Sorption of Pharmaceuticals on Microplastics

Lúcia H. M. L. M. Santos, Sara Rodríguez-Mozaz, and Damià Barceló

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Abstract

The presence of microplastics (MPs) in the environment has gained increasing attention in recent years. They are emerging environmental contaminants distributed worldwide that may have a negative impact on ecosystems, organisms, and even in human health. However, MPs are not alone in the environment and coexist with many other organic and inorganic contaminants, such as pharmaceuticals. Due to their physical-chemical properties, MPs have the ability to sorb pharmaceuticals from the surrounding water column in their surface, acting as vectors or carriers in the aquatic environment. MPs properties, such as type of

L. H. M. L. M. Santos · S. Rodríguez-Mozaz (✉)
Catalan Institute for Water Research (ICRA), Girona, Spain

Universitat de Girona, Girona, Spain
e-mail: lhsantos@icra.cat; srodriguez@icra.cat

D. Barceló
Catalan Institute for Water Research (ICRA), Girona, Spain

Universitat de Girona, Girona, Spain

Department of Environmental Chemistry, IDAEA-CSIC, Barcelona, Spain
e-mail: dbcqam@cid.csic.es

T. Rocha-Santos et al. (eds.), *Handbook of Microplastics in the Environment*,
https://doi.org/10.1007/978-3-030-10618-8_14-1

polymer, particle size, surface area, polarity, and pharmaceuticals characteristics (e.g., log K_{ow}, p*Ka*), can directly affect their sorption behavior. Moreover, MPs may undergo different aging/weathering processes in the environment, which contribute to their degradation by decreasing MPs size and changing the particle surface topography and chemistry. These processes will induce changes in the physical-chemical properties of MPs, affecting their sorption behavior. In general, it is expected that aged MPs have a higher sorption capacity for pharmaceuticals than pristine ones. This chapter focuses on the existing studies on the sorption of pharmaceuticals on MPs. A review of the current knowledge on the sorption/desorption mechanisms involved in the sorption behavior of pharmaceuticals on MPs and how this is influenced by environmental factors (e.g., aging/weathering processes, pH, salinity, dissolved organic matter) is provided.

Introduction

In 2004, Thompson and coworkers introduced the term "microplastic" to define microscopic plastic debris with a large-scale potential to accumulate in the environment and be ingested by aquatic organisms (Thompson et al. 2004). Since then, the presence of microplastics (MPs) in the environment has gained an increasing attention, becoming a worldwide concern not only from the scientific community but also from the population in general.

Nowadays, there is still some controversy relative to the definition of MPs (Hartmann et al. 2019), but generally they are defined as plastic fragments with a particle size between 100 nm and 5 mm (Fig. 1). Plastic particles with less than 100 nm are called nanoplastics, while fragments with more than 5 mm are usually defined as macroplastics.

MPs can be categorized in many groups depending on characteristics such as their chemical composition (for example, polyethylene (PE), polyethylene terephthalate (PET), polyvinylchloride (PVC), polypropylene (PP), polystyrene (PS), polyamide (PA), etc.); their form (usually fibers, fragments, spheres, films, and foams); and their color, among others (Hartmann et al. 2019; Picó and Barceló 2019). MPs can also be divided in two categories relatively to their origin: primary MPs, which are those specifically manufactured with a microsize that can be found, for instance, in cosmetic products, such as facial cleaners, or can be used in medicine as pharmaceutical vectors; and secondary MPs, which are formed by degradation of larger plastic materials to small particles, due to physical, chemical, and biological processes (Wang et al. 2018; Picó and Barceló 2019). Secondary MPs are the predominant ones in the environment.

Fig. 1 Size-based definition of plastic fragments

Nowadays, MPs have been ubiquitously reported in the environment all over the world, as has been reviewed by different authors (Avio et al. 2017; Wu et al. 2019; Li et al. 2020; Llorca et al. 2020; Wong et al. 2020; Xu et al. 2020), representing a growing concern, since the worldwide production and consumption of plastics have been drastically increasing over the years. Furthermore, plastics are very stable and have a very long and slow process of degradation, persisting long times in the environment after being discarded, thus constituting a threat for the welfare of animals, marine food web, and human health.

