

Review



The Occurrence, Distribution, Environmental Effects, and Interactions of Microplastics and Antibiotics in the Aquatic Environment of China

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Abstract: Microplastics (MPs) and antibiotics (ATs) have been detected in various aquatic environments and characterized as novel contaminants that have attracted worldwide attention. This review summarizes the characteristics of MPs and ATs, analyzes the sources of MPs and ATs in aquatic environments, reviews the concentration distribution of the two pollutants in China, and introduces the environmental effects of mixing MPs and ATs. Studies on single pollutants of MPs or ATs are well established, but the interactions between the two in aquatic environments are rarely mentioned. The physicochemical characteristics of MPs make them carriers of ATs, which greatly increase their risk of being potential hazards to the environment. Therefore, in this article, the interaction mechanisms between MPs and ATs are systematically sorted out, mainly including hydrophobic, electrostatic, intermolecular interactions, microporous filling, charge-assisted hydrogen bonding, cation-bonding, halogen bonding, and CH/ π interactions. Also, factors affecting the interaction between ATs and MPs, such as the physicochemical properties of MPs and ATs and environmental factors, are also considered. Finally, this review identifies some new research topics and challenges for MPs and ATs, in order to gain deeper insight into their behavioral fate and toxic mechanisms.

Keywords: microplastics; antibiotics; aquatic environment; sorption; interaction mechanism; combined pollution

1. Introduction

One of the great inventions of the 20th century, plastics are widely used in the foodstuff, textile, automotive, construction, and pharmaceutical industries, leading to a dramatic increase in plastic production [1]. In 1950, the global plastic production was 500,000 tons per year. However, it is projected that it could reach 1.8 billion tons per year in 2050; the plastic recycling rate is only about 9%, while 12% is incinerated, and the majority of plastic waste is discharged into the natural environment [2,3]. Plastics released into the environment are difficult to break down naturally and will degrade into microplastics (MPs) over time. Plastic particles less than 5 mm in diameter are usually defined as MPs and have been found in the ocean [3], rivers [4], atmosphere [5], soil [6], groundwater, and even drinking water [7]. Not only that, microplastics have been detected in marine organisms [8], freshwater organisms [9], soil organisms [10], and even human blood [11], seriously affecting biological health. MPs have properties such as hydrophobicity, surface charge, long molecular chain arrangement, large specific surface area, size, shape, color, and diversity of functional groups. The physicochemical properties of MPs confer strong



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Copyright: © 2024 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). adsorption and migration abilities and are considered ideal carriers for many pollutants in the environment [12,13].

Since their discovery in the 1830s, antibiotics (ATs) have significantly improved human and animal health and agricultural yields. Worldwide, ATs are used in more than 100,000 tons per year and have gained increasing interest in recent years as an emerging pollutant [14]. ATs are antimicrobial drugs that interfere with the developmental functions of bacteria and are widely used in the livestock, medical, and aquaculture industries [15]. Most ATs are not completely absorbed and degraded by organisms upon entry, and approximately 5–90% of ATs are released to soil and water as prototypes or metabolites [16]. ATs released into the environment are accidentally ingested by organisms, either directly or indirectly. Accidentally ingested ATs can accumulate in plants, animals, and humans through the food chain, causing ecosystem and health problems [17,18]. ATs have been reported in wastewater [19], hospital wastewater [20], farming wastewater [21], surface water [22], groundwater [23], oceans [24], drinking water [25], soil [26], and sediment [27]. AT residue in the environment not only endanger human health and disrupt the balance of ecosystems but may also accelerate the spread of antibiotic resistance genes (ARGs) and antibioticresistant bacteria, thereby threatening public health and global health security [28].

Due to their physicochemical properties, MPs can be used as carriers to create complexes to disperse other toxic contaminants such as heavy metals [29] and organics [30]. Among the toxic contaminants adsorbed by MPs, ATs are important organic pollutants [28]. MPs and ATs are both pollutants with high detection rates in aquatic environment. When they meet, MPs will adsorb ATs, leading to their dispersion into different environmental media, which in turn causes wider and more complex contamination [3,27]. The coexistence of MPs and ATs forms a prominent form of composite pollution, but there is a lack of dedicated and comprehensive reports on the behavioral characteristics and interactions of the two pollutants. This paper reviews published journal papers on the interactions of MPs and ATs to better understand the environmental effects of the two pollutants. A total of 139 journal articles (2018.01-2023.12) were searched in the Web of Science library using the keywords "antibiotics" and "microplastics". The keyword metrics analysis of the 139 retrieved papers was performed using CiteSpace software (v.6.2.6), and the clustering results are shown in Figure 1. The analysis of seven clusters in the cluster view and the summary of the literature revealed that the 139 existing studies mainly focused on single pollutants of MPs or ATs, and their interaction mechanisms and environmental effects have not received the same attention. Therefore, after discussing the environmental effects of MPs and ATs, this paper will provide an in-depth study of the interactions between MPs and ATs, with a view to accurately evaluate the potential risks of MPs adsorbed to ATs.

