A REVIEW ON INSIGHTS INTO ANTIBIOTICS ADSORPTION ON MICROPLASTICS: INTERACTIONS, CHALLENGES, AND REMOVAL METHODS IN AQUEOUS ENVIRONMENTS

A DISSERTATION

SUBMITTED IN PARTIAL FULFILLMENT OF THE REQUIREMENTS FOR THE AWARD OF THE DEGREE

MASTER OF SCIENCE

IN

CHEMISTRY

Submitted by:

Priynka Yadav (2K22/MSCCHE/28)

Under the supervision of

DR. RAMINDER KAUR

DEPARTMENT OF APPLIED CHEMISTRY

DELHI TECHNOLOGICAL UNIVERSITY

(Formerly Delhi College of Engineering)

Bawana Road, Delhi-110042

MAY, 2024

DELHI TECHNOLOGICAL UNIVERSITY

(Formerly Delhi College of Engineering)

Bawana Road, Delhi-110042

CANDIDATE'S DECLARATION

I, Priynka Yadav (2k21/MSCCHE/28) students of M.Sc. (Applied Chemistry), hereby declare that the project Dissertation titled "A Review on Insights into Antibiotics Adsorption on Microplastics: Interactions, Challenges, and Removal Methods in Aqueous Environments" which is submitted by me to the Department of Applied Chemistry, Delhi Technological University, Delhi in partial fulfilment of the requirement for the award of the degree of Master of Science, is original and not copied from any source without proper citation. This work has not previously formed the basis for the award of any Degree, Diploma Associateship, Fellowship or other similar title or recognition.

Place: Delhi **Priynka Yadav**

Date: 31th, May 2024

DELHI TECHNOLOGICAL UNIVERSITY

(Formerly Delhi College of Engineering)

Bawana Road, Delhi-110042

CERTIFICATE

I hereby certify that the Project Dissertation titled “Insights into Antibiotics Adsorption on Microplastics: Interactions, Challenges, and Removal Methods in Aqueous Environments" which is submitted by [Priynka Yadav, 2k22/MSCCHE/28] student of M.Sc. (Applied Chemistry) Delhi Technological University, Delhi in partial fulfilment of the requirement for the award of the degree of Master of Science, is a record of the project work carried out by the students under my supervision. To the best of my knowledge this work has not been submitted in part or full for any Degree or Diploma to this University or elsewhere.

Place: Delhi

Date: 31th, May 2024

DR. RAMINDER KAUR

SUPERVISOR

ABSTRACT

Two newly discovered contaminants that are currently often found in the aquatic environment are microplastics and antibiotics. When antibiotics and microplastics combine, combination contamination occurs, which could have detrimental effects on living organisms. This article provides a thorough analysis of the relationship between microplastics and antibiotics. There is also an explanation of the environmental effects of the composite pollutants. Our knowledge of the interactions between antibiotics and microplastics can be strengthened by this review. Provide basic information to evaluate the combined toxicity of both newly emerged pollutants and provide insight into the locations of these contaminants in the aqueous medium. Furthermore, after desorption, it looks at new and existing ways for removing antibiotics and microplastics from water, emphasizing sophisticated treatment procedures such membrane filtering, advanced oxidation processes, and adsorption. In order to effectively address the dual contamination of microplastics and antibiotics in aqueous settings, the review highlights the necessity for integrated approaches combining physical, chemical, and biological methodologies. In order to ensure safer water quality and environmental sustainability, future research directions are also suggested to improve the understanding and management of these contaminants.

ACKNOWLEDGMENT

I wish to express my deep sense of gratitude and veneration to my project Supervisor**, Dr. Raminder Kaur, Department of Applied Chemistry, Delhi Technological University**, Delhi or his perpetual encouragement, constant guidance, valuable suggestions and continued motivation which has enabled me to complete this project work.

I wish to thank **Prof. Anil Kumar, Head of the Department of Applied Chemistry, Delhi Technological University** for his encouragement.

I would like to continue by thanking Mrs, Manisha Yadav (Research scholars), for massive help in making me understand the basics of techniques and patiently responding to my all the doubts.

Ultimately, and perhaps most importantly, I would like to thank our family and friends from the bottom of my hearts for their constant support throughout the Project.

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CHAPTER 1
INTRODUCTION

In the marine environment, microplastic particles have existed since the early 1970s [1]. Because plastic can be used for so many things, its use has increased dramatically since the 1950s [2] In modern society, the adoption of plastics has largely superseded natural polymers due to their extended durability, chemical stability, and cost-effectiveness. However, the ubiquity of synthetic polymers and the resulting proliferation of plastic waste, once heralded as a technological blessing, has transformed into a global predicament. The situation deteriorates as a result of their harder natural breakdown, This finally causes plastic to accumulate in almost all environments [3]. The National Oceanic and Atmospheric Administration (NOAA) in the United States and the European Chemicals Agency both define microplastics as plastic particles smaller than five millimeters. Although the majority of studies on microplastics have historically concentrated on their occurrence in aquatic settings, it is now becoming more evident that these tiny particles can be found in a variety of environmental media, including soil, clean water, water in the ground, air, and sediment. Thus, it is becoming more and more important to look into the presence and effects of microplastics in a variety of settings. Furthermore, it is obvious that studies on the size of these microplastics are required because they are very likely to be toxic, even though the harmful consequences of the microparticles on human health remain unknown [4]. Due to their minute dimensions, and microplastics are easy to consumed by organisms and become integrated into food web. Human tissues (such as blood) and excreta are also affected by the buildup, in addition to marine organisms. Animals' growth, reproduction, metabolism, and immune systems will all be harmed by the buildup of specific microplastic amounts in their bodies. In smaller sizes, microplastics can get through the blood-brain barrier and enter particular parts of the brain, which could have a significant negative effect on human health. Therefore, there is a serious risk to human health and the integrity of ecosystems when microplastics accumulate in aquatic environments. [5]. Antibiotics play a crucial role in combating bacterial infections by either eradicating or impeding bacterial growth, thereby fostering animal growth and enhancing feed efficiency [6]. The projected daily rate per 1,000 individuals increased by 46% from 9.8 defined daily doses (DDD) to 14.3 DDD between 2000 and 2018, reflecting a substantial increase in antibiotic consumption worldwide [7]. Central Asia and eastern Europe were expected to have the highest levels of consumption, while sub-Saharan Africa had the lowest levels. Owing to the guts of humans and animals' low ability to absorb antibiotics, between 30% and 90% of parent medicines were eliminated through feces [6]. In China in 2013, the environment was exposed to over 99 percent of the antibiotics expelled by humans as well as animals, with an estimated 54,000 tons going to waste [8]. Growing human population, economic growth, and easy access to medications are the main causes of the global increase in antibiotic consumption. Additional growth promoters and antibiotics are needed to meet the increased demand for animal protein in food production. Freshwater algae, fish, zooplankton, microphytes, and macrophytes are among the aquatic habitats that these antibiotics may adversely affect [9]. It is important to note that the ongoing SARSCov-2 pandemic may have contributed to the rise in antibiotic use globally [10]. Antibiotics are typically administered to treat bacterial co-infections, even though COVID-19 is a viral illness [11]. The presence of antibiotic residues in the environment may

create serious risks to ecosystems and human health.[12]. furthermore have the potential to cause the rise and spread of antibiotic-resistant genes and microorganisms, which have the potential to transform into infections and travel from the natural world into human bodies, posing more dangerous risks [13]. Antibiotics in aquatic environments are problematic because low quantities can harm aquatic creatures and cause resistance. The Nairobi River Basin has been shown to have high concentrations of antibiotics, including sulfamethoxazole [14]. Animals excrete 30 to 90% of antibiotics due to their water solubility and limited gastrointestinal absorption. Up to 90% of them are eliminated in urine and 75% in feces due to their water solubility. Antibiotics in animal feces break down in the disposal of animal waste, decreasing their bactericidal effect and encouraging the proliferation of multi-resistant bacteria. Furthermore, antibiotics damage aquatic life and contaminate the ecosystem [15]. Due to antibiotic overuse, antibiotic resistance genes (ARGs) are becoming more and more prevalent, garnering global concern. ARGs possess a lengthy half-life and possess the capability to horizontally transfer among nearby bacteria, posing significant ecological risks. They are prevalent across diverse environments including rivers, sediments, and wastewater treatment facilities (WWTPs) [16]. Microplastics (MPs) possess a significant specific surface area, characterized by their minute particle size, hydrophobic nature, and limited degradability. This unique combination enables them to efficiently adsorb and transport antibiotics as well as various organic contaminants. This makes microplastic superior transporters of hydrophobic organic compounds [17]. MPs have the ability to both absorb and transfer ATs, changing the toxicity of both MPs and ATs separately. In blood clams, for instance, it has been observed that PS increases the bioaccumulation of florfenicol and oxytetracycline (OTC) [18]. The interplay between MPs and ATs in these activities is crucial to the overall toxicity to aquatic life [16]. Aquaculture systems are heavily contaminated by microplastics and antibiotics, which are prevalent in aquatic habitats. When these contaminants interact, they produce composite pollution, as microplastics carry antibiotics and their harmful combinations. Microplastic aging and environmental factors regulate the antibiotics' adsorption-desorption process on microplastics. Antibiotics are concurrently transferred by the digestion of microplastics, leading to bioaccumulation and subsequent amplification throughout the food chain [5]. The absorption of antibiotics on microplastics needs particular attention because antibiotics are part of the environmental chain that spreads antibiotic resistance [19]. Microplastics and antibiotics coexist in the ecosystem and may interact in a variety of ways. This raises the prospect that microplastics may be more likely than antibiotics to travel through waterways. It is crucial to comprehend the spread and transformation of interactions between antibiotics and microplastics in order to prevent and manage combination pollution. The most significant mechanism by which microplastics interact with antibiotics is adsorption, and in freshwater systems, different forms of micro plastics can adsorb different antibiotics. Microplastics and antibiotics would coexist in aquatic settings and have a wide range of effects on aquatic creatures, including microorganisms that are resistant to antibiotics and are chemically hazardous. Adsorption of organic compounds such as polychlorinated biphenyls and trichloroethane by microplastics is possible in aquatic environments [20]. This review aims to compile the most recent research findings about the processes behind the interactions between antibiotics and microplastics in aquatic settings. We will investigate physicochemical characteristics that drive these interactions, the ecological and health risks that these combined pollutants supply, and the efficiency of the available removal techniques. This review aims

to provide a thorough picture in order to guide future research and policy-making efforts that will address the combined hazards that microplastics and antibiotics pose to our water systems.