MPs can be introduced into the environment through their direct discharge in the effluents of wastewater treatment plants (WWTPs), runoff, and due to weathering breakdown of larger plastic debris (León et al. 2018). Once in the environment, MPs may have different behaviors, such as migration, sedimentation, or accumulation, which will depend on the action of external factors. Migration consists in the transport of MPs to other sites due to the action of currents, wind, tide, tsunamis in the oceans, etc. Sedimentation involves the biofouling of floating MPs in the aquatic environment, which will increase their density, resulting in the sink of the plastic particles to the river or sea bed. Sedimentation of MPs may be facilitated by the microbial colonization of their surfaces, the adherence of phytoplankton, or the aggregation with organic debris and metals (Wang et al. 2016). Usually, MPs tend to accumulate in water surface, especially those which density is lower than water (e.g., PP (0.9–0.91 g/cm^3) and PE (0.917–0.965 g/cm^3)), while MPs with a higher density, such as PS (1.04–1.11 g/cm^3), polycarbonate (PC) (1.18–1.22 g/cm3), or PVC (1.38 g/cm^3), tend to sink and are mostly found in sediments. However, MPs environmental distribution is not so linear and can be affected by both MPs density and external factors such as those previously mentioned (e.g., temperature, wind, storms, internal waves, and the surface-to-volume ratio). Thus, high-density MPs can also be found in the water column or the low-density ones in sediments (Wang et al. 2016; Di and Wang 2018).

However, MPs are not alone in the environment, coexisting with other organic and inorganic contaminants. Due to their physical-chemical properties, MPs have the ability to sorb and accumulate contaminants from the surrounding water column in their surface, acting as vectors of contaminants to organisms after ingestion. Indeed, contaminants concentration in MPs may be up to 10^5–10^6 times higher than in the surrounding water column (Wang et al. 2018) and will reflect the environmental global pollution patterns. A great concern associated with MPs is due to the desorption of sorbed contaminants and the leaching of plastic chemical additives that are introduced during the manufacture process (e.g., phthalates, plasticizers, flame retardants, UV stabilizers, pigments, etc.), into an organism after ingestion (Wang et al. 2016; Carbery et al. 2018; Wang et al. 2018). All in all, MPs as well as the desorbed contaminants and leached additives may be bioaccumulated in organisms, easily entering the food web and reaching high trophic levels and ultimately human beings (Carbery et al. 2018; Peng et al. 2020). MPs can also act as carriers of contaminants and microorganisms, facilitating their fast and far transport in the environment (Wang et al. 2016; Carbery et al. 2018). In this way, sorption and

desorption of contaminants on MPs may play an important role in the fate and distribution of pollutants in the aquatic environment.

MPs have showed the ability to sorb different kind of organic contaminants, such as pesticides (León et al. 2018; Camacho et al. 2019; León et al. 2019), persistent organic pollutants (e.g., polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs), per- and polyfluoroalkyl compounds (PFAS)) (Hüffer et al. 2018; León et al. 2018; Llorca et al. 2018; Camacho et al. 2019), personal care products (Wu et al. 2016; León et al. 2018; Camacho et al. 2019; León et al. 2019), and pharmaceuticals (Wu et al. 2016; Guo and Wang 2019; Elizalde-Velázquez et al. 2020), among others. This book chapter will focus on the sorption of pharmaceuticals on MPs, their sorption/desorption mechanisms involved as well as the effect of aging or weathering of MPs in these processes. The influence of natural factors such as pH, salinity, or dissolved organic matter (DOM) in the sorption processes will also be discussed.

Sorption/Desorption Processes Between Microplastics and Pharmaceuticals

Adsorption of contaminants such as pharmaceuticals on MPs depends on different factors linked either to the sorbent (MPs) or to the sorbate (contaminants) properties. Type of polymer, particle size, polarity, specific surface area, pore size distribution, abundance of rubbery, or degree of crystallinity, are examples of MPs properties that can have impact on the sorption capacity of pollutants. Besides that, the properties of organic contaminants (sorbates) and their greater affinity for the MPs surface than to the water column will also influence the sorption of organic contaminants onto MPs. Exposure time and the concentration of the contaminants in the surrounding water column are other factors that have to be taken into account too (Wang et al. 2016; León et al. 2018; Li et al. 2018; Wang et al. 2018).