As the world's largest developing country, how China can effectively improve its ecological environment in the process of rapid economic development has become an essential topic of human concern and research. Since 1978, China has implemented the economic policy of reform and the opening of its economy, and the rapid economic growth has created a number of environmental problems. Rapid population growth, urbanization, and the expansion of scale in the agricultural industry has been accompanied by an explosion in the production and use of chemicals, including MPs and ATs. With its large economy and population, China has a large share of the global production and use of MPs and ATs. At present, China's marine, river, and lake ecosystems are facing increasing pollution pressure from MPs and ATs. Based on this situation, this study reviewed the relevant studies on MPs and ATs in China, summarized the distribution characteristics of MPs and ATs in China, analyzed the environmental effects of MPs and ATs, and explored the interaction mechanisms and influencing factors of MPs and ATs in the aqueous environment.



Figure 1. Clustered view of the Web of Science journal literature with "antibiotics" and "microplastics" as keywords.

2. Structure, Properties, and Distribution of MPs and ATs

2.1. Structure, Characteristics, and Distribution of MPs

MPs include primary MPs and secondary MPs. Primary MPs are plastic microparticles that are inherently small in size, such as those used in skin care products, cosmetics, detergents, and medical supplies [31]. Plastics released into the environment are known as secondary MPs, which are small plastic fragments produced by the fracture and decomposition of plastics through physical (weathering, mechanical, and water interference), chemical (photo-oxidation, UV radiation, and freeze-thaw cycles), biological (microbial degradation), and other actions [32]. During the production process of plastics, numerous additives such as plasticizers, surfactants, flame retardants, antioxidants, lubricants, adhesives, etc., are added to improve their plasticity and practicality [33]. Plastic waste will gradually fragment and degrade when it enters the aquatic environment. As the MPs degrade, these additives leach out of the MPs and affect the quality of the aquatic environment [2,34]. In addition, MPs are hydrophobic substances and can adsorb various hydrophobic organic pollutants floating on the ocean [35], resulting in more severe ecotoxicity. There are many types of MPs; polyethylene terephthalate (PET), polyethylene (PE), polystyrene (PS), polypropylene (PP), polyamide (PA), and polyvinyl chloride (PVC) are the most commonly detected MPs in aquatic environments [36]. The structures and physicochemical properties of different types of MPs are listed in Table S1.

The main sources of MPs in the environment are waste disposal, surface runoff, industrial production, and wastewater treatment plants (WWTPs). WWTPs existing treatment technologies are less effective in removing nanoscale plastic particles that end up in the environment and cause multiple environmental impacts [37,38]. To date, specific treatment technologies have not been developed specifically for the removal of MPs from wastewater and sludge, in addition to the conventional treatment technologies available [39]. In general, wastewater treatment plants do not achieve good removal performance for wastewater collected with many MPs particles. On the other hand, the influent base of WWTPs is so large that even if the MPs removal rate achieves a good result, huge amounts of MPs will still be discharged into the aquatic environment [40].

MPs in the environment are easily transported, dispersed, and redistributed globally due to their small particle size, chemical stability, and hydrophobicity. For example, MPs have been found in the Mariana Trench [41], Antarctica [42], the Arctic Ocean [43], the deep

seabed [44], and pristine mountainous areas [45]. This is likely due to the long residence time of MPs, which allows sufficient residence time for currents, sedimentation, and other effects to move them into these relatively extreme environments. Certainly, the concentration of MPs and the degree of contamination vary from one geographic region to another. In general, the abundance of MPs is higher in waters near urban areas than in waters near rural areas due to population density and the level of industrial facilities. Population density, the number and type of industrial facilities, and the methods used to dispose of plastic waste are the key variables influencing the concentration of MPs in aquatic environment. The abundance of MPs in various environmental media in China is shown in Table S2 [46–76], and the visualization results are shown in Figure 2. There are three characteristics of MPs pollution in China (Table S2): (1) Spatial distribution: MPs were widely distributed spatially, and MPs were detected from beaches, surface waters to sediments. (2) Geographical distribution: The concentration of MPs is positively correlated with the level of economic development, and Beijing-Tianjin-Hebei, the Yangtze River Delta, and the Pearl River Delta are the main areas enriched with MPs. (3) Distribution characteristics of MPs: PE, PP, PS, and PET are the types with the highest detection rates in aquatic environments in China, with colors mostly colored and colorless, and morphology mostly fibers and fragments. Specifically, the most polluted river in China is the Lower Yellow River and its estuary with 65,400–93,200 n/m³ of pollutants, the most polluted ocean is the Northern Yellow Sea with 545 ± 282 n/m³, and the highest concentration of pollutants in lakes is Poyang Lake with 5000-34,000 n/m³. It is worth noting that the collection, detection, and quantification methods of MPs have not yet been unified, the above statistical results have certain limitations, and the establishment of a complete analytical method for microplastic characterization is also an urgent problem to be solved at this stage.



Figure 2. Abundance of microplastics in aquatic environments and sediments of China. (Note: Only regions with available data for the period 2010–2023 are indicated in the figure).

2.2. Structure, Characteristics, and Distribution of ATs

ATs can be classified into fluoroquinolones, tetracyclines, macrolides, and sulfonamides according to their chemical structure [77], and their structural and physicochemical parameters are shown in Table S3. Different types of ATs have differences in their physicochemical properties (octanol–water partition coefficient (Log Kow), acidity coefficient (pKa), sorption behavior, and photo reactivity), resulting in different biological properties (activity and toxicity) and forms of presence (neutral, cationic, anionic, or amphoteric) of ATs in aquatic environments [12], as supported in Table S3.

