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Review

Microplastic–Pharmaceuticals Interaction in Water Systems

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Abstract: Microplastics, MPs, in aquatic environments pose serious threats when associated with other pollutants, such as pharmaceuticals, PHs. This review is a continuation of an earlier paper on the role of MPs as containers and carriers of heavy metals, HMs, persistent organic pollutants, POPs, pharmaceuticals, PHs, and personal care products, PCPs, in marine environments and published in the *Journal of Marine Science and Engineering*. The current effort aims to elucidate the most recent data on the interaction and association of MPs with PHs and the ecotoxicological implications on food webs. This review focuses on the nature of the interaction from different perspectives, such as the hydrophobicity and hydrophilicity of the polymer and drug, the polymer surface, and the rate of weathering. The effects of environmental conditions, such as mechanical stress, photodegradation, pH, salinity, dissolved organic matter, and gastrointestinal features of marine biota, were reported. This review reports on experimental laboratory, mathematical, and field data on MPs' carrier and accumulation role in PHs and their release and ecotoxicological effects on water bodies. From the survey of the data, it emerges that the nature of the interaction and the effects on biota are very complex and variable, and perhaps only a systematic mechanic approach of data collection with a statistical approach using big data and deep learning will contribute in the future to clarify.

Keywords: pharmaceuticals; microplastics; contamination; water; sediments; food webs; toxicity

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1. Introduction

The world is producing twice as much plastic waste as two decades ago, with the bulk of it ending up in landfills, incinerated, or leaking into the environment and only 9% successfully recycled according to a new OECD, Organization for Economic Cooperation and Development, report [1]. In 2010, global primary production of plastic was 270 million tonnes, but only plastic waste improperly managed (mismanaged) is at a significant risk of leakage to the environment [2]. In 2010, this amounted to 31.9 million tonnes. Of this, 8 million tonnes, 3% of the global annual plastic waste, entered the ocean, and most of it conferred in landfills [2]. Plastics enter oceans mainly through domestic–agricultural–industrial sewages and marine-based activities [3–8]. Along their trip from the sources to the receiving media, plastic items undergo a process of chemical and physical weathering, i.e., mechanical disaggregation, wave and wind abrasion, hydrolysis, thermal degradation, ageing from UV radiation, and biodegradation [9], forming microplastics, MPs [10]. MPs in the water represent a major worldwide issue due to their high mobility and low degradability [11].

Before reaching freshwater courses and seas, they accumulate in high amounts in wastewater treatment plants where they often meet other pollutants. The co-occurrence favours polymer sorption of chemicals, such as polycyclic aromatic hydrocarbons, PAHs, polychlorinated biphenyls, PCBs, polybrominated diphenyl ethers, PBDEs, and pharmaceuticals, PHs [12–14]. Polymers can adsorb and therefore concentrate thousands of times more contaminants than surrounding waters due to their extensive solid surface and strong surface hydrophobicity [15–17]. Pellets, e.g., can concentrate pollutants up to six-fold their concentration in seawater [18].

A topical debate argues if the interaction with xenobiotics and movement up to remote areas is significant and if such interaction increases the potential toxicity through pollutant transfer to the aqueous phase or food sources and desorption in the digestive system [19–22]. The vehiculation might influence reproduction, immune response, oxidative stress, cellular toxicity, inflammation, and cancer [23]. However, as recently reported by the author [24,25], there is no unique opinion if vehiculation and bioavailability are significant.

The presence of PHs in water environments attracted the attention of scientific research over the past two decades [26]. PHs in freshwater and seawater are usually derived from wastewater effluents, discharges of livestock and poultry wastes in farmlands, and runoff from aquaculture fields [27], groundwater, and even drinking water. They represent complex organic compounds that can be decomposed into harmful, persistent secondary by-products [17].

Thus, MPs and PHs are considered emerging and ubiquitous contaminants in water bodies, and their interaction has a complex chain effect where one factor drives the other and so on [28,29]. Sorption depends on features of polymers, such as size, colour, morphology, surface area, crystallinity, functional groups, hydrophobicity and PHs as charge, and hydrophobicity. Other significant factors regard the conditions of the environment, such as pH, salinity, dissolved organic material, and dissolved organic carbon, DOC [30]. Moreover, the properties of pristine polymers change, and sorption varies with weathering, ageing, and biofilm formation [31]. Compared to hydrophobic pollutants, PHs have unique features: Only a few are hydrophobics, such as albendazole and clofibric acid, and most are polar, ionizable, and hence, have pronounced hydrophilicity with low octanol-water partition coefficients, Kow [32] and interact mainly via electrostatic and hydrogen bonding [33].

The issue of PH sorption in aqueous ecosystems on MPs has been only recently explored [34,35], and mechanisms explaining the sorption behaviours of MPs need to be clarified. The nature and type of interaction, the consideration of abiotic factors, such as temperature, UV exposure, salinity, pH, the nature of PH and MP polymers, and DOC, remain unclear [14,36]. The complex interaction and the associated molecular-level mechanism of hydrophilic compounds are somewhat unique and need further assessment since the interaction changes the transport, bioavailability, biota toxicity, and biomagnification through the food chain [37].

The current survey aims to clarify how MPs interact with drugs in water bodies, the health effects and risks associated with PH vehiculation, and the ecotoxicological consequences. This study provides a critical review of the information from the literature related to the occurrence, interaction, and toxic effects of PHs–MPs. This was initiated by conducting a literature search on the Science Direct platform focusing on articles published from 2020 to 2023. This review aims to be an agile tool for consulting scientific news referring to a limited time span.