Microplastics Properties

In general, PE showed a greater sorption capacity to environmental pollutants than other type of polymers, followed by PP and PS (Wang et al. 2018). This could be related with the abundance of rubbery segments, since, in general, rubber MPs, such as PE, allow a great diffusion of contaminants into the polymer, showing a higher adsorption capacity than the glassier ones (e.g., PP, PS, and PVC) (Alimi et al. 2018; Li et al. 2018). This is in agreement with the findings by Elizalde-Velázquez et al. (2020) that observed a higher sorption affinity of three nonsteroidal anti-inflammatory drugs (NSAIDs) (ibuprofen, diclofenac, and naproxen) to PE rather than PS and PP (Table 1). Nevertheless, PS exhibited a higher sorption affinity to the antibiotic tetracycline than PP or PE (Xu et al. 2018a). This could be justified by the increased polarity of PS. Although the three MPs (PE, PP, and PS) are considered nonpolar polymers, the presence of benzene on PS possibly increases its polarity, promoting

Table 1 Adsorption kinetics data of pharmaceuticals on pristine microplastics

Pharmaceutical	Microplastic	MP particle size	Adsorption capacity (*qe*)	Equilibrium time	Matrix	Experimental conditions	Reference
Antibiotics							
Fluoroquinolones							
Ciprofloxacin	PS	~75 μm	2.580 mg/g	24 h[a]	Ultrapure water	0.4 g/L MPs, 10 mg/L CIP, room temperature	Liu et al. (2019a)
	PVC	~75 μm	3.410 mg/g				
Ciprofloxacin	PS	50.4 ± 11.9 μm	147.72 mg/kg	–[b]	Ultrapure water	0.15 g MPs, 0.15 mg CIP, 25 °C	Liu et al. (2019b)
	HDPE	45.5 ± 12.9 μm	125.32 mg/kg				
Macrolides							
Tylosin	PE	<74 μm	666.67 mg/kg	36 h	Ultrapure water containing 0.001 M NaN_3	0.005–0.03 g MP, 0–30 mg/L TYL, 25 °C	Guo et al. (2018)
	PP		833.33 mg/kg				
	PS		1428.57 mg/kg				
	PVC		1666.67 mg/kg				
Sulfonamides							
Sulfamethoxazole	PE	150 μm	108.1 ± 2.16 mg/kg	24 h	Synthetic freshwater (ultrapure water containing 0.01 M $CaCl_2$ and 200 mg/L NaN_3)	5 g/L MPs, 1 mg/L SMX, 25 °C	Xu et al. (2018b)
Sulfamethoxazole	UHMWPE	45–48 μm	46.09 mg/kg	96 h	Ultrapure water containing 0.01M $CaCl_2$ and 0.02% (w/v) NaN_3	50, 200 or 500 mg/L MPs, 60 μg/L SMX, 24 °C	Razanajatovo et al. (2018)

(continued)

Table 1 (continued)

Pharmaceutical	Microplastic	MP particle size	Adsorption capacity (*qe*)	Equilibrium time	Matrix	Experimental conditions	Reference
Sulfamethoxazole	PA	100–150 μm	0.40 mg/g	16 h	Ultrapure water	2 g/L MPs, 2.4 mg/L SMX, 25 °C	Guo et al. (2019a)
	PE		0.10 mg/g				
	PET		0.06 mg/g				
	PS		0.07 mg/g				
	PVC		0.09 mg/g				
	PP		0.12 mg/g				
Sulfamethazine	PA	100–150 μm	0.0821 mg/g	16 h	Ultrapure water	2 g/L MPs, 2.0 mg/L SMT, 25 °C	Guo et al. (2019b)
	PE		0.0546 mg/g				
	PET		0.0530 mg/g				
	PP		0.074 mg/g				
	PS		0.0637 mg/g				
	PVC		0.0657 mg/g				
Tetracyclines							
Tetracycline	PE	60–150 μm	237.5 μg/g	–[b]	Ultrapure water containing 0.01 M $NaNO_3$, 0.003 M NaN_3	5 g/L MPs, 5 mg/L TC, 25 °C	Wang et al. (2020b)
Tetracycline	PE	150 μm	109 ± 3.62 mg/kg	24 h	Synthetic freshwater (ultrapure water containing 0.01 M $CaCl_2$ and 200 mg/L NaN_3)	5 g/L MPs, 1 mg/L TC, 25 °C	Xu et al. (2018a)
	PP	<280 μm	113 ± 4.45 mg/kg				
	PS	<280 μm	167 ± 7.74 mg/kg				