The widespread global use of ATs has led to a dramatic increase in the concentration of ATs in nature [78]. The production and use of ATs in China occupies an important position, and the management of ATs in China is a major challenge. According to the model established by Zhang et al. [79], in 2013, the total production of ATs in China was 248,000 tons, the import was 600 tons, the export was 88,000 tons, and the total use was about 162,000 tons. ATs enter surface waters mainly through leachate from landfills, fertilization, and agricultural runoff from aquaculture [80]. In addition, pharmaceutical manufacturing plants, medical effluents, and animal husbandry effluents with high concentrations of ATs are discharged into municipal sewer systems with only little treatment and eventually collected by WWTPs [81]. However, the current removal of ATs by WWTPs is extremely limited, and the concentration of ATs in treated water that is ultimately discharged in receiving waters is still at a high level [82–84]. Due to the widespread use and point source discharge of ATs, a variety of ATs can be monitored in aquatic environments and sediments in several regions of China, with fluoroquinolones, tetracyclines, and sulfonamides being the types with the highest detection rates (Table S4 [85–123] and Table S5 [72,97,100,104,124–133]). Residual ATs in the environment not only adversely affect non-target organisms but also enhance bacterial resistance, which ultimately has unimaginable consequences for aquaculture, agriculture, animal husbandry, humans, and the ecosystem [134].

The distribution and degree of pollution of ATs in different regions varies from region to region, and the pollution of ATs in China is mainly concentrated in the Yangtze River Basin, the Bohai Bay, the Pearl River Delta, and the Beijing–Tianjin–Hebei region (Figure 3). From the data collected in Table S4, the highest pollution concentrations of tetracyclines, fluoroquinolones, and macrolides were 4720.0 μ g/L, 9281.7 μ g/L, and 1112.2 μ g/L, respectively, and the above-mentioned polluted places appeared in Taihu Lake in the Yangtze River Delta region, Qinghe River in Beijing–Tianjin–Hebei region, and Zhujiang River in Pearl River Delta, respectively, which represented the highest pollution levels of several types of ATs in China. Characterization of the distribution of ATs in sediments is generally consistent with the aquatic environment (Figure 3 and Table S5). The local industrial structure, the disposal of ATs by the pharmaceutical industry, the use of ATs by the livestock industry, the climate, and the distribution of water bodies in the region are all key factors contributing to the appearance of the above data characteristics. For example, in the economically developed eastern region of China, the pharmaceutical industry is concentrated, and the discharge of large amounts of AT-rich pharmaceutical wastewater leads to a high concentration of ATs pollution in the eastern region [135]. By comparing the distribution of MPs and ATs in China, it is found that the two have similar overlapping areas and pathways, providing a realistic basis for studying their interaction.

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Figure 3. Abundance of antibiotics in aquatic environments and sediments of China. (Note: Only regions with available data for the period 2010–2023 are indicated in the figure).

3. Mixed Ecological Effects of MPs and ATs

The corrosion resistance of MPs makes it difficult for them to degrade under natural conditions, giving them enough time to interact with ATs and generate new environmental effects, and the environmental impacts and behavioral characteristics of the two are shown in Figure 4. As carriers of contaminants, MPs will interact with ATs in terms of adsorption and migration to form new combined contaminants, producing toxicity different from that of individual contaminants [136,137]. For example, an intestinal histopathology study on zebrafish found that combined exposure to oxytetracycline (OTC) and MPs attenuated intestinal damage induced by OTC alone [138]. When MPs enter the interior of an organism, they disrupt the structural integrity of cells, and the addition of tetracycline reduces the percentage of cell membrane damage by 18.4% and reduces cell-particle contact and individual toxicity [139]. A recent study suggests that coexistence of MPs with TCs reduces phenol removal efficiency in wastewater treatment plants, affects sludge characteristics, and accelerates the spread of certain ARGs [140]. The coexistence of MPs and ATs, as described above, has led to a reduction in contaminant toxicity, but exacerbation of toxicity due to mixing of the two is also common. A study on the synergistic immunotoxicity of MPs and ATs found significantly lower phagocytosis and total blood cell counts in mussels using both MPs and ATs compared to mussels using ATs alone [141]. In addition, phosphorus removal is an important process segment in wastewater treatment. In this process, MPs inhibit the phosphorus removal rate of phosphorus-accumulating organisms (PAOs) mainly through physical and oxidative damage, while the toxic mechanism of ciprofloxacin (CIP) is binding to the DNA of PAOs [142]. When MPs are present with CIP, CIP can cause PAOs to produce more extracellular polymerized material, thereby reducing the oxidative stress

of MPs on PAOs [14]. Additionally, the adsorption of MPs decreases the concentration of CIP, thereby inhibiting its binding to DNA and thus its effect to PAOs [143]. The synergistic effect of MPs and TC affects the growth of bacteria and consequently the nitrification process in the wastewater treatment process [144]. Current toxicity studies of ATs and MPs have mostly focused on the toxicity of single substances, while the environmental effects of combined exposure to MPs and ATs are still unclear. In general, the combined effects of MPs and ATs have a multidimensional impact on ecosystems, so more in-depth studies should be devoted to assessing the individual effects and the combined behavior of the pollutants.



Figure 4. Environmental effects and behavioral characteristics of microplastics and antibiotics in the environment.