2. Pharmaceuticals in Aquatic Systems

About 160 drugs, including antibiotics, non-steroidal and anti-inflammatory, NSAIDs, psychiatric, and cardiovascular [37], have been revealed in waters with a net dominance of antibiotics because of their overuse and misuse [38]. Without policy intervention, global antibiotic consumption in 2030 could be 200% higher than in 2015 [39]. It is estimated that up to 15,000 tons of antibiotics are released annually into the European environment and have a high possibility of interacting with aged MPs because of their hydrophilic, oxygen-containing functional groups [24]. Three classes of antibiotics are very common in coastal water, sulfonamides (SAs), quinolones (QNs), and macrolides (MLs), including erythromycin, oleandomycin, and spiramycin. The most diffused antibiotics in oceans are oxytetracycline, tetracycline, sulfamethoxazole, ciprofloxacin, and trimethoprim, which are endocrine disruptors, and they impact crustaceans and fish, alter the levels of hormones in the blood, and can induce cardiovascular collapse [24]. In European coastal superficial water, the most common antibiotics are erythromycin, amoxicillin, ciprofloxacin, sulfadiazine, and clarithromycin [24]. One hundred thirteen pharmaceuticals and pharmaceutical

metabolites were detected in coastal waters [24]. Marine and offshore environments can be impacted in their waters and sediments by the presence of many groups of antibiotics from aquaculture, land farming, and wastewater [24]. Antibiotics are feared in all types of water because (i) they induce resistance; (ii) resistance genes transfer horizontally between microbial populations, and (iii) antibiotics can have direct wide-spectrum toxicity to organisms [24].

It has been reported that approximately 50–90 percent of antibiotics administered by humans or animals are excreted via urine and faeces as a mixture of the parent compound and their metabolite forms (e.g., glucuronides) [40]. Then, these compounds are carried to WWTPs where some of them can be partially eliminated or pass the process unchanged. Tran et al. [40] report that the concentration of antibiotics in wastewater treatment plants', WWTPs', influent and effluent samples varied significantly from below the method quantification limit (MQL) to a few tens of micrograms per litre depending on the compound, type of wastewater sample, sampling site, and sampling date. Among the investigated antibiotic classes, sulfonamides, fluoroquinolones, macrolides, and trimethoprim frequently were detected in both influent and effluent samples worldwide.

3. MPs in Aquatic Systems

MPs have been identified in surface water lagoons and estuaries, coastal shorelines, pelagic and benthic regions of the sea, Arctic freshwaters, ice, and the ocean [41]. Within aqueous environments, MPs provide a large, solid surface, especially for hydrophobic, organic contaminants up to several orders of magnitude higher than those in the surrounding waters. Common detected MPs in water bodies are polyethylene, PE, polypropylene, PP, polyamide, PA, polyvinyl chloride, PVC, polystyrene, PS, and polyethylene terephthalate, PET [42,43]. Nylon, or PA, is one of the most common polymers detected in WWTPs due to its wide use as an essential thermoplastic in the automotive trade, the manufacture of textiles, and wind turbines [44]. Freshwater environments exhibit higher MP concentrations than marine environments. Among the water environments, most studies have been conducted on the abundance and characteristics of MPs in seawater. Only recently, studies on MPs in various water environments, including freshwater, wastewater, and groundwater, are being conducted [41]. Depending on the inherent properties of MPs (density, hydrophobicity, recalcitrance) and environmental factors, MPs can easily move between environmental media, and high-density MPs can settle and accumulate at the bottom of the water environment. Due to this phenomenon, it is difficult to investigate the concentration and characteristics of microplastics in overall water environments [41].

In general, the concentration of MPs is higher nearshore or in an estuary adjacent to land than in the open sea [41], and the concentration of MPs is higher in seas with geographical characteristics such as in semi-enclosed bays than in the open seashore [41]. Using data of the MP composition in the central Mexican Pacific according to their shape, it was confirmed that the fragments were composed of PP or PE; the fibres were composed of PE or polyester (PES), and the films were made of PE [41]. MPs found in seawater are composed of PE and PP [41]. Both types of plastics account for 60% of global plastic production. Both PE and PP can be attributed to the wear and tear of fishing gear, which is generated due to high-intensity fishing activities [43]. In WWTPs, most MPs are removed during primary treatments, including solid skimming and sludge settling processes. As the MP removal rate is affected by their size, shape, and type in wastewater treatment [41], the removal rate of MPs may vary among studies. MP emissions from WWTPs are non-significant [41], suggesting that there are other sources of pollution (e.g., marine industrial waste, land-based plastic litter from rainfall, worn tire tread from the atmosphere) that can enter the aquatic environment. While extensive research has been conducted on seawater MPs, there is a lack of information on freshwater MPs [41]. Nevertheless, research on the abundance and behaviour of MPs in freshwater is gaining momentum. The importance of freshwater cannot be underestimated given that it is a source of water that is supplied to households through water treatment. Land-based sources are essential contributors

to environmental MPs, and stormwater retention ponds also play a role in transporting microplastics from land to the aquatic environment [41].

4. Association of PHs–MPs

Even though MPs are considered relatively inert, in the presence of PHs, they can participate in many interaction processes, such as aggregation, adsorption, and transformation, that can have synergistic, adverse, or potentiating effects [45]. MPs have various morphological features, such as colours, sizes, shapes, and different polymeric compositions, functional groups, and surface charges. The charges may vary after interaction with PHs and exposure to media conditions, forming several aggregates [46].

The nature of MPs, especially their hydrophobicity and large surface area, provide these materials with a high adsorption potential [17,47]. The literature offers many examples of the association. Qu et al. [48] reported significantly higher levels of venlafaxine in simulated aqueous systems with MPs than without. In the same way, the adsorption of five antibiotics on different polymers in freshwater and seawater systems was studied by Li et al. [35]. Adsorption of antibiotics on MPs may result in their long-range transport and cause compound combination effects. In this study, the authors [35] investigated the adsorption of [sulfadiazine (SDZ), amoxicillin (AMX), tetracycline (TC), ciprofloxacin (CIP), and trimethoprim (TMP)] on five types of MPs [polyethylene (PE), polystyrene (PS), polypropylene (PP), polyamide (PA), and polyvinyl chloride (PVC)] in freshwater and seawater systems. Scanning electron microscope (SEM) and X-ray diffractometer (XRD) analysis revealed that microplastics have different surface characteristics and degrees of crystallinity. The degree of crystallinity influences their mechanical properties: Higher crystallinity results in a harder and more thermally stable MP but also a more brittle material, whereas the amorphous regions provide certain elasticity and impact resistance [24]. These polymer properties make pollutants more accessible inside the amorphous regions where chains are disordered aligned and with empty voids and free volume increasing diffusion of hydrophobic chemicals between polymeric chains. Hydrophilic chemicals can easily access high-crystalline polymers, such as PS, and hardly diffuse into low-crystallinity polymers, such as PP and PE [24]. Adsorption isotherms demonstrated that PA had the highest adsorption capacity for antibiotics with a distribution coefficient (Kd) value ranging from 7.36 ± 0.257 to 756 ± 48.0 L kg^{-1} in the freshwater system, which is attributable to its porous structure and hydrogen bonding. A relatively low adsorption capacity on the other four microplastics was observed. The adsorption decreased in the order CIP > AMX > TMP > SDZ > TC with Kf correlating positively with octanol–water partition coefficients (Log Kow). Compared to the freshwater system, the adsorption capacity in seawater decreased significantly, and no adsorption was noted for CIP and AMX. The authors reported that commonly observed polyamide particles may serve as a carrier of antibiotics in aquatic environments [35].