1.1. THE PRESENCE AND IMPACT OF ANTIBIOTICS IN AQUATIC ECOSYSTEM.

1.1.1. Sources of antibiotics in the Aqueous Environment.

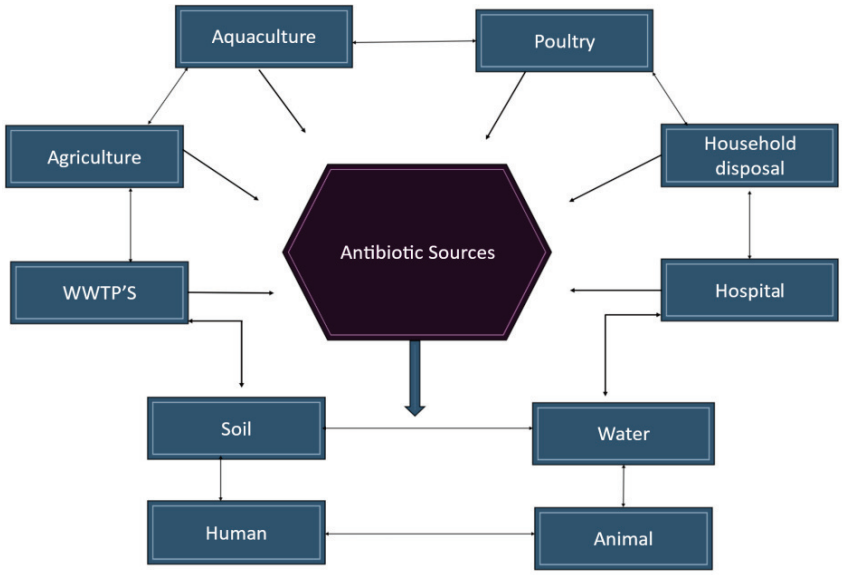

Fig 1 Showing source of antibiotics.

Antibiotics and their derivatives are always present in the natural environment. The release of animal excrement, veterinary trash, pharmaceutical plants, dairies, domestic rubbish, livestock husbandry, and municipal trash all raise the risk of antibiotic pollution. Encouragement for the expansion of fish farming, beekeeping, and livestock rearing is fueled by the prevalent use of antibiotics in agriculture. However, this practice leads to environmental contamination through the discharge of antibiotic remnants and byproducts present in chicken manure. Antibiotics such as Monensin play a role in enhancing animal growth and feed efficiency, thereby aiding dairy farms in producing milk that closely resembles natural resources. Therefore, water resources are indirectly contaminated by antibiotic residue [21]. However, plants use extremely little antibiotics as compared to animals. The presence of antibiotics in agricultural area streams is indicative of their widespread usage within agriculture [22]. Additionally, runoff and drain flow may carry them to ditches, streams, and rivers. Leaching may carry them to groundwater, and they may even make their way into the food chain [23]. The release of waste pollutants stemming from manufacturing processes, accidental spills occurring during production or distribution, and the improper disposal of outdated or expired medications pose considerable threats to environmental integrity [24]. The continual presence of these substances might subject bacteria to selective pressures, potentially fostering the development of resistance. Within the influent of sewage treatment facilities, there exists a concentration of antibiotics introduced by pharmaceutical entities, alongside effluents from residential and industrial sources [25]. Antibiotics are frequently found in all

environmental matrixes due to their manufacturing, usage, and improper disposal [26].

1.1.2. Effect of antibiotic on environment.

To mitigate the accumulation of antibiotics in aquatic ecosystems, safeguarding both human well-being and ecological balance, it is imperative to address the dissemination of antibiotic-resistant genes alongside environmental degradation. The convergence of antibiotics with pathogens, facilitating their ingress into the human system, poses significant risks. Furthermore, antibiotics exert detrimental impacts on the development and reproductive cycles of marine organisms, potentially compromising liver function. The continual presence of these substances might subject bacteria to selective pressures, potentially fostering the development of resistance. Within the influent of sewage treatment facilities, there exists a concentration of antibiotics introduced by pharmaceutical entities, alongside effluents from residential and industrial sources. Therefore, effective regulation of antibiotic utilization in aquatic environments is essential to uphold public health and mitigate ecological damage [27]. The long-term consequences of antibiotics are more detrimental than their short-term ones, and their presence in sludge and wastewater can also result in resistance [28]. The significant toxicity of antibiotic residues to algae and daphnids has been seen in natural habitats; 20% and 44% of antibiotics, respectively, are highly toxic to these species. Various types of antibiotics, such as tetracyclines, sulfonamides, and macrolides, have the potential to negatively impact the growth and maturation of algae. Abscisic acid secretion that can be induced, protein biosynthesis suppression, and disruption of chloroplast formation are examples of growth-inhibiting processes. Exposure to rapamycin can cause uninflated swim bladders, yolk sac edema, and behavioral changes in zebrafish, embryos, and larvae [29]. Antibiotics possess capacity to impact bacterial colonies present in wastewater networks. Antibiotics also suppress the activity of bacteria in sewage treatment systems, which can be detrimental to the decomposition of organic molecules [30],[31]. Antibiotics have the potential to significantly impact the breakdown of organic matter by modifying the population of bacteria in sediments both quantitatively and qualitatively. The sulfate reduction process can be impacted by the concentration and aggregation of antibiotic chemicals in sediments, which can inhibit the sulfate reducing bacteria's ability to proliferate and function. Antibiotics' impact on sediments is crucial to take into account because of the concentration and aggregation of these anti-bacterial substances in the sediments at the bottom of fish farming sites [32].

1.2.THE PRESENCE AND IMPACT OF MICROPLASTIC IN AQUATIC ECOSYSTEM.

1.2.1. Sources of Microplastic.

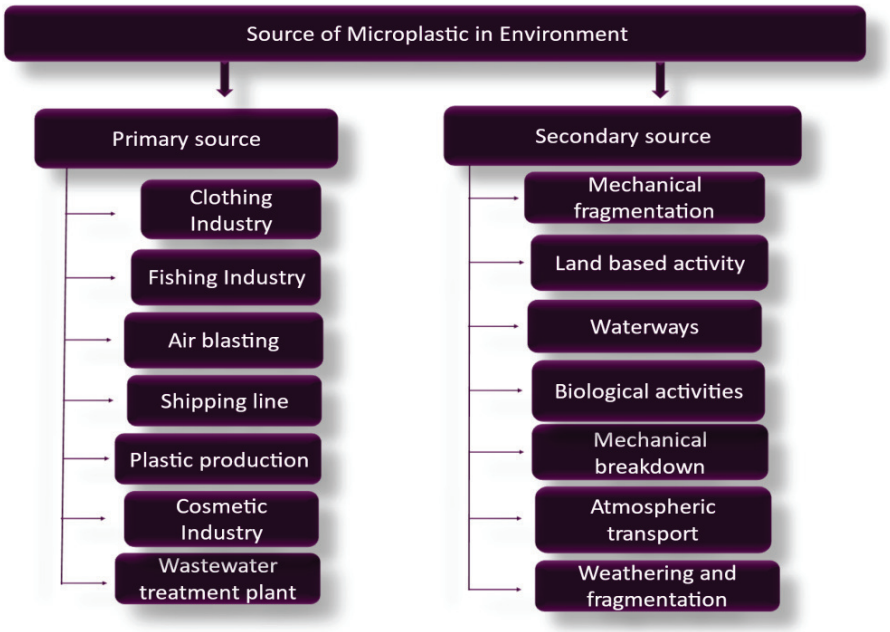

Fig 2 Showing source of Microplastics.

It's essential to comprehend the origins of microplastics (MPs) to mitigate their potential adverse impacts on the environment [33]. Primary MPs are intentionally manufactured for various purposes, while secondary MPs originate from the deterioration of textile and tire microfibers, along with macro- and meso-plastic fragments. MPs are predominantly terrestrial in nature, attributed to their role in commercial fishing and littering, constituting more than 80% of the plastic waste discovered in the ocean [34]. Wastewater treatment facilities serve as the primary sources of microplastics (MPs), encompassing household items like cosmetics, exfoliants, cleaning agents, and textiles, as well as urban activities such as paint erosion, tire wear, and the plastic/textile sectors [35]. Textile microfibers discharge atmospheric microplastics into the atmosphere, where they might deposit and end up on land, in rivers, and in the ocean. MP paths to marine ecosystems should take poorly managed plastic garbage into account. The primary sources of soil pollution are landfills and plastic films; further downstream, wastewater treatment plant (WWTP) sewage sludge contributes to MPs [34].