A significant correlation was found between the concentration of ARGs and ATs in aqueous environments [145]. The improper treatment of ATs leads to the proliferation of ARGs, making bacteria more resistant and able to evade the controlling effects of antimicrobial drugs [146–148]. MPs and ARGs coexist widely in aquatic environments, and MPs can act as storage units to influence the type and abundance of ARGs [149–151]. For example, in aquaculture, MPs can act as carriers of ARGs and enter aquatic organisms through the food chain, posing a significant risk to aquaculture and human health [152]. In wastewater treatment, MPs can block UV beams and affect the UV disinfection efficiency of ARGs [153]. In riverine environments, MPs can carry and disperse ARGs in different locations, and urban rivers pose a higher potential environmental risk compared to rural and peri-urban areas [154]. In waste leachate, MPs can selectively enrich for specific pathogens, and the carriage of 11 pathogens was positively correlated with ARGs, suggesting that the presence of MPs greatly increases the enrichment of ARGs and pathogens [155]. In addition, MPs provide new substrates for biofilm formation and may act as new carriers of ARGs into the aquatic environment, creating ecological risks and adversely affecting human health [156]. The interaction between MPs and ARGs has been shown to affect community structure and produce toxic effects in aquatic ecosystems. Overall, MPs in aquatic environments enriched ARGs; caused contamination, proliferation, and accumulation of ARGs; and stimulated a vicious cycle of ARGs, leading to greater pathogenic potential and more severe ecological impacts.

4. Factors Influencing the Interaction between MPs and ATs

In aquatic environments, MPs and ATs interact with each other in various ways. The interaction process is influenced by the physicochemical properties of MPs, the physicochemical properties of ATs, and environmental factors (Figure 5).



Figure 5. Factors influencing the mechanism of interaction between microplastics and antibiotics.

4.1. Physicochemical Properties of MPs

4.1.1. Types

The diversity of MP types determines the diversity of their properties. The type of MP plays an important role in determining the polarity, pHpzc (point of zero charge), crystallinity, and functional group species; the properties of different types of MPs are shown in Table S1. The functional group type determines the type of interactions between MPs and ATs, such as hydrogen bonds, π - π interactions, and halogen bonds. The polarity of MPs is not a direct factor affecting their adsorption capacity, but the polar groups of MPs and polar ATs may generate electrostatic reaction energy between them through dipole-dipole and dipole-induced dipole attraction, leading to a stronger adsorption effect [157]. The pHpzc can be used to reflect the nature of charge properties exhibited by the MPs particles, which in turn affects the electrostatic interaction of MPs with ATs. The different types of MPs lead to differences in the physicochemical characteristics of the polymers, which in turn lead to differences in their adsorption properties.

4.1.2. Crystallinity

The crystallinity of MPs may also affect the interaction between MPs and ATs. According to the available literature, there are controversial points on the effect of the crystallinity of MPs on the adsorption of ATs. One theory suggests that the crystallinity may affect the ability of MPs to adsorb organic micropollutants because the crystalline fraction has lower oxygen permeability and is less susceptible to oxidation [158]. In the amorphous part, the polymer molecules are disordered and move randomly, and there is space and conditions for the adsorption of pollutants, and the ratio of this region determines the adsorption rate of MPs on ATs and their concentration at equilibrium [159,160]. For example, in a study of norfloxacin (NOR) adsorption by PS, PP, PE, and PA, it was found that PS, which had a lower crystallinity, had more amorphous regions and higher surface activity, and therefore had the strongest adsorption capacity [161]. After subsequent aging treatments of the four MPs, the crystallinity increased, and the adsorption capacities were all weakened [161]. However, it has also been suggested that there is no significant relationship between the crystallinity of MPs and their adsorption properties. The research shows that MPs with high crystallinity have obvious diffraction peaks. Based on this feature, the pure crystallinity of MPs order is PP > PA > PS [162]. However, the order of the actual adsorption capacity for pollutants was PA > PP > PS, which was not significantly related to the size of crystallinity, indicating that crystallinity of MPs affects the interaction of MPs with ATs under certain conditions, but further experimental verification is needed.

4.1.3. Size

Variation in size affects the adsorption of MPs in different ways, mainly by changing the number of adsorption sites and the size of the specific surface area required to influence their adsorption of ATs [32]. Degradation of plastics under natural conditions is usually accompanied by changes in size, and these changes give them different adsorption properties. In a comparative study between MPs and nanoplastics, it was found that the rate constant for CIP adsorption decreased significantly when the size of PS changed from micron to nanoscale, which may be due to strong surface competitive adsorption. However, when the diameter of PVC was decreased from 74 μ m to 1 μ m, the adsorption of CIP was significantly accelerated [164]. Another result on the effect of particle size on the adsorption of Triclosan on PS showed that the amount of Triclosan adsorbed gradually increased as the PS particle size decreased [165]. Overall, the adsorption behavior is completely different as the size of the plastic decreases, which is highly dependent on the type of MP and contaminant.