Due to specific polymer–pollutants interaction, there is no rule to predict sorption ability. Xu et al. [49] studied the accumulation of tetracycline on PS, PP, and PE, revealing that the adsorption was polymer–drug specific. For example, it has been reported that the sorption of antibiotics, such as sulfamethoxazole [50,51], to PS, PE, PP, PVC, and polyethylene terephthalate (PET) was lower than sorption to polyamide, PA. This was related to the PA porous structure and complex hydrogen bonds [52].

4.1. Mechanisms of Interaction

Atugoda et al. [17] summarized that PH sorption onto MPs relies on six types of interaction: hydrophobic, electrostatic, H-bonding, van der Waals force, pore filling, and π–π, Figure 1. The hydrophobic interaction occurs if both the PHs and polymers are hydrophobic with a high log Kow, a value > 4, which is less attracted by water molecules [45,53]. However, most PHs present hydrophilic and polar groups in their structure, which indicates mechanisms other than hydrophobic ones. If they are hydrophilic and of non-aromatic/aliphatic origin, as most of them are, electrostatic and van der Waals attraction

forces occur [54]. The π–π interaction comes into play in the interaction between aromatic MPs and PHs [55]. Hydrogen bonding is less common and occurs between a very electronegative atom and a hydrogen atom covalently bonded to a very electronegative atom, N, O, or F atom [35]. For instance, ciprofloxacin, amoxicillin, tetracycline, sulfamethoxazole, and steroidal hormones bind to PA microplastics through hydrogen bonding [56]. Electrostatic interactions, hydrogen bonds, and van der Waals are weaker than covalent and ionic bonds [57] and are dominant in MPs–PHs interaction [58]. Table 1 summarizes recent findings on the mechanisms of interaction of MPs with PHs. Generally, PE, polystyrene PS, polypropylene PP, and PVC MPs are the most studied polymers for which two main adsorption mechanisms are reported: hydrophobic and electrostatic interactions.

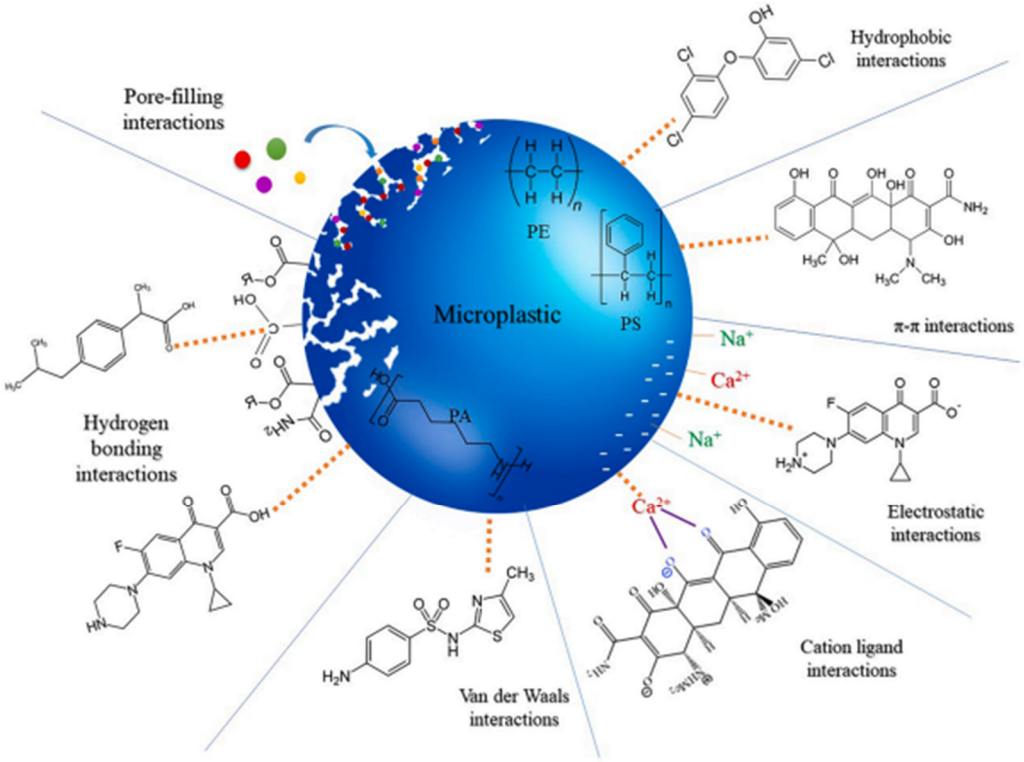

Figure 1. Different types of interface interactions between PH and MP surfaces. Reproduced from [17] with permission of Elsevier 2023.

Table 1. Mechanisms of interaction of MPs with PHs.

	Pharmaceutical	Interaction Mechanism	Reference
Polyamide	Sulfadiazine, amoxicillin, tetracycline, ciprofloxacin, trimethoprim	Hydrogen bonding, hydrophobic interaction, van der Waals force, and electrostatic interaction	[35]
Polyethylene	Tetracycline	Electrostatic interactions, hydrophobic interactions, π–π interactions, and polar interactions	[49]
Polyethylene	Ciprofloxacin	Hydrophobic and electrostatic interactions	[59]
Polyethylene	Sulfadiazine, amoxicillin, tetracycline, ciprofloxacin, and trimethoprim	Hydrogen bonding, hydrophobic interaction, van der Waals force, and electrostatic interactions	[35]
Polypropylene	Tetracycline	Electrostatic interactions, hydrophobic interactions, π–π interactions, and polar interactions	[49]
Polypropylene	Sulfadiazine amoxicillin, tetracycline, ciprofloxacin, and trimethoprim	Hydrogen bonding, hydrophobic interaction, van der Waals force, and electrostatic interactions	[35]

Table 1. *Cont.*

	Pharmaceutical	Interaction Mechanism	Reference
Polystyrene	Oxytetracycline	Electrostatic interaction, multivalent cationic bridging mechanisms, and H-bonding interaction	[60]
Polystyrene	Tylosin	Electrostatic interaction, surface complexation, and hydrophobic interactions	[54]
Polyvinyl chloride	Ciprofloxacin	Intermolecular hydrogen bonding, partitioning, and electrostatic interactions	[58]

4.1.1. Hydrophobic Interactions

This mechanism mainly concerns antibiotics, which are normally neutral molecules and can contact neutral polymers, such as PE and PP, or weakly polar ones, such as PS. The hydrophobic non-covalent forces cause the aggregation of non-polar moieties in water [61].