Tetracycline	PE	150–250 μm	120.48 mg/kg	10 days	Ultrapure water	10 g/L MPs, 10 mg/L TC, 20 °C, pH 7	Shen et al. (2018)
Oxytetracycline	PS (foam)	0.45–1 mm	–[a]	54 h	Ultrapure water containing 0.01 M NaCl and 25 mg/L NaN_3	1.7 g/L MPs, 20 mg/L OTC, 25 °C	
Psychiatric drugs							
Carbamazepine	LDPE	250–280 μm	32 mg/kg	48 h	Synthetic freshwater (ultrapure water containing 0.01 M $CaCl_2$ and 0.01% (*w/v*) NaN_3)	0.2 or 1.0 g/L MPs, 1 mg/L CBZ	Wu et al. (2016)
Carbamazepine	PE	150–250 μm	78.9 mg/kg	10 days	Ultrapure water	10 g/L MPs, 20 °C, pH 7	Shen et al. (2018)
Sertraline	UHMWPE	45–48 μm	88.80 mg/kg	96 h	Ultrapure water containing 0.01 M $CaCl_2$ and 0.02% (w/v) NaN_3	50, 200 or 500 mg/L MPs, 60 μg/L SRT, 24 °C	Razanajatovo et al. (2018)
Cardiovascular drugs							
Propranolol	UHMWPE	45–48 μm	64.38 mg/kg	96 h	Ultrapure water containing 0.01 M $CaCl_2$ and 0.02% (w/v) NaN_3	50, 200 or 500 mg/L MPs, 60 μg/L PRP, 24 °C	Razanajatovo et al. (2018)
Amlodipine	PS	50.4 ± 11.9 μm	0.22 mg/g	48 h	Ultrapure water containing 10 mM NaCl	5 g/L MPs, 10 mg/L AML, 25 °C, pH 6.5	Liu et al. (2020a)
	PE	~45 μm	0.10 mg/g[a]				
	PP	~50 μm	0.15 mg/g[a]				

(continued)

Table 1 (continued)

Pharmaceutical	Microplastic	MP particle size	Adsorption capacity (*qe*)	Equilibrium time	Matrix	Experimental conditions	Reference
Nonsteroidal anti-inflammatory drugs (NSAIDs)							
Ibuprofen	UHMWPE	2–10 μm	0.053 mg/kg	48 h	Ultrapure water	10 g/L MPs, 2.5 mg/L of each NSAID, 25 °C, pH 6.9	Elizalde-Velázquez et al. (2020)
	AMWPE	300–400 μm	0.052 mg/kg				
	PS	600–800 μm	0.040 mg/kg				
	PP	~1 mm	0.030 mg/kg				
Diclofenac	UHMWPE	2–10 μm	0.067 mg/kg				
	AMWPE	300–400 μm	0.063 mg/kg				
	PS	600–800 μm	0.065 mg/kg				
	PP	~1 mm	0.039 mg/kg				
Naproxen	UHMWPE	2–10 μm	0.035 mg/kg				
	AMWPE	300–400 μm	0.033 mg/kg				
	PS	600–800 μm	0.026 mg/kg				
	PP	~1 mm	0.017 mg/kg				
Lipid regulators							
Atorvastatin	PS	50.4 ± 11.9 μm	1.24 mg/g	48 h	Ultrapure water containing 10 mM NaCl	5 g/L MPs, 10 mg/L ATV, 25 °C, pH 6.5	Liu et al. (2020a)
	PE	~45 μm	0.45 mg/g[a]				
	PP	~50 μm	0.35 mg/g[a]				
Sex hormones							
17α-Ethinylestradiol	LDPE	250–280 μm	50 mg/kg	48 h	Synthetic freshwater (ultrapure water containing 0.01M $CaCl_2$ and 0.01% (*w/v*) NaN_3)	0.2 or 1.0 g/L MPs, 1 mg/L EE2	Wu et al. (2016)