4.1.4. Aging Behaviors

MPs show aging behavior under environmental conditions such as oxidation, thermal radiation, UV radiation, wind, etc., thus changing their microstructure and adsorption behavior [166,167]. Aged MPs are characterized by a rough surface and large surface area, while oxygen-containing functional groups such as hydroxyl and carboxyl groups are formed under the action of aging factors such as light and oxygen [168]. Aging causes the appearance of hydrophilic groups, microcracks, pores, and pits on the surface of MPs, which enhances the adsorption of pollutants [169]. For example, weathered MPs undergo surface interfacial behavior with hydrophilic ATs, and the weathered MPs adsorb much more CIP than the original MPs [170]. By studying the adsorption behavior of naturally aged MPs in freshwater and simulated seawater, it was found that aged MPs can be carriers of ATs and cause long-term effects on organisms and the aquatic environment [171]. A recent study demonstrated that the adsorption capacity of PE to chlortetracycline and amoxicillin (AMX) increased 1.08–14.24-fold after aging [172]. In a study simulating the aging of MPs, it was demonstrated that the aging process induces oxygen-containing functional groups such as -OH, C=O, and C-OH [168,173]. Firstly, the physicochemical properties of MPs such as hydrophilicity, polarity, and surface charge are influenced by the aforementioned functional groups, which in turn enhance the hydrophobic, intermolecular, and electrostatic interactions between MPs and ATs [168,173]. However, the presence of functional groups also forms hydrogen bonds with water molecules in the surrounding environment, which reduces the adsorption sites of pollutants and hinders the adsorption process [173].

The aging behavior of MPs mentioned above mainly changes the physicochemical properties of MPs themselves, which in turn affects the interaction between MPs and ATs. And during the aging process, biofilms are formed on the surface of MPs, and the formation of biofilms will indirectly change the roughness, surface charge, and surface free energy of MPs, thus affecting the interaction between MPs and ATs [174]. Although it has been found that the bacterial organisms present in the biofilm formed on the surface of MPs have a certain degradation effect on ATs [175], the degradation effect is poor and the degradation period is long compared to its adsorption, such that the bacterial degradation in the biofilm is negligible [176]. In summary, MPs undergoing the aging process—resulting in increased surface areas, increased adsorption sites, the production of oxygen (increased polarity), oxygen-containing functional groups, biofilms, and fouling—will also increase the charge, roughness, and porosity, and will accumulate greater concentrations of ATs or other contaminants [177]. MPs that undergo aging behavior enhance the adsorption of ATs, indicating that the research direction should lean toward the adsorption behavior of aging MPs and presume that aging is the most common phenomenon of MPs in natural aquatic environments.

4.2. Physicochemical Properties of ATs

The type of ATs determines physicochemical parameters such as hydrophobicity, functional group species, pKa, Log Kow, etc., which can affect the adsorption of MPs. It was shown that the Log Kow of ATs is crucial to determine their adsorption range on MPs [173,178]. The intensity of ATs adsorption by MPs was found to be significantly and positively correlated with the Log Kow of ATs in a study of the interaction of MPs and ATs [162]. ATs with higher hydrophobicity (having higher Log Kow values) have a higher affinity for MPs. As ionizable compounds, the pKa of ATs are usually veritably different due to the different functional groups possessed by different species of ATs. Therefore, under specific pH conditions, various ATs exhibit different cationic, amphoteric, and anionic forms, which are important factors affecting the interaction between ATs and MPs. Table S3 in the supporting material summarizes the structural formula, Log Kow, pKa, and other parameters of various types of ATs in order to facilitate the interpretation of understanding the interaction mechanism between MPs and ATs.

4.3. Environment Factors

4.3.1. pH

The pH primarily influences the electrostatic interaction of MPs and ATs to modify adsorption behavior, which is determined by the degree of ionization of the substance in the solution. Also, the pH of a solution and the acid dissociation constant (pKa) of a chemical substance are important parameters for the degree of ionization. For example, TC has multiple ionizable functional groups, including amino and hydroxyl groups, in multiple forms throughout the pH range [179]. In aqueous solutions, TC exhibits three pKa values of 3.3, 7.7, and 9.7. When pH < 3.3, the solution is acidic, and the dimethylamine group attracts protons under this condition, and TCH3+ is the main component of the TC group, resulting in the TC molecules in the solution exhibiting a cationic nature [180]; When 3.3 < pH < 7.7, the dominant group in TC is TCH20, which exhibits no external electrical properties; When pH > 7.7, TC is dominated by negatively charged TCH2- and TC2-, resulting in the anionic nature of TC molecules in solution [181,182]. Changes in pH also affect the adsorption capacity of MPs by influencing their zeta potential, i.e., surface potential [183]. MPs polymers are always negatively charged in alkaline solutions and tend to protonate on the surface of MPs as the pH decreases [184,185]. The natural aquatic environment has a pH range of 5–10, where most MPs are negatively charged, and thus the pKa of ATs determines the adsorption or repulsion of MPs with ATs [3]. The pKa values of common ATs are provided in Table S3 to facilitate the understanding of the states of ATs present at different pH conditions (including anionic, cationic, molecular, and amphiphilic ions) for the preliminary prediction of electrostatic interactions between ATs and MPs.

4.3.2. Ionic Strength

The ionic strength is a key factor affecting the interaction between MPs and ATs. The mechanism of ionic strength action is to control the electrostatic and non-electrostatic interactions between the surfaces of MPs and ATs by affecting the thickness and interfacial potential of the bilayer, which in turn affects the binding of MPs and ATs. Ionic strength influences electrostatic interactions to some extent because electrolytes can compete with ATs for electrostatic sites, resulting in weaker MPs adsorption to ATs [186]. When the ionic strength in the environment is high, positively charged ions will replace the ATs adsorbed on the negatively charged MPs. In the adsorption study of MPs, the adsorption of TC by PE was found to decrease with increasing ionic strength, indicating that the enhancement of ionic strength is not favorable to the adsorption behavior of TC on PE [187]. The reason may be that the enhanced ionic strength compresses the thickness of the bilayer and weakens the electrostatic force between MPs and ATs, leading to a decrease in their adsorption capacity [187,188]. Previous studies have shown that hydrophobic interactions between ATs molecules can overcome repulsive electrostatic interactions when the ionic strength is high, leading to the aggregation of ATs molecules [189]. The ATs molecules become larger after polymerization and have difficulty in accessing the adsorption sites inside the MPs, leading to a weaker adsorption capacity than at low ionic strength [189]. In summary, the increase in ionic strength mainly played an inhibitory role in the adsorption properties of MPs.