4.1.2. Electrostatic Interactions

In addition to being neutral zwitterionic molecules, PHs, depending on the pH, can exist as cations and anions and interact with polar functional groups of MPs, such as PA and PVC, through electrostatic interaction. This mechanism represents a possible interaction pathway with an MP pH PZC < pH of the average environmental conditions and hence bearing a net negative charge [62]. If the pH increases, the adsorption decreases as solutes shift from positively charged to negatively charged species [35]. Polymers with a polar group display dipole–dipole or dipole-induced interactions with the polar groups of drugs [63]. This type of interaction will be of minor importance in the case of non-polar polymers, such as PE, and assumes importance when plastic degrades with the formation of oxygen-containing groups, such as carboxyl, ester, and ketone groups on polymer [55].

4.1.3. π–π Interactions

This interaction involves non-covalent bonds in aromatic compounds in the polymer backbone as in that of PS and the aromaticity of the drug, ciprofloxacin, tetracycline, trimethoprim, and sulfadiazine [35].

5. Factors Driving the Interaction

The main factors can be divided into four categories: (1) polymer type, such as structure, crystallinity, and surface charge; (2) ageing; (3) physical–chemical properties of the pharmaceutical, pKa and Kow; and (4) environmental conditions of the water, such as temperature, pH, salinity, ionic strength, dissolved organic matter, biofouling, and biofilms [64,65]. Many of these properties are closely related to each other, e.g., water pH, which may change the ionization states of PHs and influence their sorption processes on MPs.

5.1. Polymer Type

Each polymer has its size, specific structure, crystallinity, and chemical composition in terms of functional groups, acid and base properties, surface charge, surface area, and molecular arrangement, all features that influence interactive mechanisms [54].

5.2. Particle Size

Particle size plays a minor role compared to other physical parameters [62]. Normally, PH sorption increases for smaller particles with a high surface area [36]. But this is not the rule in some cases: Xu et al. [54] reported the least sorption of tetracycline by PE and a general decrease in sorption as particle dimension reaches the nanoscale size [64] due to aggregation. Furthermore, the variation in particle size affects the time required to reach equilibrium. Particle size may also affect the adsorption/desorption rate and the rate of

equilibrium stabilization with small particles increasing diffusion rates and attainment of steady-state equilibrium conditions [64].

5.3. Structure and Cristallinity

The structure relies on the order of the polymer chain and intermolecular attraction. Polymers rich in rubber have higher interaction and hence adsorption capacity with PHs [66] due to a large void or free space inside the chain, facilitating sorption [67]. According to the glass transition temperature, Tg, there are polymers considered rubbery, such as PE, while PVC and PS are considered glassier. Elizalde-Velazquéz et al. [36] report that non-steroidal, anti-inflammatory drugs, i.e., naproxen, ibuprofen, and diclofenac, have a high affinity with the rubbery domains of PE rather than the glassy sites of PP and PS [68]. Although PS has a robust bond, π–π interaction, and a large void inside the polymer chain, the absorption capacity of PE is higher than PS because of the rubbery structure of PE. The authors report that glassier plastics, such as PET, PVC, and PS, have stiffer chains and higher intermolecular attraction, resulting in more closely positioned chains and a small free volume [24].

The crystallinity of polymers can be categorized as amorphous, crystalline, and semi-crystalline according to how the polymeric chains are arranged. They are crystalline when they are made of tightly structured and well-organized molecular chains with a range of crystallinity varying from 0 to 90%. The rate of PH adsorption increases for polymers with a low degree of crystallinity [69]. However, polymer crystallinity does not seem significant for sorption concerning other intrinsic properties of MPs, such as polarity, size, and weathering [17].

5.4. Surface Charge

Surface charge is influenced by the functional groups and affects the adsorption of the more polar compounds. Antibiotics like sulfamethoxazole, ciprofloxacin, amoxicillin, or tetracycline have a higher affinity for PA–MPs due to the presence of amide in their structure [37]. Surface charge and hence the chemical composition of the polymer is strongly influenced by the environmental conditions and, first, by ageing and weathering processes. MPs divide into two groups: non-polar aliphatic polymers, such as HDPE, LDPE, and PP, containing monomers with only C and H atoms with C–C and C–H covalent bonds, such as in PE, giving high structural strength and chemical resistance. Other polymers can be polar due to the incorporation of polar functional groups in the C and H backbone or side chains. This is the case of PA, PS, PC, PVC, and PP having polar functional groups CO–NH–, benzene, –COOH, –Cl, and $–CH_3$ [54].

5.5. Physical–Chemical Properties of Drugs

PHs are amphoteric compounds and depending on the pH, can be in multiple species as cations, anions, or zwitterionic molecules, and thus hydrophobicity generally is somewhat low, log Know < 1, and are thus considered hydrophilic or polar compounds [70]. There are some exceptions, such as sertraline with a Kow of 5.29. Thus, their sorption mechanism is different from that of hydrophobic compounds. Non-polar/weakly polar substances are adsorbed by physical entrapment in the inner matrices of the MPs, while polar compounds are retained by surface adsorption [58]. This results in significant differences in bioavailability and mobility.

6. Environmental Factors

6.1. Effect of Temperature

Temperature can modify solubility, surface tension, and sorption thermodynamics and hence the interaction and especially in extreme temperatures above 45 °C or below 0 °C. High temperatures, 75 °C, during ageing may favour the presence of oxygen-rich functional groups C–O, C–OH, and CO [71] and the interaction with hydrophilic PHs as well as their desorption [72].