1.2.2. Microplastics effect on the environment.

Microplastics pose a severe risk because of how easily they consumed and spread throughout the food web. Though it is unknown how much of an impact there will be on public health and aquatic environments, a growing number of reports point to detrimental effects on freshwater and marine biota. When microplastic trash is ingested by organisms through their food, it can induce bioaccumulation, which can lead to problems with the immune system, digestive tract, respiratory and circulatory systems, organ failure, impotence, and even death. Humans and other species have been demonstrated to be negatively impacted by microplastic

accumulations, with injuries occurring to planktons, copepods, zooplanktons, crabs, small fish, turtles, fish larvae, seabirds, and mammals [36]. Depending on the particle size, these microparticles can get past biological barriers by phagocytosis and persorption, two endocytosis processes. The animal may die, exhibit altered behavior, or experience a false feeling of fullness if microplastics obstruct its digestive system. Because of its vulnerability, the animal is more exposed to predators. Furthermore, microplastics can damage cells, DNA, generate oxidative stress, trigger immunological responses, and inflame the body [37]. Microplastics are ingested by all marine organisms, according to studies, although the amount depends on the species. MPs pose a threat to marine biota, thus it's critical to keep an eye out for their excessive use and to implement regulations and guidelines to control the sources of plastic litter [38]. Microplastics have been linked to a host of detrimental consequences on human health, including toxicity, disturbance of hormones, and gastrointestinal issues. Gastrointestinal issues pose a significant concern linked with exposure to microplastics, leading to potential inflammation, digestive irregularities, disturbances in gut flora, and changes in intestinal barrier function. In the digestive tract, microplastic accumulation causes physical discomfort and obstructions. The hormonal balance, reproductive health, development, and general well-being can all be negatively impacted by microplastics' disruption of the endocrine system. Furthermore, by drawing bacteria and other species, they can serve as a vector for pathogens and promote the development of intricate biofilms. Diseases can spread by contaminating food chains and water sources with these biofilms. When pathogenic bacteria and microplastics are consumed, they can build up in the digestive system and lead to infections or inflammatory reactions. There is evidence linking certain pathogenic microorganisms on microplastics to skin disorders, respiratory infections, and gastrointestinal disorders in human beings. Investigations are necessary to determine the presence and spread of these harmful bacteria on aquatic microplastics, as evidenced by a Hong Kong study that discovered bacterial populations developed biofilms on polyethylene microbeads in raw sewage [39].

CHAPTER 2

EXPLORING THE INTERACTION OF MICROPLASTIC-ANTIBIOTIC ADSORPTION, DESORPTION AND THEIR IMPACT ON ENVIRONMENT.

Aquatic habitats are seriously threatened by two new types of water pollution: MPs and antibiotics. Each year, a lot of antibiotics are discharged into aquatic habitats. China alone was responsible for the environmental release of 53,800 tons of antibiotics in 2013 [40]. MPs can be divided more generally into veterinary and medical categories. Because MPs have such detrimental impacts on the microbial ecology, researchers have focused their efforts on this topic worldwide [41]. In recent years, there has been a growing focus on investigating the sorption behaviors of antibiotics onto microplastics, driven by concerns about their potential environmental impact. To find out if the ease of disintegration affects the sorption capacity, researchers have also looked into less prevalent plastics that are thought to be "biodegradable," including polylactic acid (PLA) [42].The sorption of ciprofloxacin and tetracycline onto microplastics has been extensively researched. Tetracycline is the antibiotic that is produced and consumed the most in China and the second most internationally [43].

2.1. Sorption kinetic.

Sorption onto microplastics is accelerated by studies on sorption kinetics, which also provide equilibrium timeframes and concentrations. These investigations may provide insight into the possible natural behavior of microplastics and antibiotics. Because they can transfer higher concentrations, interactions between antibiotics and microplastics may be more dangerous in cases where there is a higher sorption capacity. Eighty percent of antibiotics achieve equilibrium in less than 48 hours, with antibiotic equilibrium concentrations ranging from three hours to six days. Although other researches have looked into this, higher initial antibiotic doses might not have an impact on equilibrium time. Adsorption kinetics delineates the correlation between the quantity of adsorption (qt) and the duration of contact (t). This yields crucial insights into the rate of adsorption. Antibiotics absorbed by microplastics have a very lengthy equilibrium period (teq), as Table 1 demonstrates. It typically requires more than two or three hours. days to accomplish the adsorption equilibrium [44]

Table 1. Studies on the kinetics and isotherm of antibiotic adsorption onto microplastics.

Antibiotics	Types of Microplastics	The Kinetic models	The Isotherm models	References
Cephalosporin C/SMX/SMT	Polyethylene (PE) and Polystyrene (PS)	MO	Linear > Freundlich (F), Langmuir (L)	[45]

SMX	Polyamide (PA), Polyethylene Terephthalate (PET), Polystyrene (PS), Polyvinyl Chloride (PVC), Polypropylene (PP)	EMRT>IMRT, AAS	Linear, Freundlich > Langmuir	[46]
Sulfonamides	Thermoplastic Polyurethane (TPU)	Pseudo-First Order (PFO) > Pseudo-Second Order (PSO)	Linear~F	[47]
TC	Nylon	PSO > PFO, IDM	Linear ~ L ~ F	[48]
NOR	PS, Polybutylene Succinate (PBS), PE	PSO > PFO, Intraparticle Diffusion Model (IDM)	L > F > Linear	[49]
SDZ/AMX CIP/TMP/TC	PVC, PE, PS, PP, and PA	Not Available (N.A.)	Linear	[50]

Typically, the pseudo-second-order kinetic model is used to explain the adsorption mechanism, as Table 1 shows. However, the pseudo-first-order method may offer superior conformity in specific circumstances. Significantly, the adsorption kinetics of SMT on PA, PE, PVP, and PP were better represented by the pseudo-first-order model, while the adsorption kinetics of SMT on PS and PET were best fitted by the pseudo-second-order model [45]. These scenarios highlight the theoretical significance and specific application requirements of each model. A more extensive model, encompassing elements from both the pseudo-first-order and pseudo-second-order models, has been developed [52]. Furthermore, a mixed order model—a broader version of the pseudo-first-order and pseudo-second-order models—has been created [53]. Mass transfer at the exterior and internal levels as well as sorption at active sites are all components of adsorption processes. Adsorption is multistage, and this is described by phenomenological mathematical
models and kinetic models such as intraparticle diffusion. The predominant factor hindering the rate of transfer is identified as peculiar mass transfer, as indicated by phenomenological models. Meanwhile, intraparticle diffusion models solely address the initial adsorption of CIP onto PS microplastics [44].

2.2. Adsorption isotherms.

To understand the mechanisms and processes involved in antibiotic sorption, researchers use sorbent isotherm models to examine factors such as maximal sorption concentration. The most studied adsorption isotherm models include the Linear, Freundlich, and Langmuir models. Table 1 highlights that antibiotics and microplastics exhibit different types of optimal sorption isotherms [50]. When analyzing data on antibiotic adsorption equilibrium, the linear model is the mathematical approach most frequently employed [54]. Table 1 presents findings from several studies that observed a significant linear relationship in the adsorption of various antibiotics onto different microplastics. The adsorption behavior of SMT and SMX onto five types of microplastic polymers is well-represented by the linear model, exhibiting partition coefficients between 11.5 and 38.7 L kg−1 for SMT and from 22.2 to 284 L kg−1 for SMX. Similar results were reported for the TC adsorption isotherms on nylon microplastic [47]. Adsorbate dispersion between solid and liquid phases is reflected by linear models. liquid phases resulting from hydrophobic, van der Waals, and electrostatic interactions. Several studies have investigated the nonlinear isotherms of Temkin, Freundlich, and Langmuir. The Langmuir isotherm describes chemical adsorption in a single layer, while the empirical Freundlich isotherm is suitable for both chemical adsorption and multi-layer physical adsorption at 50% coverage. In contrast, the Temkin isotherm explains multi-layer sorption where the interaction heat decreases linearly with an increase in coverage rate. Table1 demonstrates the fitting accuracy of the Freundlich model to various antibiotic adsorption data on microplastics, comparing it with the Langmuir and Temkin isotherms [44].While starting values of 0.2-50 mg/L are used in most investigations, Antibiotics can be present in the environment at concentrations ranging from nanograms per liter (ng/L) to micrograms per liter (µg/L). To determine the maximum sorption capacity of microplastics, experiments often utilize high doses of antibiotics. This approach helps to understand the environmental behavior of three different antibiotics [55]. looked at the variations in PE's sorption capacity. The antibiotic sorption capacity of weathered microplastics was found to be higher in older microplastics, as demonstrated by studies [51] and [42]. Future research should focus on the interaction between antibiotics and microplastics in natural streams, considering the complexities of natural matrices in their experimental approaches [43].

2.3. Desorption of antibiotics from microplastic.

Many researchers have studied desorption and factors affecting desorption of antibiotics from microplastics. Xiulei Fan et. al. his study investigates how antibiotics desorb from microplastics (MPs) in different environments, specifically comparing simulated intestinal fluid and Milli-Q water. Polylactic acid (PLA) and PVC, both in their original and aged forms, were examined to understand the adsorption-desorption behavior of tetracycline (TC) and ciprofloxacin (CIP). It was observed that the desorption of MPs was more pronounced in simulated intestinal fluid than in Milli-Q water, attributed to the presence of intestinal surfactants. Additionally, degradable PLA demonstrated a higher desorption capacity compared to PVC, indicating a greater potential threat to aquatic ecosystems and their inhabitants. The damage caused by microplastics to aquatic life is significant, and the ageing process exacerbates this impact. Xiulei's findings highlight the increased risk posed

by PLA in aquatic environments due to its higher desorption rates [56]. Miguel pleiter found that macrolide antibiotics Azithromycin and clarithromycin desorb from microplastic PEC, PLA and polystyrene(PS) in presence of cyanobacterial culture, which leads to toxicity [57]. The release or desorption of antibiotics is significantly influenced by temperature and various other environmental parameters, including as pH, density, and salinity, might impact the kinetic rates of antibiotics release. These factors can effectively increase or decrease the rates of antibiotic release depending on the kind of polymer. Furthermore, the rate of antibiotic release from UV radiation is significantly influenced by the presence or absence of UV stabilization in a polymer chain [58].

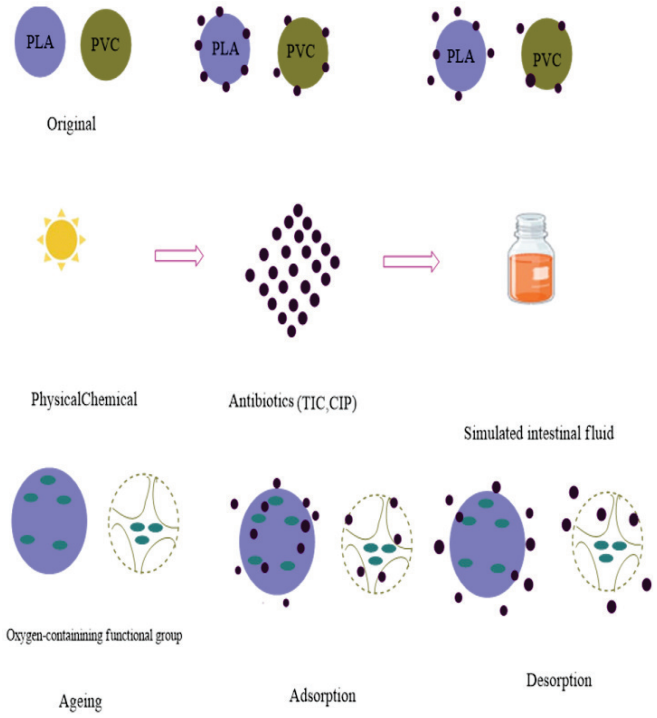

Fig 3 Showing desorption of antibiotics from microplastics.