4.3.3. Salinity

Based on the available research data, salinity has both a positive effect and a negative effect on inhibiting the interaction between MPs and ATs. It is hypothesized that the presence of salt compresses the bilayer on the surface of MPs, leading to the formation of nano-plastic clusters, similar to the coalescence mechanism of Fe³⁺ and Al³⁺ salts. The clusters of nanoplastic particles may exert stronger electrostatic interactions on the CIP compared to individual particles, thus improving the adsorption capacity. However, the concentration of Na⁺ increases gradually with increasing salinity, and the positively charged Na⁺ is more easily adsorbed on MPs by electrostatic attraction due to the negatively charged surface of MPs [190,191]. In this process, the acidic groups of MPs can be replaced by H^+ , which can affect the formation of hydrogen bonds, thus hindering electrostatic interactions and reducing the adsorption capacity [168,192]. This was similarly concluded in a previous study which found that the adsorption capacity of PE decreased with increasing salinity, suggesting that high salinity is detrimental to the adsorption behavior of TC on PE [187]. It was found that the adsorption capacity of Sulfamethoxazole on PA, PE, PET, PVC and PP all showed a decrease in the presence of salt, while the adsorption capacity of PS on Sulfamethoxazole increased slightly with decreasing salinity and then decreased again with increasing salinity to 35% [193]. The NaCl concentration gradient indicated that the amount of SMT adsorbed on MPs decreased with increasing salinity, suggesting that the presence of salt decreases the adsorption of MPs on ATs [157]. Furthermore, increasing salinity reduces the solubility of non-polar and weakly polar ATs in water, resulting in relatively easy pollutant adsorption by MPs [194,195]. Overall, the sorption of ATs by MPs was susceptible to the effect of salinity and mostly exhibited inhibited sorption behavior between MPs and ATs.

4.3.4. Dissolved Organic Matter

It has been found that the aqueous environment contains a variety of heterogeneous organic compounds, such as humic acid (HA), microbial products, bovine serum proteins and polysaccharides, etc [196–198]. The interaction of MPs with dissolved organic matter (DOM) can lead to the formation of an ecological corona on the surface of MPs, which in turn can alter the physical and chemical properties of MPs and affect their interaction with ATs [196,199]. Specifically, functional groups on HA molecules can alter the surface charging properties of MPs, which leads to a decrease in the adsorption affinity of MPs for TC. In addition, HA also competes with the adsorption sites on the surface of MPs, thus decreasing the adsorption of

TC. In addition to interacting with MPs, DOM in aquatic environment also interacts with ATs, which in turn affects the interactions between MPs and ATs. It was found that the presence of DOM all significantly decreased the adsorption of MPs to TC and enhanced the inter-sorption of TC with DOM, probably because the affinity of TC for DOM was higher than that for MPs [200]. On the other hand, in the adsorption of ATs on MPs, DOM plays the role of a "bridge" in the process and enhances the adsorption of ATs on MPs. For example, Zhang et al. [201] found that the π - π conjugation between HA and the surface of MPs leads to the enhancement of electrostatic attraction to OTCs upon the addition of HA. Similar results were found in another study of NOR adsorption by MPs, in which the adsorption capacity of NOR by MPs increased with the increase of HA concentration when the concentration of HA was higher than 10 mg/L [199]. The main reason is that MPs interact with the aromatic structure of HA through π - π conjugation to form highly conjugated copolymers, resulting in larger intermolecular forces [161]. Overall, DOM in the environment affects the adsorption behavior of MPs and ATs from two perspectives. The first scenario is that DOM occupies adsorption sites on the surface of MPs, which in turn inhibits adsorption between the two. In the second case, the DOM in the environment wraps around the MPs, affects the spatial resistance and electrostatic effect of the MPs, and acts as a "bridge" to enhance the adsorption effect between the MPs and ATs.

5. Factors Influencing the Interaction between MPs and ATs

The interaction between ATs and MPs has various principles of action, mainly including hydrophobic interactions, intermolecular interactions, and electrostatic interactions, among other forces. The dominance of the above forces varies between different types of MPs and ATs, depending on MPs properties, ATs properties, and environmental factors. In addition, micropore filling, charge-assisted hydrogen bonding (CAHB), cation- π bonding, halogen bonding, and CH/ π interactions may occur between some specific types of MPs and ATs, but further studies are needed to confirm their involvement processes and importance. The sources and interaction mechanisms of MPs and ATs in aquatic environment are shown in Figure 6.



Figure 6. Sources of microplastics and antibiotics in aquatic environments and their mechanisms of interaction. (Note: Confirmed interaction mechanisms are in the green dashed boxes, and possible but unconfirmed interaction mechanisms are in the blue dashed boxes).