6.2. Effect of pH

pH represents the most important factor regulating MP sorption since it affects MP surface charges and drug speciation as well [73]. A significant group of drugs, such as antibiotics, are polar and ionizable, and their speciation is driven by pH. Thus, drugs can be cationic, anionic, and zwitterionic species and will be sorbed depending on the surface charge [74]. They are 85–95% ionizable and weak acids or weak bases [75]. The electrostatic interaction is the main sorption mechanism of PHs by MPs. This is a physical mechanism driven by the pH of the aquatic system, normally 5–9, when the surface charge of the MPs varies among positive, negative, or neutral, and the polymers tend to aggregate [73]. However, at the common pH value of estuaries and coastal water, 8.1, there would be a tendency for many PHs to desorb from the MPs due to the repulsion [76].

The point of zero charges, pH PZC, of MPs is below 7, and hence, at the common pH of water systems, the surface is negatively charged. The pH dependence of the surface charge was shown by the point of zero charges, PZC, experiments: When the pH of the system was above the PZC of PE, PP, and PS, the polymers carried negative charges [49,77]. The effect of solution pH on the sorption of tetracycline by PE, PP, and PS was studied by Xu et al. [49]. Tetracycline is present in cations at low pHs, anions at high pHs, and zwitterions at pH 5–7, with the highest electrostatic attractions for the zwitterionic species and the maximum sorption at a pH level of 6. When the pH increased, sorption decreased as tetracycline and polymers became negatively charged and repulsed. In the same way, Guo et al. [50] showed that the sorption of sulfamethoxazole increased with pH from 3 to 5; at pH 5–7, sulfamethoxazole is neutral and sorbed by hydrophobic interactions with the non-polar polymer surface. At pH > 7, sulfamethoxazole is anionic and is repulsed by the increased electronegativity of the polymer. This is explained by the electrostatic and hydrophobic attractions since the antibiotics became positively charged in an acidic pH, whereas the electrostatic interaction became less significant at an alkaline pH as the antibiotics became negatively charged. Zwitterionic or neutral drugs interact with non-polar plastic surfaces through hydrophobic and van der Waals forces at a neutral pH. At a high pH, as in marine environments, drugs are present as anions, which enhances their electrostatic repulsion with polymer surfaces. Thus, the pH of the aqueous system affects zwitterionic strength and electrostatic repulsion. Other similar results are shown in the literature. Guo et al. [54] studied the sorption of Tylenol on PS, PVC, PE, and PP. They found that the sorption capacity in the case of PS and PVC decreased gradually with the increment of pH from 3.0 to 7.0, while PE and PP showed a minimal difference. This is likely due to the electrostatic attraction. In another study [78], the pH decreased to less than 3.0, and protonation of PE and PS surfaces and enhanced adsorption of PFOS, perfluoro octane sulfonic acid, was observed. Thus, the pH is critical in influencing the sorption process between MPs and PHs depending on their surface charges [79]. Elizalde-Velázquez et al. [36] studied the sorption of three non-steroidal, anti-inflammatory drugs, ibuprofen, naproxen, and diclofenac, on PS, ultra-high-molecular weight PE, average molecular weight, medium density PE, and PP. The sorption process was pH-dependent, probably due to the molecular equilibrium effects that affect not only MPs' surface charge but the compound's speciation as well. Only under acidic conditions were the pharmaceuticals highly sorbed onto the MPs, a process ruled by hydrophobic interactions. Thus, solution pH affects overall chemical reactivity, biochemical and physicochemical properties, equilibrium condition, and toxicity [80]. McDougall et al. [81] studied the desorption of three groups of PHs, cationic, anionic, and neutral, from PE-simulating gastric and intestinal fluids. The pH of the solution was the most important factor driving PH desorption, influencing speciation of the active compounds and MP surface charge. The desorption of the cationic compounds in gastric fluids was more evident and up to 50% due to the reduced surface charge of the polymers under low pH, revealing that PHs sorbed to MPs can be bioavailable [81].

6.3. Effect of Ionic Strength

The role of ionic strength on PH sorption depends on the type of adsorbent, adsorbate, electrolyte, and solution chemistry [17]. Sorption decreases with ionic strength, as was the case of ciprofloxacin [17], sulfamethoxazole [74] and sulfamethazine. Polymers are normally negatively charged and hence hydrophilic and influenced by the levels of cations, such as Na^+ and Ca^{2+}, in an aqueous medium that can bind electrostatically, disturbing the charge equilibrium of surfaces. Salts also increase the viscosity and density of water, hindering the mass transfer of drugs with ions being sorbed more easily [54]. Wan et al. [82] observed that $MgCl_2$ promoted the aggregation of PS and hence decreased tetracycline sorption via electrical double-layer compression or elimination and modulating repulsive forces and reducing accessibility to PHs [17].

An opposite effect of ionic strength can be also observed: the so-called salting-out and ionic complexation effect [83]. Lu et al. [76] observed a threefold increased sorption of 17β-estradiol and 17α-ethynylestradiol when the salinity of seawater was doubled and explained using solubility decreases of hydrophobic PHs with an increase of salts and the enhanced hydrophobic interactions of drugs with MPs. The presence of ions also promotes cationic bridging, especially with multivalent ions. On the other hand, polyvalent ions may favour the formation of ternary complexes through the bonding of the drugs with specific PH functional groups of the adsorbent, which enhances the sorption properties [84]. In the case of non-steroidal, anti-inflammatory drugs, NSAIDs, Elizalde Velasquez [36] reported higher sorption of ibuprofen, diclofenac, and naproxen on PP, PS, and PE in freshwater than that in seawater due to the high salinity content of seawater. The increase in salinity affects the aggregation state of the polymer. In parallel, it also increases the hydrophobic interaction between the drug and the polymer via the salting effect, i.e., decreasing the concentration of PHs in the solution [55].