PS polystyrene, *PVC* polyvinylchloride, *HDPE* high density polyethylene, *PE* polyethylene, *PP* polypropylene; *UHMWPE* ultrahigh molecular weight polyethylene, *PA* polyamide, *PET* polyethylene terephthalate, *LDPE* low-density polyethylene, *AMWPE* average molecular weight medium-density polyethylene

[a]Extrapolated from kinetics graphics

[b]Not indicated

its sorption capacity through polar interactions and π-π interactions (Xu et al. 2018a). The presence of a benzene ring in the PS monomer also increases the distance between the polymer chains, facilitating the attachment and integration of pharmaceuticals into this polymer (Alimi et al. 2018). The type of polymer will also influence in the polarity of MPs, due to the presence of functional groups in their structure. Although MPs polarity can affect the adsorption of polar chemicals, just by itself it is not capable of explaining differences in the adsorption capacity as was proved by Li et al. (2018). These authors observed that the polar PA MPs had a high-sorption capacity for four antibiotics (ciprofloxacin, trimethoprim, amoxicillin, and tetracycline), while the polar PVC MPs showed a lower affinity for the same antibiotics. MPs may establish different interactions with pharmaceuticals: Some of the most common binding mechanisms are partitioning, hydrophobic interaction, van der Walls forces, π-π interactions, hydrogen bonding, electrostatic interaction, and cation competition (Elizalde-Velázquez et al. 2020). For instance, PS can undergo nonspecific van der Waals and π-π interactions with pharmaceuticals at the aromatic surface, while PE and PP can only undergo van der Waals interactions, since they do not have ionizable functional groups (Li et al. 2018; Qu et al. 2018; Shen et al. 2018; Xu et al. 2018a). PA, PS, PVC, and PP are also polymers more polar than PE and PET, because they have polar groups (e.g., -CO-NH- in PA, benzene ring in PS, -Cl in PVC, and -CH_3 in PP), which enhance the electrostatic interaction between MPs and polar compounds such as pharmaceuticals through dipole-dipole and dipole-induced dipole (Guo et al. 2019b). Guo et al. (2019a) compared the adsorption capacity of the antibiotic sulfamethoxazole on six MPs (PA, PE, PET, PS, PP, and PVC), showing that sulfamethoxazole had a higher affinity for PA than for the other MPs, probably due to the formation of hydrogen bonding between the antibiotic and the group amide of PA. Sulfamethazine also showed a greater affinity for PA MPs, but in this case, the sorption processes were dominated by both electrostatic and van der Walls interactions (Guo et al. 2019b). PA MPs also proved to have a high sorption capacity for the antibiotics ciprofloxacin, amoxicillin, and tetracycline, which can be attributed to the formation of hydrogen bonding between the amide group of PA and the carbonyl group present in these antibiotics (Li et al. 2018). The antibiotic tylosin also showed a high adsorption capacity for polar MPs, showing the following order: PVC > PS > PP > PE (Guo et al. 2018). Tetracycline showed a greater adsorption capacity on PS MPs rather than on PE and PP, due to the presence of benzene ring in both tetracycline and PS, which favored polar interactions and π-π interactions (Xu et al. 2018a).

The specific surface area of MPs influences in the adsorption of pharmaceuticals by providing adsorption sites. Smaller particles have a larger surface area, increasing significantly their sorption capacity for MPs with the same chemical composition as was shown by Shen et al. (2018). These authors proved that PE MPs with smaller size have a higher adsorption capacity for the antibiotic tetracycline than those with a larger size. Elizalde-Velázquez et al. (2020) also reported an increase in the amount of pharmaceuticals (NSAIDs) sorbed to MPs with a decrease in the particle size.

A high degree of crystallinity represents less amorphous regions on MPs, comprising closely packed polymer chains. Usually, this fact limits the adsorption

capacity of MPs, resulting in a lower adsorption capacity (Liu et al. 2019a; Elizalde-Velázquez et al. 2020). In general, the low crystallinity of MPs favors the adsorption of more hydrophobic organic contaminants (Li et al. 2018).