5.1. Hydrophobic Effect

Plastics are polymers formed from monomers by addition or condensation reactions, which determines the structure of MPs to be rich in alkyl groups and strongly hydrophobic. Hydrophobic interaction is the phenomenon by which hydrophobic groups cluster together to avoid water and is a weak, non-covalent interaction between non-polar molecules [202]. The strong hydrophobicity exhibited by MPs allows for a higher adsorption capacity for hydrophobic contaminants. For example, it has been suggested that the adsorption rate and capacity of PE particles on Sulfamethoxazole may be positively correlated with the hydrophobicity of the compound [203]. An experiment on the adsorption of ATs by MPs showed results indicating that the ability of different ATs to adsorb specific types of MPs varies greatly [162]. Except for PA, the adsorption of the other four MPs (PE, PS, PP, and PVC) on the five ATs decreased in the order of CIP > AMX > trimethoprim > sulfadiazine > TC. Coincidentally, the Log Kow values of the studied ATs decreased in the order of CIP > trimethoprim > AMX > sulfadiazine > TC. Definitely, exceptions cannot be ruled out, and the above correlation does not hold for PA, because PA has amide functional groups [204]. Hydrogen bonds can be formed between the amide group of PA and the carbonyl groups of AMX, TC, and CIP, when hydrogen bonding forces are dominant compared to hydrophobic interactions dominate. In general, the strength of adsorption of the four MPs to the five ATs, except PA, maintained a great correlation with the hydrophobicity of the ATs. A literature review shows that hydrophobic interactions occupy an important position in the adsorption process of MPs on ATs [171,173,195].

5.2. Intermolecular Interactions (Hydrogen Bonding, π - π Interactions, and Van Der Waals Forces)

The physicochemical properties of MPs and ATs with high detection rates in aquatic environments are shown in Tables S1 and S3, which will be more useful for understanding the hydrogen bonding, π - π interactions, and Van der Waals forces during the adsorption of ATs by MPs. Hydrogen bonding, Van der Waals forces, and π - π interactions have been identified as the driving forces affecting the adsorption of ATs on MPs [164,168,205]. Firstly, Van der Waals forces are a prevalent force between molecules and do not involve weak interactions forces that occur between molecules with covalent or ionic bonds. For example, aliphatic PE can only have intermolecular interactions with ATs by Van der Waals forces, while the adsorption of non-polar PP and PS on CIP can only be driven by Van der Waals forces, but PS containing benzene rings can also have π - π interactions [206]. Since MPs contain a large number of alkyl groups, the formation of hydrogen bonds is common in MPs, often formed between or within molecules, and hydrogen bonds are intermolecular interactions. For example, in a study on the adsorption of ATs by MPs, it was found that the adsorption of AMX, TC, and CIP by PA was higher than that of PE, PS, and PP [162]. The reason is that the amide group of PA can form hydrogen bonds with the carbonyl groups of AMX, TC, and CIP, which is the main reason for the high adsorption of PA [207]. π - π interactions are a special spatial arrangement of aromatic compounds, and MPs containing aromatic groups exhibit π - π interactions [208]. For example, aliphatic PS may have π - π interactions with ATs, and studies have shown that the stronger adsorption ability of aromatic PS to ATs may be caused by π - π bonds that enhance the interaction between MPs and ATs [32,173].

5.3. Electrostatic Effect

In electrostatic interactions, the interaction force between ATs and MPs is mainly due to their charge characteristics, and then, the attractive or repulsive force occurs [195]. The magnitude of the electrostatic force between MPs and ATs can be determined by the degree of ionization of the substance in the solution. It has been shown that the main factors affecting the electrostatic attraction/repulsion interaction between MPs and ATs are the pH of the solution, the pKa of the ATs, and the pHpzc of the MPs particles [203], because the pKa and pH of the solution may determine the isoelectric charge of the compound [209]. When the value of pHpzc of the MPs is lower than the pH of the solution, their surfaces

become negatively charged and readily attract positively charged chemicals, at which point the electrostatic effect of MPs on ATs manifests itself as a gravitational force and vice versa as a repulsive force [210]. Most MPs, such as PP, PS, and PE, have a pHpzc lower than the pH of most aquatic media (4.26, 3.96, and 4.30, respectively), which enhances the adsorption of positively charged ATs [200]. Overall, the relationship between the pKa of ATs, the pH of the solution, and the pHpzc of MPs determine the electrostatic attraction/repulsion interactions and may consequently affect the adsorption process between ATs and MPs.

5.4. Other Mechanisms of Action

The adsorption performance of biochar is related to its physicochemical properties such as its own surface parameters and microporous structure, which are similarly reflected in this study of the interaction mechanism between MPs and ATs. In a study on TC adsorption by PE, it was found that a large number of free binding sites (micropore filling) existed in PE for the adsorption of TC molecules in the initial stage of adsorption [187]. When all the adsorption sites on the PE surface are occupied, the adsorption rate of PE decreases significantly. Microporous filling generally occurs when the following two conditions are met [211,212]: (1) The number of MPs micropores is sufficient; (2) The MPs micropores are 1.7–3 times the size of ATs molecules. When the above conditions are met, micropore filling should be considered as an important mechanism of action.

CH/ π interactions exist between the hydrogen atom of the alkyl group (H donor) and the π -face of the aromatic ring (H acceptor), and they are considered to be an important driving force between polyolefinic substances and aromatic compounds [213]. Existing studies have found that alkyl groups (-CH) on PVC provide protons to the off-domain aromatic rings of bisphenol analogues, leading to changes in the adsorption behavior of bisphenols on PVC [214]. Therefore, MPs containing alkyl groups may undergo CH/ π interactions with aromatic ATs to enhance adsorption.