6.4. Effect of Dissolved Organic Matter

The effects of dissolved organic matter, DOM, the fraction of organic matter in a solution that passes through a 0.45 µm filter, on the binding of drugs are poorly studied [36]. DOM can induce contrasting effects on PH sorption by MPs in the function of their properties enhancing or reducing sorption [49]. Few studies show increased sorption. Zhang et al. [60] studied the sorption of oxytetracycline on beached polystyrene foams in the presence of humic acid, HA, and fulvic acid, FA, as the two representative dissolved organic materials and showed increasing sorption with DOM. They found that fulvic acid promoted significant oxytetracycline sorption to the beached foams due to the complexation role of humic acid, acting as a bridge with both the beached foam surfaces and oxytetracycline molecules. Lu et al. [76] recently observed increased sorption of hormonal steroid compounds by MPs due to DOM–MPs complexation. Chen et al. [85] showed the formation of a copolymer between the carbonyl group of DOM and the aromatic moieties of the polymer through π–π interaction, which enhanced the electrostatic sorption of the positively charged oxytetracycline to the copolymer. Migration and stability of MPs might be considerably aided by DOM [86] due to the formation of a coating, eco-corona layer on the MPs, which inhibits the aggregation of plastic particles via electrostatic repulsion and steric forces [86]. However, mobility and stability can be reduced with the presence of DOM, as they can create a coating layer on the plastic surface. There are studies by Xu et al. [49] reporting decreased sorption of tetracycline on PS, PP, and PE when DOM increased due to complexation with the hydrophobic or hydrophilic DOM parts, which changes the partitioning between the polymer surface and water [87]. DOM and drugs may compete for the limited sorption sites on MPs [88] with the desorption of PHs from MPs [89] or can block the pores on the MPs' surface with their large molecules, hindering the access of drugs [90]. Another interactive role is played by the concomitant presence of polyvalent metals: Through PHs, stable ternary complexes can form in the water metal cations and DOM [91].

6.5. Effect of Biofouling

Biofouling is the process of microbe colonization of MPs through extracellular polymeric substances, EPS. This induces changes in morphology and physicochemical properties [92]. There is still very limited knowledge on the effect of biofouling on MPs–MPs interatom. PHs must pass from the water into the biofilm and then to the polymer material [93]. PHs interact via hydrophobic partitioning into the biopolymer or through binding onto the sorption sites of the heterogenic EPS [94]. EPS is rich in ionizable functional groups, such as carboxyl, phosphoryl, amino, and hydroxyl, that increase the sorption of metal ions via MPs [95], and in this way, metals have a synergistic effect on the sorption by forming a PHs–metal–EPS complex through an ionic bridging effect [96].

6.6. Effect of MP Ageing

MPs in water are exposed to physical and chemical weathering, such as photooxidation, temperature variations, friction, and saltwater corrosion. They crack into smaller particles with reduced hydrophobicity [73]. Ageing occurs through ultraviolet-induced photodegradation, physical impacts, and biodegradation [97]. Many kinetic models and batch sorption data reveal how modified MPs have a higher sorption capacity than pristine polymers [73]. Weathering processes increase surface area, accessible binding sites, and hence PHs sorption [97]. In this regard, however, the data are not always in agreement, Table 2. Huang et al. [97] studied the role of MP ageing, PS, on the interaction with sulfamethoxazole, SMX, and the β-blocker propranolol (PRP) and using red tilapia as the model fish. The authors reported a 0.27- and 0.16-fold increase in the specific surface area and average pore volume, respectively, and the formation of more carbonyl on the aged PS. Ageing increased PRP accumulation by 82.3% in the brain, whereas it decreased the SMX level by 46.1% in the gills. Even the response of the model was different: In the case of PS–PRP, the stress was alleviated by the ageing with reduced neurotoxicity and lipid peroxidation damages, whereas in the case of SMX-aged PS, co-exposure resulted in higher inhibition of cytochrome P450 enzyme activities. Thus, the interactive effect of MPs and the drug varies with the intrinsic features of the drug, exposure strategy, selected endpoints, biological models, and environmental conditions. One key issue, as stated by Huang et al. [97], is that there is no significant literature on the interactive effects of aged MPs and drugs on aquatic organisms where the experiments are carried out with commercially pristine MPs, that is, with homogeneous size distribution and regular shape. If ageing in laboratory settings has shown evident increases of SSA and the oxygen load and consequently more intensive drug sorption on polymers, it is very likely to hypothesize side chain effects of the toxicological interaction between MPs and drugs.

Table 2. Interactive effects of MPs–PHs on various aquatic organisms.

Drug	MPs	Organism	Effect	Reference
Roxithromycin	PS (10–100 μg/L)	*Red tilapia*	Reduced inhibitory effect	[98]
Cephalexin	No-specified (0.184 mg/L)	*Polatoschistus microps*	Increased inhibitory effect	[99]
Procainamide	No-specified (0.75–48 mg/L)	*algae*	Increased inhibitory effect	[100]
Venlafaxine	PVC (50 mg/L)	*loach*	Increased inhibitory effect	[101]
Propranolol	PS (10 μg/L)	*Red tilapia*	Reduced inhibitory effect	[97]
Sulfamethoxazole	PS (50 μg/L)	*Red tilapia*	Increased inhibitory effect	[97]

The effects of the biotic and abiotic degradation events depend on the chemical nature of the plastic, the loads, the hydrophilicity of the surfaces, and the environmental conditions [102].

In water, the excess of hydrogen atoms favours the formation of a phenolic hydroxyl group on aged MP surfaces [103]. Through chemical transformation, the richness of oxygen-containing functional groups increases, carboxyl, hydroxyl, ketone, and ester, and this confers polymers hydrophilic properties [104], making them capable of more adsorb

the hydrophilic molecules of PHs [29]. If the weathering process increases, the negative charges of the polymer surfaces and the sorption of cationic PHs via electrostatic interaction increases [73]. The increased hydrophilicity was highlighted by Liu et al. [58] who showed functional group alteration on PS and PVC via UV irradiation and ciprofloxacin sorption increase. Zhang et al. [60] also showed a twofold increase in the adsorption capacity of beached PS foams for oxytetracycline, OTC, concerning virgin PS foams, and the adsorption included electrostatic interaction, multivalent cationic bridging mechanisms, and H-bonding interaction. In the same way, ageing can also sustain other mechanisms, such as π–π, electrostatic interaction, hydrogen bonding, ion exchange, and complexation [60]. Other authors, reported decreased desorption of atorvastatin and amlodipine from weathered PS due to the increased electrostatic interaction [79].

Ageing also modifies polymer crystallinity since the weathering process enhances the percentage of the crystallization region and reduces the amorphous plastic portion, but the effects on PHs sorption are poorly known [105].