Pharmaceuticals Properties

On the other hand, adsorption of pharmaceuticals to MPs may be related with the hydrophobicity of the compounds as shown by Razanajatovo et al. (2018). These authors proved that the adsorption of sulfamethoxazole, propranolol, and sertraline on PE was positively related with the octanol-water partition coefficient (log K_{ow}) of the three pharmaceuticals. The same behavior was seen in the adsorption of antibiotics (Li et al. 2018; Zhang et al. 2018) and NSAIDs (Elizalde-Velázquez et al. 2020) on MPs of different polymers. In fact, it is expected that pharmaceuticals with high log K_{ow} had stronger sorption affinities to MPs due to their high hydrophobicity (Wu et al. 2016; Xu et al. 2018b). Elizalde-Velázquez et al. (2020) compared the sorption capacity of diclofenac, ibuprofen, and naproxen on different MPs (PE, PS, and PP), showing that diclofenac, which had the highest log K_{ow} (4.51), exhibited the strongest sorption capacity in all the tested polymers (Table 1). On the other hand, Wu et al. (2016) showed a less pronounced sorption of polar and hydrophilic pharmaceuticals (carbamazepine and 17α-ethinylestradiol) (log $K_{ow} = 2.45$ and 3.67, respectively) on PE, due to a weak interaction with the hydrophobic surface of these MPs. The same behavior was observed for the antibiotic sulfamethazine (log $K_{ow} < 2$), since this hydrophilic antibiotic would have limited interaction with the hydrophobic surface of MPs (Guo et al. 2019b).

The sorption processes of pharmaceuticals on MPs may also be affected by other factors such as electrostatic interactions, which are closely related with the p*Ka* of pharmaceutical, pH of the solution, and the pH point of zero charge (pH_{pzc}) of the MPs (Razanajatovo et al. 2018). p*Ka* of pharmaceuticals together with the pH of the medium will directly influence in the speciation of the compounds and hence in their sorption on MPs (Li et al. 2018). For instance, in a freshwater system (pH = 6.7–7.1), the sorption of pharmaceuticals such as the antibiotic ciprofloxacin, the antidepressant sertraline, or the β-blocker propranolol will be enhanced, because there is electrostatic interaction between the positively charged pharmaceutical and the negatively charged surface of MPs (Li et al. 2018; Razanajatovo et al. 2018). Whereas the adsorption of NSAIDs or the antibiotic sulfamethoxazole in MPs will be decreased at the same pH, given that both pharmaceuticals and MPs will be negatively charged, exerting electrostatic repulsion (Razanajatovo et al. 2018; Elizalde-Velázquez et al. 2020).

Overall, the sorption process of pharmaceuticals on MPs will be affected by MPs type and size, and by contaminant type, which will also influence the sorption equilibrium time. Table 1 summarizes the sorption kinetics data of pharmaceuticals on MPs, giving an overview of the equilibrium time and adsorption capacity of MPs for pharmaceuticals of different therapeutic groups. Most of the data available on literature refers to the adsorption of antibiotics on several MPs. Moreover, due to the

variability of the experimental conditions used between sorption experiments, comparability of the results is, in general, difficult.

Sorption Kinetics and Isotherms

The sorption kinetics may include three mass transfer steps: (1) external mass transfer, which corresponds to the diffusion of pharmaceuticals in the liquid layer around MPs; (2) internal mass transfer that implies the diffusion of pharmaceuticals inside MPs; and (3) adsorption of pharmaceuticals on active sites located on the surface of MPs (Guo et al. 2019a). These steps will significantly influence the equilibrium time of pharmaceuticals on MPs that can be reached within 16 h (e.g., sulfamethoxazole and sulfamethazine on different MPs (PA, PE, PET, PS, PP, and PVC)) to 10 days (e.g., tetracycline on PE) (Table 1). The sorption kinetics of pharmaceuticals on MPs may include two or three steps, suggesting different adsorption stages during the sorption process. For instance, the adsorption of sulfamethoxazole on six MPs (PA, PE, PET, PS, PP, and PVC) drastically increased in the first hour, and then the sorption rate slowed down until it reached the equilibrium within 16 h. The diffusion of sulfamethoxazole in the liquid film around MPs was identified as the rate-limiting step, which could be attributed to the hydrophobicity of MPs surface. The diffusion of sulfamethoxazole in the pores inside the MPs also occurred (Guo et al. 2019a). The adsorption kinetic of sulfamethazine on the same MPs showed a similar behavior, being controlled by external and internal diffusion (Guo et al. 2019b). Different sorption stages were also identified in the sorption of tetracycline in PE MPs, corresponding initially to film diffusion followed by intraparticle diffusion, which played an important role on the adsorption of this antibiotic on PE MPs (Wang et al. 2020b). The sorption of tylosin to several MPs (PE, PP, PS, and PVC) could be described in three stages, including a quick adsorption onto the surface of MPs, which could involve hydrophobic interaction, covalent forces, and van der Walls forces, followed by a later diffusion into the pores located in the interlayer structure of MPs, which was identified as the rate-limiting step (Guo et al. 2018). Moreover, the mass transfer steps of a sorption process can be changed due to environmental conditions (Guo et al. 2019a).