The halogen bond is formed between the halogen atom and the π electron of the benzene ring, which is a non-covalent interaction [215]. PVC branched chains contain halogen atoms Cl, which can behave as electron-deficient (electrophilic, electron acceptor) centers and undergo mutual attractive forces with electron-rich (nucleophilic, electron donor) substances [216]. Phenolic compounds contain benzene rings and hydroxyl groups that allow halogen bonding with PVC, which was also confirmed in a subsequent study [217]. TC, OTC, and AMX also have the same conditions for halogen bonding by containing benzene rings and hydroxyl groups, so we hypothesize that halogen bonding can also occur between MPs and ATs, but further evidence is needed.

Cation– π interactions are a key force in the adsorption of fluoroquinolone ATs on thermogenic carbonaceous materials. For example, on graphite and graphene-based materials, the amino group on the R3 ring of CIP obtains H+ and undergoes protonation, producing cation-bonds with electrons [218]. In this process, cation– π interactions induced species conversion (CIP (0) combines protons to form CIP (+1)), and the above process has been verified experimentally and theoretically [218]. MPs and ATs have conditions for the occurrence of cation– π interactions, but further experimental verification is needed.

CAHB refers to the charge distribution between two substances that form a hydrogen bond, as opposed to a normal hydrogen bond, which can directly form a CAHB with a much stronger bond than a normal hydrogen bond and with properties comparable to those of a covalent bond [219,220]. It was found that CAHB, as a class of low-resistance hydrogen bonds or salt bridges similar to cation bridges, are commonly found in environmental processes [219]. Meanwhile, the researchers borrowed CAHB to explain some unexplained experimental phenomena in the adsorption of ionic compounds by carbonbased adsorbents [221]. Under natural conditions, anionic ATs can undergo CAHB with MPs as the acceptor H- donor, and such a charge distribution enables the formation of more stable hydrogen bonds between the donor and acceptor due to the negative charge on the acceptor proton group, but the occurrence of this behavior requires a certain pH of the aquatic environment [218,219,221].

6. Conclusions and Perspectives

Taking the aquatic environment in the Chinese region as the research background, this paper explores the mechanisms and influencing factors of the interaction between MPs and ATs and synthesizes the effects of the interaction between MPs and ATs on the environment. The main findings are as follows: (I) The diversity of types of MPs and ATs leads to the diversity of their structures and properties, which is a key factor affecting the interaction of the two pollutants. The pollution of MPs and ATs in Chinese regions is mainly concentrated in Beijing-Tianjin-Hebei, the Yangtze River Delta, the Pearl River Delta, etc. The degree of pollution is related to the level of economic development, industrial structure, water allocation, climate, and other factors. In addition, the similarity in the regional distribution of MPs and ATs is mainly due to the similar sources and migration pathways of the two pollutants. (II) The adsorption of ATs by MPs is driven by a variety of mechanisms that have been confirmed by studies, including hydrophobic interactions, intermolecular interactions (hydrogen bonds, Van der Waals forces, and π - π interactions), and electrostatic interactions. (III) The process of interaction between MPs and ATs is influenced by several factors: pH and ionic strength mainly affect the electrostatic interactions between MPs and ATs to modify the adsorption behavior, and changes in salinity affect the solubility of nonpolar and weakly polar ATs in water and may affect the formation of hydrogen bonds. Aging MPs have changes in their surface morphology and microstructure, leading to changes in the adsorption behavior of MPs. In addition, the crystallinity, polarity, and size of MPs are also important factors that affect the adsorption of ATs by MPs. Based on the existing studies, we propose three directions for future research:

(1) Some factors influencing the interaction process between MPs and ATs are still unclear. For example, the influence of the crystallinity of MPs on the adsorption of ATs is controversial and needs to be further investigated. In addition, the morphology of MPs also affects the interaction with ATs. The present study shows that colloidal MPs have high surface area and have stronger enrichment effects on pollutants such as ATs in the water column, and that colloidal MPs have stronger mobility [136], so colloidal MPs may be an important direction for future research.

(2) The adsorption mechanism of ATs by MPs should be further investigated, including but not limited to hydrophobic interactions, hydrogen bonding interactions, π - π interactions, Van der Waals forces, electrostatic interactions, etc. In addition, the effects of MPs on the environmental behavior of other pollutants, such as organophosphates, heavy metals, perfluorooctane sulfonate, etc., should be carried out to gain a more comprehensive understanding of MPs as a new pollutant. In addition, the above studies are only at the laboratory stage and future research should focus on the sorption and environmental impacts of mixed pollutants in the natural environment, where laboratory results may differ significantly from those in the natural environment.

(3) China is the largest developing country in the world, and its production and use of MPs and ATs are among the highest in the world. The Chinese aquatic environment is facing serious pollution risks, and the establishment of pollution evaluation standards and treatment methods for MPs and ATs to better balance the relationship between economic development and environmental management is an important reflection of China's role in global environmental governance. Meanwhile, China's experience and research results on the management of these chemicals can provide useful references and suggestions for other countries.

(4) MPs and ATs as new pollutants have attracted extensive attention from scholars, but there is no uniform standard on the detection of MPs. In addition, how ATs may be extracted from MPs under natural conditions is also a difficult problem. Due to the variety and low concentration of ATs under natural conditions, extracting ATs from MPs accurately and correctly is a problem that needs to be solved urgently.

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