7. Sorption Models

Linear, Freundlich, and Langmuir models have been used to describe sorption depending on the type of polymer and drug. Sulfamethoxazole sorption on PE fitted both linear isotherms and a pseudo-second-order kinetic model [49]. Tylosin adsorption isotherms were described by the Freundlich equation rather than the Langmuir model and also fit the pseudo-second-order model [54]. Furthermore, Godoy et al. [43] conducted an extensive study on the sorption and desorption of PE, PET, PP, PS, and PVC towards amoxicillin, paracetamol, and vancomycin. The kinetic study revealed that the sorption process was slow, 28 days for amoxicillin to reach equilibrium. The modelling showed a better fit of the Langmuir model. The sorption and desorption of sulfamethoxazole, propranolol, and sertraline by PE MPs in water were investigated by Razanajatovo et al. [106]. After 96 h, the sorption decreased in the order of sertraline 28.61% > propranolol 21.61% > sulfamethoxazole 15.31%, and the sorption kinetics fitted the pseudo-second-order model. The authors concluded that hydrophobicity and electrostatic interactions ruled the sorption process. Sorption kinetics are strictly related to the initial PH concentration with rapid sorption at low adsorbate concentrations and slow sorption when active sites become saturated, and sorption rapidly reaches a steady-state equilibrium [16].

8. Bioaccumulation

MPs can be ingested by planktonic organisms, planktivorous fish, and benthic invertebrates, impacting the feeding rate, growth, development, obesity, oxygen consumption, chromosomal alteration, impaired reproduction, and cancers [107]. Most research examining the impact of MPs on biota focuses on fish species with the observed effects ranging from oxidative stress and disruption of metabolite composition [108] to strong inflammatory responses and tissue damage [109]. PHs exert similar negative impacts on water organisms, such as decreases in reproduction and growth, oxidative stress, changes in behaviour, disturbed circadian rhythm, decreased locomotion, and a decrease in survivor rate [110]. Due to the continuous exposure of aquatic organisms to co-contaminated MPs, their adverse effects can be magnified [111]. Both pollutants enter the tissues or the circulatory system and depend on the physiological conditions and features of MPs and PHs. Some authors [112] define MPs as tiny "Trojan horses" and hence as a hidden source of contaminants, as they can be unintentionally taken up by biota. The effect of sorption can have enhanced or reduced effects on the distribution, metabolism, bioavailability, and excretion of aquatic organisms [113]. This depends on the release of the chemical in the organisms and hence on the strong or weak adsorption ability of MPs and may result in unpredictable ecological consequences [114].

Many studies have been published so far revealing a transfer of PHs from MPs to marine organisms. The adsorbed PHs may be desorbed much faster in the gut of fish than in seawater [53], constituting an important vector of contamination.

Zhang et al. [16] reported that the antibiotic roxithromycin was bioaccumulated in Red tilapia in a greater proportion when it was adsorbed onto PS than when it was directly ingested, while it could mitigate the oxidative damage and neurotoxicity caused by ROX. The co-exposure via ingestion of MPs with venlafaxine and O-desmethylvenlafaxine increased their levels in fish tenfold [115]. Zhang et al. [16] and Zhou et al. [113] reported that MPs may facilitate the bioaccumulation of PHs in fish and bivalves [113]. PS–MPs can enhance the bioaccumulation of roxithromycin in the fish *Oreochromis niloticus* with a similar pattern of the antibiotic alone: gut > liver > brain > gills [16] with bioaccumulation increasing with the increase of PS–MP concentration. Similarly, the bioaccumulation of the antidepressant venlafaxine and its metabolite O-desmethylvenlafaxine in *Misgurnus anguillicaudatus* liver increased with co-exposure to PVC–MPs [115]. The results reveal the capability of MPs to retain drugs and release them to biota after ingestion, promoting their bioaccumulation. However, this is not always the rule, and MPs can decrease ibuprofen bioaccumulation in the microalgae *Chlorella pyrenoidosa* exposed to PS–NPs. This might be due to the strong retention of ibuprofen PS–NPs or to the sustained degradation of ibuprofen in *C. pyrenoidosa* via enhancement of the enzymatic activity of the microalgae [62].

After ingestion of polluted MPs, PHs may be desorbed under gastrointestinal conditions with mechanisms still poorly known. Gut environmental conditions play an important role. MP releasing capacity increases if the pH is low [115], the temperature is high, and the DOM is low [53], and thus, the rate of biotic transfer is higher in intestinal and gut fluid than that in ambient waters [116]. Other authors considered the effect of weathering and ageing and reported opposite results, i.e., a reduction of PH desorption, decreasing bioaccessibility in digestive fluids due to stronger interaction between PHs and aged MPs [79]. Liu et al. [117] reported that ageing inhibited PH desorption by decreasing hydrophobicity and π–π interactions and increasing electrostatic interactions between the weathered polymer and drugs.

Liu et al. [79] assayed desorption of atorvastatin, log Kow 6.4, and amlodipine, log Kow 2.1, in simulated gastric fluid, pH 2, intestinal fluid, pH 7, and seawater, pH 7, and observed the ageing effect. Desorption was greater for atorvastatin in intestinal fluid and amlodipine in gastric fluid, and ageing hindered the process due to the reduction of hydrophobicity given by the oxygen functional groups, whereas high temperature increased desorption. Similarly, the uptake of PVC–triclosan particles by lugworm *Arenicola marina* increased the mortality rate by over 50% of the population [118]. Thus, the bioaccessibility of PHs depends on their solubility, competition of gastrointestinal components, such as pepsin and bile salts, for MP adsorption sites, and physiological conditions, such as pH and temperature, of the organism [106]. Liu et al. [119] focused on the desorption mechanisms in the stomach and intestine of marine organisms and revealed the role of pepsin on PH solubilization and its competition for adsorption sites on MPs via π–π and hydrophobic interactions. The temperature of the organism also plays an important role [79]. Wagstaff and Petrie [120] assayed desorption of the antidepressant fluoxetine from PET MP marine water and simulated gastric and intestinal fluids and highlighted a rate of desorption of 37% at 20 °C and 41% at 37 °C at up to 4 h of incubation in gastric fluid and intestinal fluids.

Even biofouling influencing sorption, desorption, and degradation of co-occurring drugs affects their potential toxicity toward aquatic organisms. MPs biofouled carrying PHs can induce a shift in the bacterial community selecting pollutant-degrading bacteria [31]. Biofouling can increase bioavailability through increased ingestion of pollutant-loaded particles and subsequent desorption in the digestive system or by pollutant transfer to the aqueous phase or food sources, potentially inducing increased exposure for certain organisms [116] or ingestion by filtering organisms due to emission of chemical attracting signals [99].