2.4. Mechanisms of microplastic-antibiotic compound interactions.

The two main ways that the antibiotics-microplastics combination interacts are through adsorption and desorption. Antibiotics work by a variety of mechanisms, including hydrophobic, Van der Waals and π-π interactions and so on.

2.4.1. Hydrophobic interactions:

Non-polar compounds exhibit an affinity for each other in polar environments such as water, facilitated by hydrophobic interactions. This phenomenon is particularly relevant in the context of microplastics, where materials like PS, PE, PP, and PET dominate the landscape. Consequently, hydrophobic interactions emerge as a prevalent mechanism governing sorption interactions [59].Hydrophobic antibiotics have a natural affinity for hydrophobic microplastics, resulting in the adsorptive removal of the microplastics as a means of reducing water contact. Conversely, the repulsive forces between hydrophilic antibiotics and hydrophobic microplastics are primarily governed by hydrophobic interactions [60]. Studies on antibiotics have shown significant partitioning to PE as a result of hydrophobic interactions [19]. Researchers noted that variances in the hydrophobic characteristics of sulfamethazine, sulfamethoxazole, and cephalosporin C impacted their adherence to PE. They noted a trend where the antibiotics' hydrophobicity decreased in this sequence: cephalosporin C > sulfamethoxazole > sulfamethazine. Due to PE's inherent hydrophobic characteristics, the antibiotics adhered to it in a manner consistent with their hydrophobic nature, with cephalosporin C exhibiting the least absorption and sulfamethoxazole displaying the highest absorption rates [61]. High log kow hydrophobic antibiotics should have a high affinity for MP. [45] Some researchers have proposed that the interaction between the compounds and polyethylene (PE) microparticles might be influenced by their hydrophobic nature [62]. An investigation on the sorption of tylosin onto PS highlighted the importance of hydrophobic interactions. The significant sorption rates reported are the result of an interaction between the hydrophobic layer on of PS and the hydrophobic components of tylosin, also [63].

2.4.2. Hydrogen bonding Interaction.

Antibiotics containing amine, hydroxyl, and carboxyl groups possess the capability to form hydrogen bonds with carbonaceous substances [64]. One alternative approach to combating, involves investigating the adsorption mechanisms on MPs. Proton donors were the amide groups in PA. When it came to AMX, TC, and CIP, PA outperformed PE, PP, and PVC in terms of adsorption capacity. However, the carbonyl compounds in CIP, TC, and AMX served as proton acceptors, which allowed the antibiotics to establish hydrogen bonds with them [16]. Hydrogen bonding interactions are observed between steroidal hormones such as 17β-estradiol and various antibiotics like ciprofloxacin, amoxicillin, tetracycline, and sulfamethoxazole, as well as with PA. This interaction is particularly notable during the process of plastic surface weathering which introduces oxygen-containing groups onto PS, PP, and PVC surfaces. These groups encourage hydrogen bonding with triclosan, ciprofloxacin, and oxytetracycline. These groups include the carboxyl group, ester, and ketone groups. As a result, this phenomenon controls how PPCP compounds sorb on old microplastics [65]. A more detailed study is necessary to identify the exact atoms or molecular groups in polyethylene (PE), polypropylene (PP), and polyvinyl chloride (PVC) that contribute to hydrogen bond formation. The presence of a 532.8 eV subpeak in the X-ray photoelectron spectroscopy spectra, which signifies −OH/−O−C−O, suggests a potential hydrogen bond between chlorinated polyethylene (CIP) and polystyrene (PS) [66].

2.4.3. Electrostatic interactions.

There are two types of electrostatic forces: repulsion and attraction. When molecules have opposite charges, they attract each other electrostatically. Conversely, molecules with similar charges repel each other. A number of variables, including pH-dependent interactions, affect how well antibiotics adsorb onto microplastics. The type of electrostatic forces that exist between antibiotics and microplastics depends on their charged states. The pH of the solution, the existence of coexisting ions or dissolved organic matter, the point of zero charge of microplastics, the pKa values of antibiotics, and other factors all affect this process. Tetracycline (TC), for instance, can exist in both neutral and cationic forms in acidic environments, while microplastics made of polyethylene (PE) have negative zeta potential [55]. Both PVC and polystyrene (PS) become negatively charged when the surrounding solution's pH falls to less than 7.1. These microplastics (MPs) attract tylosin through electrostatic interactions caused by their negative charge [67]. Furthermore, coexisting ions or dissolved organic matter (DOM) can affect the electrostatic characteristics of microplastics and antibiotics. For example, when humic acid is applied to the surface of polyethylene (PE) microplastics, the contact forces between ciprofloxacin (CIP) and microplastics shift from electrostatic repulsion to attraction. [68] Ciprofloxacin (CIP) was primarily found in its cationic form in freshwater environments with pH values between 6.7 and 7.1. This condition improved adsorption by increasing the electrostatic interaction between the cationic CIP and the negatively charged microplastics (MPs). Antibiotics (ATs) exhibited the maximum adsorption onto microplastics (MPs) when present in their cationic or zwitterionic forms, thereby diminishing electrostatic repulsion [69]. The adsorption capacity of microplastics (MPs) for the majority of adsorbates (ATs) increases initially and subsequently diminishes as the pH of the solution rises [16]. As the pH level rises, there is a reduction in adsorption attributed to the transformation of solutes from cations to anions. Moreover, the presence of oxygen-containing functional groups like carboxyl, ester, and ketone groups on a polymer leads to plastic degradation. Interactions characterized by dipole–dipole or dipole–induced forces between polymers and pharmacologically active polar groups are also noted. However, in the instance of non-polar polymers like PE, these interactions are less prominent [69].

2.4.4 Van der Waals interactions.

Through π–π interactions and van der Waals forces, microplastics (MPs) influence the sorption of antibiotics [70]. Through these processes, aliphatic and aromatic polymers can create non-covalent linkages. Because polystyrene (PS) has greater π-π interactions than polyethylene (PE), PS absorbs antibiotics including ciprofloxacin, trimethoprim, and sulfadiazine more effectively [19]. One popular technique for determining π–π interactions and hydrogen bonding during the adsorption process is to look at the structures of both adsorbents and adsorbates. Although weak, these interactions frequently take place between aromatic structures that are parallel to one another, such as PET and PS. Antibiotics commonly include aromatic compounds. Hydrogen bonding and π–π interactions are supported by methods such as FT-IR, XPS, molecular dynamics simulations, and DFT computations. These techniques are helpful for researching the interfacial processes that take place

when antibiotics are adsorbed by microplastic adsorbents. The results show that Van der Waals interactions predominate in the adsorption process of SMT-microplastics systems, whereas electrostatic interactions play a more important role in the adsorption of SMT on PA, PS, and PVC. The significance of nonbonding interactions is shown by these findings [44]. π-π interactions are suggested to be the main mechanism for MP adsorption with benzene rings. The π-π conjugation force generated by the benzene ring is responsible for PS's greater affinity for TC in TC adsorption when compared to PE and PP [71].

2.4.5 Pore-filling interactions.

Near their solubility limits, glassy and rubbery polymers experience interactions that lead to the filling of their nano and micropores with contaminants [72][73]. Liu et al. [75] identified pore filling and partition effects as the primary mechanisms behind ciprofloxacin adsorption on PS and PVC. Their research demonstrated that these processes dominate the adsorption behavior. Additionally, they found that the diffusion rate onto a plastic surface is influenced by the pore diameter and the size of the diffusing molecule. Pores with larger diameters facilitate quicker and more extensive molecule occupancy compared to smaller pores, resulting in a gradual reduction in sorption rate due to pore-filling interactions [74]. In situations where weathering has taken place, pore-filling gives the plastic the ability to produce additional pores on its surface, which is crucial for the interactions between microplastic and pollutants [75].

Table 2. Antibiotics and virgin MPs' interactions.

Microplastic Type	Size	Concentration	Antibiotics Studied	Medium	Interaction Types	References
Polyamide (PA)	75–180 μm	0.5, 1, 5, 10, and 15 mg/L	Tetracycline, Amoxicillin, Ciprofloxacin, Sulfadiazine, and Trimethoprim	Filtered seawater or ultrapure water	Electrostatic interaction, hydrogen bonding, and surface properties	[70]
Polyethylene (PE)	28, 48, 125, 250, and 590 μm	0,5 mg/Ml	Tetracycline	Solution	Surface properties and hydrogen bonding	[76]

Polyethylene (PE)	150–425 µm	10 mg/Ml	Chlortetracycline hydrochloride Oxytetracycline hydrochloride and Tetracycline		Electrostatic attraction, van der Waals force and hydrogen bonding	[55]
Various (PET, PVC, PP, PS, PA, PE)	100–150 µm	2 mg/mL	Sulfamethazine	Distilled water	Electrostatic and Van der Waals interaction	[77]
Polyethylene (PE)	150 µm	5 mg/mL	Sulfamethoxazole	Solution (0.01 mol/L CaCl2 And 200 mg/L NaN3)	Hydrophobic, electrostatic, and van der Waals interactions	[78][79]
Various (PET, POM, PS)	3–5 mm	50 mg/mL	Clarithromycin and Azithromycin	Milli-Q water	Hydrophobic interaction	[57]

Table 3- Antibiotics and aged MPs' interactions.

