrated Membrane–Electrocoagulation System for Removal of Celestine Blue Dyes in Wastewater. Membranes 10(8):184. https://doi.org/10.3390/membranes10080184
93. Lu J, Zhang W, Zhang X, Si G, Zhang P, Li B, Su R, Gao X (2021) Efficient removal of Tetracycline-Cu complexes from water by electrocoagulation technology. Journal of Cleaner Production 289:125729. https://doi.org/10.1016/j.jclepro.2020.125729
94. Alam R, Sheob M, Saeed B, Khan SU, Shirinkar M, Frontistis Z, Basheer F, Farooqi IH (2021) Use of Electrocoagulation for Treatment of Pharmaceutical Compounds in Water/Wastewater: A Review Exploring Opportunities and Challenges. Water 13(15):2105. https://doi.org/10.3390/w13152105
95. Ensano B, Borea L, Naddeo V, Belgiorno V, Luna M, Balakrishnan M, Ballesteros F (2019) Applicability of the electrocoagulation process in treating real municipal wastewater containing pharmaceutical active compounds. Journal of hazardous materials 361:367-373. https://doi.org/10.1016/j.jhazmat.2018.07.093
96. Butler E, Hung YT, Yeh RYL, Suleiman Al Ahmad M (2011) Electrocoagulation in Wastewater Treatment. Water 3(2):495-525. https://doi.org/10.3390/w3020495
97. Baran W, Adamek E, Jajko M, Sobczak A (2018) Removal of veterinary antibiotics from wastewater by electrocoagulation. Chemosphere 194:381-389. https://doi.org/10.1016/j.chemosphere.2017.11.165
98. Benjamin OO, Busisiwe NZ, Babatunde AK, Luthando T, Gbenga MP, Nonhlangabezo M, Minghua Z, Omotayo AA (2020) Solar photoelectrocatalytic degradation of ciprofloxacin at a FTO/$BiVO_4$/ MnO_2 anode: Kinetics, intermediate products and degradation pathway studies. Journal of Environmental Chemical Engineering 8:103607. https://doi.org/10.1016/j.jece.2019.103607
99. Peleyeju MG, Arotiba OA (2018) Recent trend in visible-light photoelectrocatalytic systems for degradation of organic contaminants in water/wastewater. Environ. Sci. Water Res. Technol. 4:1389–1411. https://doi.org/10.1039/C8EW00276B
100. Vinoth V, Sivasankar T, Asiri AM, Wu JJ, Kaviyarasan K, Anandan S (2018) Photocatalytic and photoelectrocatalytic performance of sonochemically synthesized $Cu_2O@TiO_2$ heterojunction nanocomposites. Ultrason Sonochem 51:223–229. https://doi.org/10.1016/j.ultsonch.2018.10.022
101. Martins AS, Cordeiro-Junior PJM, Bessegato GG, Carneiro JF, Zanoni MVB, de V. Lanza MR (2019) Electrodeposition of WO_3 on Ti substrate and the influence of interfacial oxide layer generated in situ: a photoelectrocatalytic degradation of propyl paraben. Appl Surf Sci 464:664–672. https://doi.org/10.1016/j.apsusc.2018.09.054

102. Liu H, Yang W, Wang L, Hou H, Gao F (2017) Electrospun $BiVO_4$ nanobelts with tailored structures and their enhanced photocatalytic/photoelectrocatalytic activities. Cryst Eng Comm 19:6252–6258. https://doi.org/10.1039/C7CE01478C
103. Zhang M, Pu W, Pan S, Okoth OK, Yang C, Zhang J (2015) Photoelectrocatalytic activity of liquid phase deposited α-Fe_2O_3 films under visible light illumination. J Alloys Compd 648:719–725. https://doi.org/10.1016/j.jallcom.2015.07.026
104. Reddy KR, Reddy CV, Nadagouda MN, Shetti NP, Jaesool S, Aminabhavi TM (2019) Polymeric graphitic carbon nitride (g-C_3N_4)-based semiconducting nanostructured materials: synthesis methods, properties and photocatalytic applications. J Environ Manage 238:25–40. https://doi.org/10.1016/j.jenvman.2019.02.075
105. Mishra A, Mehta A, Basu S, Shetti NP, Reddy KR, Aminabhavi TM (2019) Graphitic carbon nitride (g–C_3N_4)–based metal-free photocatalysts for water splitting: a review. Carbon 149:693–721. https://doi.org/10.1016/j.carbon.2019.04.104
106. Li Y, Zhang C, Zhao G, Su P, Wang J, et al (2024) A critical review on antibiotics removal by persulfate-based oxidation: Activation methods, catalysts, oxidative species, and degradation routes. Process Safety and Environmental Protection 187:622-643. https://doi.org/10.1016/j.psep.2024.05.001
107. Zhu K, Li X, Chen Y, Huang Y, Yang Z, Guan G, Yan K (2024) Recent advances on the spherical metal oxides for sustainable degradation of antibiotics. Coordination Chemistry Reviews 510:215813. https://doi.org/10.1016/j.ccr.2024.215813
108. Singh J, Palsaniya S, Soni RK (2020) Mesoporous dark brown TiO_2 spheres for pollutant removal and energy storage applications. Applied Surface Science 527:146796. https://doi.org/10.1016/j.apsusc.2020.146796
109. Liu R, Shi Y, Lin L, Wang Z, Liu C, Bi J, Hou Y, Lin S, Wu L (2022) Surface Lewis acid sites and oxygen vacancies of Bi_2WO_6 synergistically promoted photocatalytic degradation of levofloxacin. Applied Surface Science 605:154822. https://doi.org/10.1016/j.apsusc.2022.154822