9. Toxicity

As already seen for bioavailability, even toxicity is still controversial. The interaction processes and toxicological outcomes of MPs–PHs mixtures are complex and variable with potential biotoxicological outcomes.

Studies are comparing the interaction of MPs and PHs in a single test and combining exposure scenarios and toxic effects. The influence of MPs on PH toxicity for aquatic species will depend on the mixing PHs–MPs as well as on the different sensitivities of the affected organisms. Three different scenarios are possible: (1) MPs decrease the toxicity of PHs; an example is given by the inhibitory effect of ibuprofen on the growth rate of the freshwater microalgae *C. pyrenoidosa*, which was alleviated in the presence of PS–NPs [13]; (2) MPs can potentiate the toxicity as reported by Guilhermino et al. [111]. This is the case of the inhibition of the feeding activity of the bivalve *C. fluminea* by the antibiotic florfenicol, which was more pronounced in the presence of MPs; or (3) microplastics might not have an impact on the toxicity. The exposure of brown trout (*Salmo trutta f. fario*) to the antidepressant amitriptyline caused a significant effect on the fish development (fish were smaller and weightless), affected fish swimming behaviour, elevated acetylcholinesterase activity, and inhibited the activity of carboxylesterases, though any of these adverse effects were changed by the presence of PS–MPs [121].

MPs carrying PHs can be ingested by aquatic organisms from food and drinking water and gill respiration [122]. Then, the mixtures of MPs and drugs move to the digestive system, oesophagus, stomach, and then the intestine where they affect the gut microbial community and especially the smaller particles [123]. Once released, the drugs reach the blood circulation, damaging organs, such as the intestinal barrier, which is the first line of defence against MPs–PHs invasion. They subsequently bioaccumulate in the food chain and harm biological health [124]. Some authors [111] exposed a PS–antibiotic mixture to the bivalve *Corbicula fluminea* and observed a higher toxicity than the individual toxicity regarding neurotoxicity and oxidative damage. Fonte et al. [99] observed that co-exposure to MPs and cefalexin inhibited the post-exposure predatory performance of common goby. Other studies showed opposite outcomes as those of Oliveira et al. [125] with the plastic polymer promoting biotransformation or bioavailability and hence delaying organism mortality.

The interactions may be synergistic, additive, or antagonistic [113]. Zhang et al. [98] revealed that in the combined presence of PS and roxithromycin, roxithromycin-induced neurotoxicity was attenuated, and there was a significant increase in superoxide dismutase activity, meaning that the MPs were antagonistic. In the synergistic and additive effect, MPs exacerbate the biotoxicity of pollutants. Many studies show increased toxicity of MPs–PHs association for aquatic biota than single exposure trials [99,103,126]. Alemida et al. [126] tested MPs in association with the antibiotic cephalexin and the common goby *Pomatoschistus microps* and the anticonvulsant carbamazepine and the Mediterranean mussel *Mytilus galloprovincialis*. Qu et al. [101] studied the association of MPs with methamphetamine, commonly known as a narcotic, which increased the toxic and enantioselectivity of methylamphetamine against aquatic species. Mao et al. [103] carried out individual and combined tests of toxicity with polyethylene and ciprofloxacin on free-floating freshwater macrophytes and reported that sensibility was species-specific and co-pollution influenced macrophyte population dynamics and community structure. The authors hypothesized that co-pollutants of MPs and antibiotics have stronger toxicity on floating macrophytes due to MPs enriching more antibiotics through a series of surface adsorption and desorption processes. Ingestion of MPs co-contaminated with PHs by marine invertebrates and vertebrates leads to physical effects and physiological stress [125]. MPs can cause the blockage of the intestinal tract and secretion of gastric enzymes, reduce the levels of steroid hormones, and delay ovulation, hindering food passage [23].

Toxicity is also affected by biofouling. There is a chance that the degradation routes of sorbed drugs on biofouled polymers may be altered and lead to compounds of higher toxicity. The antibiotic sulfamethoxazole is partially degradable naturally, generating

products of acute toxicity much higher than the initial substance [127]. Zhang et al. [98] exposed fish livers, *Oreochromis niloticus*, for 14 d to antibiotic roxithromycin-associated MPs and observed considerable variation of cytochrome P450 activities to RDX alone, suggesting that the polymer may affect the metabolism of the antibiotic in tilapia. In some cases, MPs can be considered hubs and carriers of microbial pathogens and antimicrobial resistance genes, ARGs, with the help of signalling molecules [128]. These biofilms facilitate the horizontal transfer of genes and further exacerbate AMR [129]. The association may also cause the production and spread of antibiotic resistance genes, ARGs, by attaching cohesive microorganisms on surfaces and hence inducing horizontal gene transfer among species [130]. The effects of the antibiotic–polymer-attached biofilms on gene transfer across strains are poorly known [131].

10. Conclusions

Interaction of polymers with drugs is significant and very complex with electrostatic and hydrogen bonding as dominant mechanisms, due to polar and ionizable nature of PHs and hence pronounced hydrophilic feature. The electrostatic interaction in an aqueous system is driven mainly by pH, range 5–9. In this range, MPs surface charge varies among positive, negative, or neutral, and polymers tend to aggregate. However, in the case of estuaries and coastal waters, the common pH is ~8.1, and there would be a tendency for many PHs to desorb from MPs due to repulsion. Regarding toxicity, many studies report on the Trojan horse effect of MPs on drugs and hence consider MPs as a hidden source of toxicants that can be unintentionally taken up by biota. However, the effects on the bioavailability and toxicity of aquatic organisms remain unpredictable as well as their ecological consequences.

There is a lack of research on the co-existence of different groups of PHs and MPs in simulated or real marine scenarios. Most of the research has been simulated at bench scale, underestimating the role of ageing and with antibiotics sorption to polar polymers, and data are needed on β-blockers, antidepressants, NSAIDs, analgesics, steroidal hormones, and antimicrobials. Steroidal hormones are one of the drug classes whose MPs sorption has not yet been thoroughly investigated. Future studies should include how biofilms interact with polymers and drive the accumulation, degradation, and ecotoxicological behaviour of drugs.

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