MPs Types	Size range	Concentration	Antibiotics studied	Medium	Interaction Types	Reference
Nylon	1 mm	2,5 mg/mL	Tetracycline	Distilled water	Van der Waals forces	[48]
Polyvinyl chloride (PVC), Polypropylene (PP), Polystyrene (PS), and Polyethylene (PE)	~74 µm	0,2 - 1,2 mg/mL	Tylosin	Milli-Q water (0.001 mol/L NaN3	Electrostatic attraction, hydrophobic interaction, and surface complexations	[67]
Polystyrene (PS)	40 µm	0,5 mg/mL	Ciprofloxacin	Deionized water	Hydrophobic, π π interaction, electrostatic	[66]

					hydrogen bonding	
Polyvinyl Chloride (PVC), Polystyrene, (PS)	~75 µm	0.4 g/L	Ciprofloxacin		Partitioning, hydrogen bonding, bonding, cation exchange, π-π interactions, electrostatic attraction	[80]

2.5. Factors influencing the way antibiotics adsorb on microplastics.

It is crucial to consider the physicochemical features of MPs when determining the extent of adsorption interaction with antibiotics. [82][50]. Microplastics' (MPs') properties, which range greatly and affect how pollutants cling to various MP kinds, include their polarity, rubber-like areas, and independent functional groups [83]. It's generally known that polyethylene (PE) has a greater affinity for organic contaminants than polypropylene (PP) and polyvinyl chloride (PVC) [84]. MPs' interactions with antibiotics affect their capacity to absorb medications. For instance, hydrophilic antibiotics like SMX are more attractive to polar MPs like polyamide (PA) [83]. Similarly, out of PS, PP, and PE, PVC showed the largest sorption capacity for tylosin [67] Interactions between the functional groups of pollutants and microplastics (MPs) and π-π interactions, in particular, have a considerable impact on the sorption process. In polyamide (PA) and polypropylene (PP), for example, polar functionalities such as -CO-NH- and -CH3 are important in promoting electrostatic interactions with polar contaminants [77]. Furthermore, to improve sorption, functional groups in PVC, such as -Cl, act as electron acceptors [85]. Furthermore, PVC demonstrates a preference for both polar and non-polar antibiotics [70]. Another important element influencing MPs' adsorption behavior is crystallinity [86]. Amorphous, semicrystalline, and crystalline polymers can be distinguished from one another by the way their molecular chains are arranged. Organic pollutants have been seen to have a strong adherence to amorphous polymers [87]. As crystallinity decreased, the crystalline MPs' ability to adsorb organic contaminants increased gradually [88]. The amount of the polymer's non-crystalline area determines the equilibrium concentration and rate of organic material adsorption on plastic polymers [89] However, because amorphous areas have free volume and empty spaces within polymers, which allow molecules to migrate and transfer quickly, they are more favorable for adsorption [90]. Laboratory experiments have demonstrated that microplastics with low crystallinity exhibit varying affinities for hydrophobic organic contaminants, with polyethylene (PE) showing the highest affinity, followed by polypropylene (PP), then polyamide (PA), and finally polystyrene (PS) [91]. Furthermore, the model prediction results indicate that the rubbery state of the amorphous region is where adsorption and desorption are most likely to occur, as opposed to the glassy state [87]. Antibiotic adsorption capacity of microplastics is determined by both their size and shape. Microplastics have a higher specific surface area when they get smaller. As a result, this augmentation offers more adsorption sites, increasing their potential for

adsorption. [92]. Macarena et al. found a relationship between the reduction of particle size and the adsorption of antibiotics in their investigation of the capacity for adsorption of 20–1000 μm PS-MPs on metronidazole (MNZ). Similar trends were observed for other antibiotic compounds such as 1,3,6,8-tetrabromocarbazole (1,3,6,8-BCZ) and 3,6-dibromocarbazole (3,6-BCZ). However, the chemical properties of small particles render them susceptible to agglomeration, leading to a decrease in specific surface area and a consequent reduction in the number of effective adsorption sites for organic contaminants. Particle size and agglomeration work together to influence MPs' adsorption capacity [87]. Antibiotic-microplastic interactions have been found to be significantly influenced by surface area [19]. Surface area may be influenced by the particle's shape. Because microplastics feature pores and cracks from weathering or biodegradation, antibiotics have been seen to accumulate on their surface. These microplastics can have a larger surface area. Zhang et al. [75] An investigation on the sorption capacity of PS foam revealed that samples taken from north Chinese beaches had much greater rates of oxytetracycline sorption than virgin PS foam. The final factor influencing MPs' adsorption ability is their specific surface area; particle size only matters in a limited range [86]. Aging is the phenomena wherein MPs' performance deteriorates with time due to artificial or natural factors such temperature, water corrosion, and UV radiation[93][94][95]. Common microplastics' surface shape is significantly altered by this, leading to a significant number of wrinkle, pieces, and grooves. Additionally, its microstructure—which includes functional groups like carboxyl groups, aldehyde groups, hydroxyl groups, and carbon-carbon double bonds—will alter. The procedure modifies the surface potential and crystallinity of microplastics, which eventually alters how they behave in the environment [96] [97] In order to evaluate thermoplastic polyurethanes (TPUs) and polyamides (PAs) for their potential to adsorb sulfonamide antibiotics (SAs), Jiang et al. aged the materials using UV light and UV light mixed with hydrogen peroxide (UV + H2O2). [47] The findings indicated that the older TPU-MPs and PA-MPs had larger oxygen contents than the original MPs, increasing between 11.43% and 12.01% to 14.96% and 23.53%, respectively. This demonstrated that MPs underwent oxidation and destruction as they aged [93]. Aging MPs alter surface charge and hydrophobic properties, which affect antibiotic adsorption capacity. Having a greater water-octanol partition coefficient (KOW), The original MPs are more hydrophobic and have the capacity to enhance organic pollutants. Functional groups that include oxygen become more intense with age, strengthening their polar and hydrophilic characteristics. The electronegativity of MPs is also increased with age. PLA-MPs and PVC-MPs aged by UV radiation both operate better as antibiotic transporters, despite having less zeta potential than PVCMPs. The adsorption of ciprofloxacin and tetracycline is increased by 2.19 and 1.18 times, respectively, with time [87]. Different MPs with and without degradation shown different antibacterial adsorption capacities. Compared to nondegradable MPs, degradable MPs had a greater affinity for antibiotics. According to Jiang et al., degradable PBS-MPs had an approximately twofold higher adsorption capacity for norfloxacin (NOR) than non-degradable PE-MPs [49]. Nevertheless, there aren't many relevant investigations, therefore further research is required to validate this [59].

2.6. Environmental Factors Impacting Antibiotics Adsorption onto Microplastics.

The following is a summary of the primary variables that may impact antibiotics' ability to bind to microplastics.

2.6.1. Effect of pH

Antibiotics are clearly affected by the pH of the solution when they are sorbent by microplastics, but the particular antibiotic/microplastic combination also affects this process. This concept is demonstrated quite well by Puckowski et al. [98] work, which uses norfloxacin to sorb onto LDPE, HDPE, PP, and PVC. While the highest sorption of norfloxacin in PP and PVC occurs at pH 12, that of LDPE and HDPE occurs at pH 4 and 2, respectively. The fact that the results vary according to pH suggests that the polymer surface charge is essential to the sorption antibiotics.

In contrast, when the pH value increases from 7 to 11, the ability of nanoscale PS particles to adsorb CIP simila rly decreases [99]. The quantity of TCS on the PS reduces when pH rises from 4 to 11 because of the electrostatic repulsion between TCS and the PS surface at pH 10. The highest possible adsorption capacity [49]. At pH<7, electrostatic attraction is the primary cause of the tylosin and PS/PVC adsorption response[67]. Furthermore, it was discovered that pH had little influence on the interaction when sulfamethoxazole's acid effect on polyethylene was investigated. The sorption balance was not considerably impacted by pH increase above 8 because both PE microplastics and sulfamethoxazole were negatively charged and electrostatic repulsion was not a major factor in the sorption process. [79]. It is necessary to carefully investigate how pH value affects other ionizable antibiotics in addition to studying its hydrophobicity (the presence of oxygen-containing functional groups) and electrostatic interactions because there aren't many studies with comparable findings [100].

2.6.2. Salinity.

Salinity affects the way mixed pollutants sorb; by competitive adsorption, salinity ions take up active sites on microplastics. Antibiotic adsorption is inhibited by sodium ions that adsorb onto microplastics' surface when salinity rises. By means of electrostatic attraction, sodium ions can take the place of acidic groups on the surfaces of microplastics, thereby diminishing the sorption effect. Research has indicated that the hydrophilic chemical compounds norfloxacin and ciprofloxacin are not able to adsorb on salt. Antibiotic sorption characteristics are greatly influenced by the inherent structure and surface area of microplastics. Salinity might not have a major effect on how antibiotic contaminants and microplastics interact, though. There hasn't been any clear evidence of a salinity influence on sulfamethoxazole adsorption on PE microplastics in research [101]. Research indicates that the sorption capacity of microplastics is greatly affected by the various combinations of electrolytes, adsorbents, adsorbates, and the chemistry of the solution [102]. Ciprofloxacin's ability to sorb onto PVC gradually diminished as the concentration of NaCl rose from 8.8% to 35% [80]. Studies have demonstrated that at varying NaCl concentrations (10%, 20%, and 35%), the absorption of

sulfamethazine and sulfamethoxazole on microplastics including PE, PS, PA, PVC, PP, and PET changes. The presence of salinity can either enhance or inhibit the adsorption of antibiotics onto microplastic surfaces. Studies indicate that antibiotic adsorption on microplastics (including PP, PS, PE, and PVC) increases slightly at a salinity of 0.01 M before decreasing again. In order to limit the possibility that microplastics would serve as antibiotic carriers and the likelihood that they will serve as breeding grounds for bacteria and genes resistant to antibiotics (ARBs), various ex-situ remediation strategies may be developed by simulating higher salinity conditions [63].

2.6.3. Dissolved organic matter.

Medical contaminants and microplastics may interact differently in natural water bodies when dissolved organic stuff is present. At higher concentrations, antibiotics and microplastics generally exhibit a greater tendency to interact with each other, which diminishes their adsorptive efficiency due to competitive adsorption. Alternatively, organic macromolecules might facilitate the adsorption of antimicrobial pollutants onto microplastics. Humic acid (HA) has been observed to significantly diminish the affinity of ciprofloxacin for polyethylene (PE) microplastics [60]. But HA might have also aided in the microplastics PS's adsorption with oxytetracycline [75]. Natural organic matter is mainly composed of fulvic acids and humic acids. There are various ways in which they can be adsorbed on microplastics. Antibiotics that are cationic or zwitterionic may be more electrostatically attracted to conjugated co-polymers. The adsorption capability of CIP by PS nanoplastics was enhanced, for example, by the increasing electrostatic attraction with CIP+ and PS nanoplastics/humic acid. However, due to the spatial obstruction caused by humic acid and the clustering of nanoplastics, negative effects were observed at elevated concentrations of humic acid [66]. It was also discovered that fulvic acid (FA) had very little effect on the way that sulfamethoxazole and polyethylene (PE) interacted [79].

2.6.4. Temperature.

Because their molecules travel more slowly on solid surfaces during the exothermic process of adsorption, heat is released. By reducing equilibrium and raising rate, temperature has an impact on adsorption. Certain adsorption mechanisms, however, are unable to quickly reach equilibrium at low temperatures, which results in enhanced adsorption. Wu and associates observed that heat absorption caused the adsorption of the antibiotics fluoroquinolone and tetracycline to increase as temperature rise[87]. Chen and Ai et al. discovered similar outcomes while examining the adsorption behavior of lipopeptide antibiotics (LAs) and sulfonamide antibiotics (SAs) [103][104]. Microplastics and medicinal pollutants sorb well in the presence of temperature. Higher temperatures don't improve tetracycline's adsorption onto high-density polyethylene and ordinary polystyrene. [105]. The same findings as the previous research are found in the study on the impact of temperature on PS and ciprofloxacin adsorption [99]. As the temperature rises, the equilibrium capacity might not drop as it did in the cases of PE and PVC, where the temperature actually increased the adsorption capacity of bisphenol A. The characteristics and makeup of microplastics and antibiotic pollutants are closely related to this phenomenon.

[106].

2.7. The impact of the interaction between microplastic and antibiotics on aquatic ecosystems.

By serving as carriers, MPs alter the spatial distribution of these drugs, leading to heightened levels of environmental pollution [87]. Antibiotics are transferred into the organism by the swallowed MPs, creating a synergistic contamination effect [107]. The study of chemical toxicity to aquatic creatures is significantly influenced by this effect [16]. According to Tang et al [102]. The combined pollution could pose a greater hazard and lead to negative consequences if aquatic organisms ingest microplastics that have absorbed antibiotics. The possible effects that microplastics containing antibiotics may have on living things can be better understood by conducting research on a variety of contaminants. For example, Stollberg et al. studied the effects on the tissues of blue mussels' digestive glands [108]. Researchers examined microplastics contaminated with fluoranthene, a type of polycyclic aromatic hydrocarbon. As a result of the microplastics' contamination, the researchers discovered that the blue mussels exposed to these microplastics had considerably greater amounts of fluoranthene in their tissues than the mussels in the control group. According to Feng et al.[109] Tetracycline adsorption decreased the electrostatic repulsion between the negatively charged polystyrene microplastics (PS-SO3H) and marine unicellular diatoms. As a result, the microplastics' acute toxicity rose over the course of a day, and the connections between the particles and cells improved. Furthermore, some microorganisms may become hazardous due to the antibiotics present on microplastics. Zhu and colleagues. [110] examined the effects of growth suppression and oxidative stress to evaluate the combined toxicity of Triclosan (TCS) and four distinct forms of microplastics on the microalgae Skeletonema costatum. According to their research, TCS and every kind of microplastic (toxicity ranked PVC800 > PVC > PS > PE) severely impeded the microalgae's ability to proliferate. PVC800 was identified as the most impactful microplastic. When combined with PVC and PVC800, TCS is less harmful than when combined with PE and PS. This discrepancy can result from TCS's higher adsorption ability on PVC and PVC800. Tetracycline adsorption, according to Feng et al.[109], the adsorption of tetracycline enhances the surface hydrophobicity of positively charged polystyrene microplastics (PS-NH2) , leading to reduced toxicity and diminished cell-particle interactions [111]. Furthermore, MPs work as "sinks" in the environment for bacteria and genes resistant to antibiotics (ARBs) [70]. Zhang et al. [112] Research indicates that the dense biofilms formed by antibiotic-resistant bacteria (ARBs) on microplastic (MP) surfaces lead to a concentration of ARBs that is 100 to 5000 times greater than in the surrounding water. In aquatic environments, microplastics can form unique microbial communities and selectively accumulate both intracellular and extracellular antibiotic resistance genes [115]. Harmful compounds are released into the bodies of aquatic creatures when they eat microplastics (MPs) including antibiotic-resistant genes (ARGs) and anthropogenic toxins (ATs). This process causes contaminants from the environment to migrate into the body, having a negative compound effect. [116]. Besides, the coexistence of MPs and ARGs (Antibiotic Resistance Genes) intensifies the genetic exchange of antibiotic-resistant bacteria (ARB) in the ecosystem. This phenomenon leads to a significant portion of ARB acquiring immunity to various Antibiotic Treatments (ATs), thereby fostering the evolution of single-gene expression

resistance into multi-drug resistance [117]. In ecological scenarios characterized by the prevalence of microplastic biofilms, the well-being of organisms experienced notable repercussions. This observation was underscored by Wang et al. [118] who noted that microplastic biofilms exhibited elevated concentrations of ARGs and heightened levels of pathogenic metabolic pathways compared to naturally occurring minerals. Moreover, PS containing zinc, magnesium, and copper was discovered to prevent microalgae from growing. [119], and the development of zebrafish embryos was adversely affected by PS containing cadmium [120]. The molecular-level interaction between microplastics and antibiotics remains largely unexplored, despite recent research efforts. The potential synergistic effects of microplastics and antibiotics on toxicity could be influenced by their interactions. Due to their insensitivity when compared to traditional physiological measures, metabolic assessments have the potential to provide light on the intricate mechanisms behind the toxicity of microplastics. Using metabolomic analysis and cellular reactions, you and your colleagues investigated the individual and combination toxicity of polystyrene microplastics and CIP. By generating ROS, microplastics prevented cell survival, and CIP interfered with nucleotide metabolism. An antagonistic impact based on particle size was seen in the CIP adsorption on microplastics [121]. Antimicrobial resistance (ARGs), which increases bacterial resistance, can arise from improper use of antibiotics (ATs). Aquatic ecosystems coexist with microplastics (MPs) and an abundance of ARGs, with MPs having the ability to affect ARG type and abundance. Via the food chain, MPs can infiltrate aquatic creatures, endangering both human health and aquaculture. In addition, MPs have the ability to selectively enhance particular pathogens in waste leachate, influence the effectiveness of UV disinfection in riverine environments, and block UV beams in wastewater treatment. [122]. It is known as in aquatic environments, interactions between MPs and ARGs can have harmful consequences and alter community structure. In aquatic settings, MPs have been found to enrich alternative rhizomes (ARGs), contribute to ARG contamination, proliferation, and accumulation, and activate a negative feedback loop that intensifies ARGs' pathogenic potential and ecological effects. The majority of current studies on the toxicity of microplastics (MPs) and antibiotics (ATs) focus on these compounds separately. The effects of concurrent exposure to MPs and ATs on the environment, however, are still poorly known. Because of the intricate and varied ways in which these pollutants interact with ecosystems, a deeper examination of their individual effects as well as their interactions is necessary. Over all though, there has been little and inadequate research done to clarify the underlying mechanism.

CHAPTER 3

METHODS TO REMOVE MICROPLASTIC AND ANTIBIOTICS SEPARATELY.

3.1. Technique used to remove microplastic from aqueous water.

To get rid of microplastics from water, several methods have been devised [123].
Physical removal methods and advanced treatment techniques are the two general categories into which these operations may be separate

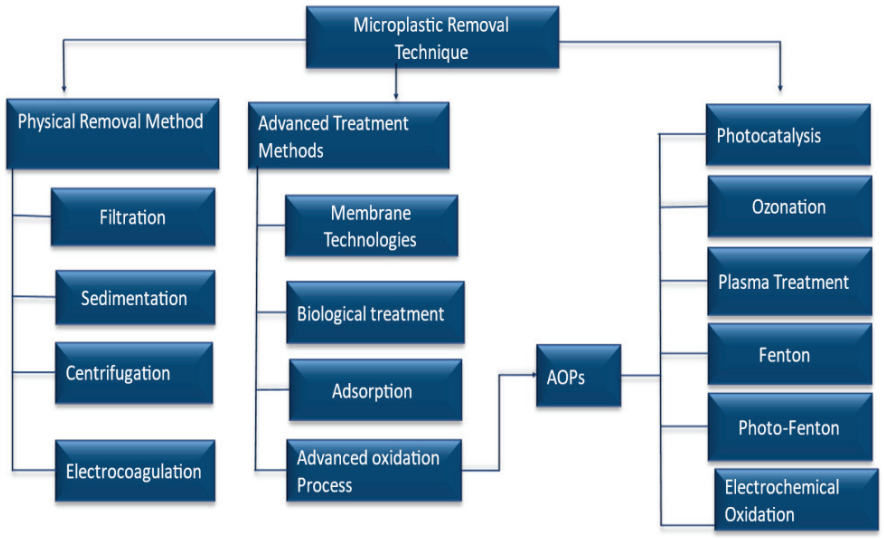

Fig 5 Showing Removal method of microplastics.

3.1.1. Physical removal method.

Physical techniques for eliminating microplastics involve methods or procedures aimed at physically extracting or segregating microplastic particles from diverse surroundings. These approaches typically leverage the inherent physical properties of the particles, including dimensions, mass, and movement patterns, to effectuate the separation of microplastics [124]. Filtration serves as a prevalent, straightforward method for eliminating microplastics (MPs) from both wastewater and water sources. A disc filter made up of thirteen polyester mesh discs with an 18 µm pore size was shown to be effective in eliminating 89.7% of MPs larger than 10 µm in a 2019 study by Simon et al. However, it was noted that the accumulation of large plastic particles on the filter surfaces led to pore blockage, thereby reducing the efficiency of the filtration process. Similar findings were

reported by Talvitie et al. in 2017, indicating a filtration efficacy ranging from 40% to 98.5%. Another promising filtration method involves the utilization of fast sand filters, which have demonstrated effectiveness in trapping various MPs within the sand grains. Sand layers with a maximum size of grains of 35 mm are usually used in these filters. For example, 0.5 mm of quartz and 1 mm of gravel are used. Notably, this approach is particularly suitable for removing MPs larger than 20 μm, offering a practical, cost-effective solution for MP removal [125]. Microplastics, when present in a container or bodies of water, possess the capability to accumulate at the seabed or lake floor as sediment. The density differential between microplastics and water is the basis for this technique. To improve the settling process, flocculants or coagulants might be added. Numerous academic research have examined the process of sedimentation as a means of removing microplastics from water. Shen et al. [126] discovered that microplastics may be effectively removed from wastewater by electrocoagulation (EC) with an aluminum anode. They attained elimination rates more than 80% in each of their studies. In their investigation of electrocoagulation techniques, Elkhatib et al.[127] studied electrocoagulation techniques and discovered that up to 99% of micro plastics could be successfully eliminated by varying the pH and current density in synthetic solutions. Similarly, Akarsu et al. [128] used membrane filtering in conjunction with the electrocoagulation-electroflotation (EC/EF) process to obtain removal efficiencies of 100% across a range of polymer types. Xue and associates showed that raising the alum dosages frequently resulted in more effective elimination of microspheres small than 90 μm [129]. throughout the drinking water treatment procedures to remove carboxylated polystyrene microspheres. Lee and Jung ultimately explored the coagulation technique as a solution for removing microplastics from seawater. They attained remarkable clearance efficiencies exceeding 60% through the utilization of a variety of coagulants [124]. In centrifugation, microplastics are separated according to density by rapidly spinning water. The clearance of the microplastics is aided by their movement towards the outer edge due to centrifugal force [130]. Murray and colleagues looked into how centrifugation affected the wastewater treatment process's ability to remove nanoplastics. Their findings revealed that variations in centrifuge speed, duration, and sample volume significantly enhanced the elimination efficiency. Longer centrifuge periods resulted in a more gradual rate of growth, reaching 99 ± 1% after 10 minutes of centrifugation. An alternate method for sampling microplastics is suggested: continuous flow centrifugation, which provides size- and density-selective sampling, effective removal of microplastics, and volume reduction [124]. Through the use of an electric current, microplastic particles are destabilized and aggregated in this manner. After that, the agglomerated microplastics are easily removed using filtration or sedimentation techniques [131][132]. Several studies have looked into how well electrocoagulation (EC) removes microplastics and have pinpointed important elements that lead to high removal rates. Notably, it has been demonstrated that using aluminum anodes is more effective than using iron anodes for the elimination of microplastics [126]. Additionally, a pH of 7.2 might result in a removal effectiveness of over 90% [127], and fiber microplastic removal works better than granular microplastic removal [128]. Researchers have discovered that applying a greater voltage density and raising the electrolyte concentration are useful strategies to further improve the removal effectiveness of microplastics [133].

3.1.2. Advanced Treatment Methods.

3.1.2.1 Membrane technologies.

Microplastics can be successfully removed by advanced membrane filtration methods including ultrafiltration and nanofiltration. With their particular pore diameters, these membranes can let water pass through while selectively excluding microplastics [134]. Membrane technology stands out as a potent solution for eradicating microplastics (MPs) from wastewater. Through the utilization of various electrode configurations, pH adjustments, and varying reaction durations, the electrocoagulation-electroflotation (EC/EF) method has demonstrated an exceptional efficacy in completely removing two different types of polymers, achieving a notable 100% elimination rate. An important example is this procedure. A 100% removal efficiency for MPs was also demonstrated using membrane filtering, which again showed remarkable efficacy [124]. Additionally, the membrane-based fertilizer-driven forward osmosis (FDFO) technique shown remarkable success in eliminating all microplastics (MPs) and nanoplastics (NPs) from wastewater, yielding irrigation water of superior quality [135]. Furthermore, membrane fouling caused by extracellular materials was successfully reduced by the FDFO procedure [136]. Additionally, it was remarkable how well MPs and NPs were removed using air flotation as well as nano-ferrofluid techniques. These methods effectively removed over 90% of fragment particles when combined with membrane techniques such as microfiltration and ultrafiltration [137].

3.1.2.2. Biological method.

Biological methods utilize the ability of living things to break down environmental microplastics (MPs) and tackle the problem of MP contamination. Researchers have studied in great detail how different species may break down microplastics (MPs) found in natural water sources as well as wastewater. The biological entities that are studied the most in terms of MPs degradation potential are microorganisms. According to Harrison et al. [125], microbial activity can have a substantial impact on the decomposition of microplastics (MPs), offering a potential therapeutic option for aquatic environments. This strategy might be used in vitro as well as in situ, providing a way to lessen the negative effects of MPs on aquatic life. To break down microplastic pollution, many biological therapies are employed, using a broad range of species such sea clams, corals, algae, fungus, bacteria, enzymes, and marine microorganisms like eukaryotes and archaeans. Further helping to remove microplastics are biopolymers including chitin, cellulose, starch, and lignin, which form bigger flocs that may be removed later.

3.1.2.3. The Adsorption.

Materials with a strong affinity for microplastics, including activated carbon or specialty resins, are used in adsorption processes. These substances possess the capacity to draw in microplastics and cling to their surfaces, therefore eliminating them from aquatic environments. [138]. Chemical bonds, such as covalent, ionic, and hydrogen bonds, are formed during chemical adsorption. Physical adsorption, on the other hand, is dependent on intermolecular forces like Van der Waals forces [139]. Usually occurring at lower temperatures, physical adsorption is characterized by its rapid adsorption rates, low heat of adsorption, and indiscriminate adsorption inclinations. Chemical adsorption, on the other hand, involves the formation and breaking of chemical bonds and requires high temperatures in order to function selectively [124]. Novel composite materials that efficiently

absorb microplastics (MPs) have been created by researchers. These composites are classified as either sponge composites [140][141] or powder composites [142][143]. The method used by each group to absorb MPs varies. Powder composite materials are usually separated using a variety of physical techniques, such as magnetic extraction or high-speed centrifugation, after initially being mixed with MPs. One effective environmentally friendly adsorbent, for instance, was created by adding iron ions to fly ash (FA) in order to extract polystyrene microplastics [140]. High-speed centrifugation was used to separate the adsorbent from the MP suspension after it had been added and mixed. The iron-modified FA and MPs exhibited strong interactions, as demonstrated by thorough characterizations. The three primary mechanisms for adsorption were complexation, π–π interactions, and electrostatic attraction.[124].

3.1.2. The Advanced Oxidation Processes (AOPs).

Polymeric chain breakage is a common application of photocatalytic oxidation. A photocatalyst—typically a semiconductor—was stimulated with photonic radiation in this process. One can receive photonic energy from either artificial or natural light sources. The electrons and positive holes are separated by the excitation. Hydroxyl radicals are created when positive holes contact with water, and super oxide is created when electrons combine with oxygen. The polymeric chains are broken down by superoxide and hydroxyl radicals. ZnO nanoparticles were employed to break down low density polyethylene microplastics. The findings demonstrated that MPs' elasticity characteristics vary, and these variations are directly related to modifications in chemical bonding [144]. MPs were photocatalytically degraded using TiO2-based micro and nanodevices. The removal efficiency was demonstrated to be markedly affected by the surface area and the interplay between microplastics (MPs) and photocatalysts.. Additionally important factors in the breakdown of MPs are salinity and light [145][146]. Hydrothermal hydrolysis and combined carbocatalytic oxidation on magnetic spring-like carbon nanotubes have been used to assess MPs degradation. Solid pyrolysis utilizing nitrogen dopants and encapsulated magnetic nanoparticles was used to create novel nanocarbon springs. By catalytically activating peroxymonosulfate, carbon hybrids produce reactive radicals [147]. The ability of the N-TiO2 photocatalyst to degrade high density was investigated utilizing two alternative preparation methods. MPs made of polyethylene found in aquatic media. Utilizing cutting-edge analytical methods, wastewater analysis verified the MP deterioration [148]. The wastewater's clarity is essential to the photocatalytic process since it contains photonic energy must be capable of penetrating. When radiation is applied from an artificial source, it suggests a substantial influence on life because energy use. Lamp cleaning and replacement on a regular basis increase labor costs. The existence of additional MPs and organic pollutants both have an impact on the photocatalytic process [149].

3.2. Removal method of antibiotics from aqueous water.

Several treatment techniques are employed for removing antibiotics from various water sources and addressing emerging pollutants. Illustrated in Figure 6, these methods can be categorized into six primary blocks, each containing specific elimination strategies. Among these, the Coagulation-Flocculation (C-F) technique holds prominence in chemical reactions, often serving as the initial step in the process. They can be used at several

water treatment stages, including: industrial wastewater must first be pretreated before it may enter municipal sewers[150]. Urban wastewater treatment and drinking water treatment facilities are the two primary techniques for handling urban waste water [151].The potential of leveraging adsorption, particularly through activated carbon, for the removal of pharmaceutical residues from wastewater. Results from research indicate that it can remove ciprofloxacin, cellulose membranes, and antibiotics with removal percentages ranging from 90% to 97%. UV/H2O2, FP, EO, OP, and Electrochemical Oxidation (EO) are methods for eliminating contaminants using physically Advanced Oxidation Processes (AOPs). When employed with various media, including zeolite, powdered activated carbon (PAC), granular activated carbon (GAC), and cellulose membranes, these techniques have demonstrated efficacious outcomes in the elimination of pollutants. These techniques show tremendous promise for application in the pharmaceutical industry and other drug-related wastewater remediation [152].

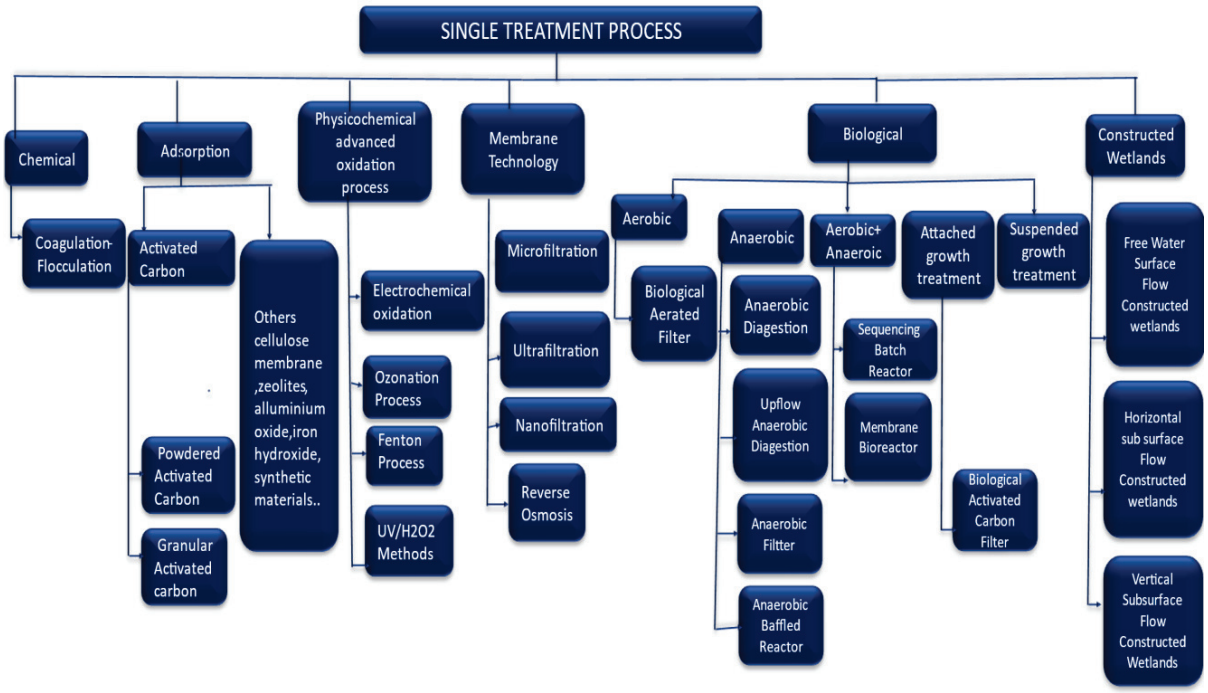

Fig 6 Showing Removal method for antibiotics.

AOPs based on ultraviolet radiation (UV / H2O2) are a good substitute, but they come at a high expense because of the necessity of upstream pretreatment and downstream H2O2 cooling, which is quite irksome [153]. However, because it requires less energy and chemicals and has an efficiency similar to the prior one, ozone is offered as a really intriguing substitute [154]. At the same time, it is also less expensive. Reading over these studies, which were all carried out in wastewater treatment facilities as well as frequently in pilot plants and municipal treatment facilities, are the antibiotic elimination studies conducted with this approach [153] [155]. The technology of membranes comes in fourth. Pollutants are caught in this process as wastewater travels through tiny membrane holes. Microfiltration (MF), Nanofiltration (NF), Ultrafiltration (UF), and Reverse Osmosis (RO) are the classifications that allow them to be separated. Notably, membrane filtering methods have been tested in lab environments as well as in real-world applications for the removal of medicinal

compounds—like antibiotics—from various liquids [152]. Three categories of biological treatments (anaerobic, aerobic, and hybrid approaches) are categorized in fifth place. The primary anaerobic technique utilized is the Biological Aeration Filter (BAF) system. This distinctive approach to wastewater treatment employs biological processes to integrate both oxidation and filtration [156]. The process involves three distinct phases: the gas phase allows for air entry, the liquid phase is where the solid material is submerged, and the solid phase supports microbial growth. Examples of anaerobic technologies include the upflow anaerobic sludge blanket (UASB), anaerobic baffled reactor (ABR), anaerobic digestion (AD), and anaerobic filter (AF). However, these methods might have adverse environmental effects. The technologies of Sequencing Batch Reactor (SBR) and Membrane Bioreactor (MBR) integrate both anaerobic and aerobic processes. With MBR, you can get better nitrification performance, low sludge formation, flexible operation, and extended sludge retention times., while SBR runs in five sequences. Membrane separation technology and biological technology are combined in MBR. Utilizing biological methods, such the Biological Activated Carbon filter (BAC) to purify drinking water, are used in attached and suspended growth treatments. Strong, easy to construct, and low energy use characterize these systems [152]. Man-made wetlands rank sixth. The wetland is fed by wastewater that is treated artificially through an ecosystem composed of plants, bacteria, and soil. Wastewater is purified by a number of techniques, including filtration, coprecipitation, ion exchange, plants adsorption, and microbial breakdown. [157]. Three types of treatments can be distinguished by the direction of the water flow: Constructed Wetlands with Free Water Surface Flow (FWS CWs), Horizontal Subsurface Flow (HSF CWs), or Vertical Subsurface Flow (VSF CWs) [158].

3.3. Management approaches and future perspective.

The fundamental ideas behind control techniques for antibiotic and microplastic pollution are water separation and entrance prevention, control, and management. By cutting back on production and consumption, organizing the disposal process, and recycling, pollutants can be controlled throughout their entire life cycle. Future study tought to, in our opinion, concentrate on removal techniques, instrument improvement, and strategies for reducing pollution emissions. The current regulatory framework addresses each aspect of the plastic life cycle, with a particular emphasis on the stages of product processing, consumption, and disposal. It excludes plastic waste reuse, recycling, and the synthesis of plastics from raw materials, and it excludes the integration of plastics from aquatic habitats into the existing recycling value chain. To avoid and manage plastics from entering the aquatic environment, we should consider developing an efficient benefit guiding system and controlling plastics throughout their whole life cycle. Treatment options that show promise include raw material replacement, reuse of products, waste classification, disposal of rural trash, recycling of waste plastic, and resource usage. In addition to reducing or eliminating antibiotic use at the source, other crucial objectives include improving wastewater treatment technologies for use in homes, hospitals, and industries [159]. The technology's high equipment costs and energy usage are a drawback, though. A number of antibiotics (pollutants) can be eliminated from wastewater and the water purified by using adsorbents with well-developed pore structures, like activated carbon and biochar. Despite the fact that MP-antibiotic separation (removal) from aquatic habitats has reached an objective degree of technology, study on MP-antibiotic combinations has not

been published. Currently, there is no published research specifically addressing the technology appropriate for microplastic (MP) and antibiotic combinations. Investigating how these combinations can be isolated in water will thus be an important research focus in the future. The fate of microplastics consumed by aquatic organisms and their ecotoxicological impacts on wildlife are still largely uncertain, particularly regarding potential health risks to humans. Given the increasing concerns about the toxicity of microplastics absorbed through antibiotics, further research is underway to understand their effects on humans and higher trophic level animals

CHAPTER 4

CONCLUSION

These pollutants' combined impacts increase their toxicity and duration while making it more difficult to remove them from water bodies. Antibiotic resistance genes in the microbial population may develop more quickly as a result of the transfer of antibiotics made possible by microplastics. The environmental and health effects of this relationship may be increased, which makes it an important field of research. There is a pressing need for efficient techniques of removing these pollutants. Physical filtering, chemical treatments, and biological methods are some of the current methods; each has benefits and drawbacks of its own. More advanced methods, such the creation of materials with multiple uses and nanotechnology, have the potential to increase removal efficiency. Nonetheless, the complex nature of the interactions between antibiotics and microplastics demands the creation of creative, integrated treatments that can successfully handle the dual contamination. Antibiotics and microplastics interact, and future studies should focus on understanding the mechanisms underlying these interactions as well as their long-term impact on human health and aquatic ecosystems. Furthermore, more comprehensive and standardized approaches are required to assess how well different treatment systems remove these pollutants. With the expansion of knowledge and improvement. It is clear from a review of the literature that a variety of physicochemical processes enable antibiotics to adsorb easily onto microplastic surfaces. The fact that these interactions depend on variables including antibiotic qualities, microplastic features, and environmental circumstances highlights how complicated this process is. Furthermore, antibiotics may become more persistent and bioavailable in aquatic systems as a result of their adsorption onto microplastics, which could be dangerous for both human and environmental health. But fully comprehending and reducing the effects of antibiotic adsorption on microplastics is a problem that the field must overcome. Several obstacles need to be addressed, such as the absence of established research methods for examining adsorption processes, insufficient information on the movement and destiny of microplastics containing antibiotics in various environmental matrices, and uncertainties about the ecological consequences of these interactions on aquatic life. Interdisciplinary teams working together, creative research strategies, and coordinated efforts to close current knowledge gaps are needed to address these difficulties. The amount of antibiotic-loaded microplastics that build up in aquatic habitats has been reduced by the development of numerous removal procedures in response to the pressing need for workable mitigation strategies. The physical, chemical, and biological techniques that are included in these methodologies each have their own advantages and drawbacks. Certain methods concentrate on eliminating microplastics exclusively, while others target related pollutants, such as antibiotics, to tackle wider environmental issues. Therefore, research projects that attempt to clarify the fate, transportation, and ecological consequences of antibiotics' adsorption on microplastics must be given top priority. This makes it necessary to keep working to create reliable analytical

methods, set uniform procedures, and improve interdisciplinary teamwork. In addition, it is imperative to take proactive steps to address the root cause of microplastic pollution, with a focus on promoting circular economy concepts and sustainable waste management methods. In order to effectively address this complicated issue, comprehensive approaches are required, as this review highlights the complex interactions between antibiotics and microplastics in watery settings. By enhancing our comprehension of these connections and applying focused mitigation strategies, our goal should be to shield the general public's health and aquatic ecosystems from the negative effects of microplastic pollution.

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