Sorption isotherms are frequently used to model the sorption of pharmaceuticals on MPs and can fit different models, namely, linear, Freundlich, and Langmuir, depending on the interactions established between sorbent and sorbate. The linear isotherms are defined by a linear correlation between the concentration of pharmaceutical sorbed on the MPs and the concentration of the pharmaceutical in the aqueous phase at equilibrium. Freundlich isotherms define a nonlinear relationship between the amount of pharmaceutical sorbed and the concentration of pharmaceutical in the solution at the equilibrium, while Langmuir isotherms describe the sorption process by a rational basis of a monolayer coverage (Elizalde-Velázquez et al. 2020). In the nonlinear adsorption isotherms (Freundlich and Langmuir), not only hydrophobic interactions play an important role in the sorption process, but also other interactions such as electrostatic interactions have to be considered (Xu et al.

2018a). Table 2 summarizes the sorption equilibrium data of pharmaceuticals on MPs. In general, for most of the compounds, the linear and Freundlich models are those that better represent the sorption equilibrium data of pharmaceuticals on MPs. This suggests that sorption processes are mainly dominated by the partitioning of pharmaceuticals between MPs and the aqueous phase (linear model), or by heterogeneous multilayer sorption processes (Freundlich model). Nevertheless, it is difficult to compare between experiments, since there is a huge variability in the experimental conditions considered.

Besides the MPs characteristics, including type of polymer and particle size, and the contaminant type, external factors such as weathering/aging of MPs, presence of dissolved organic matter (DOM), pH, and salinity may also affect the sorption behavior between MPs and pharmaceuticals. These factors will be discussed in the following sections.

Effect of Aging or Weathering of Microplastics in the Sorption/Desorption Processes

Different aging/weathering processes as well as the foulants attached to the plastic debris will influence in the sorption capacity of MPs. Although the aging of MPs increases their ability to sorb contaminants (Wang et al. 2016; Wang et al. 2018), just a few studies focused on the impact of these processes in the sorption of organic contaminants on MPs.

The processes of aging and weathering contribute to the degradation of MPs and can be driven by abiotic and biotic factors. Examples of abiotic factors are photodegradation by UV light, seawater corrosion, physical abrasion, thermal degradation (rarely occurs in the environment), and chemical oxidation, while biotic factors concern the microbial colonization of MPs surface and the biodegradation by living organisms. All these factors induce changes in the particle size and in the particle surface topography and chemistry, which can be reflected by: (i) increasing the surface area, due to cracking and fragmentation of the particles, as well as the number of sites available for sorption of contaminants and microbial colonization; (ii) changing the polarity and surface properties due to an increase in oxygen functional groups (e.g., carboxylic, aldehyde, ketone, and hydroxyl groups); (iii) affecting the crystallinity; or (iv) formation of biofilms on the MPs surface (Wang et al. 2016; Carbery et al. 2018; Liu et al. 2019a; Elizalde-Velázquez et al. 2020).

One of the most evident effects of aging/weathering processes is the fragmentation of MPs into smaller-sized particles as well as the generation of cracks on their surface. This will increment the particle surface area, which might have a higher sorption capacity to contaminants. In fact, it was proved that sorption properties of aged MPs are positively correlated with their surface area, which is an important factor concerning the adsorption of hydrophilic contaminants such as pharmaceuticals (Liu et al. 2020b).

Polarity of MPs surface can be increased by the reaction with oxygen, which contributes for the occurrence of hydroxyl bonds, carbon-oxygen bonds, and

carbonyl bonds on the surface of weathered MPs, resulting in an increased hydrophilicity (Wang et al. 2016; Xu et al. 2018b; Liu et al. 2019a). The presence of carbonyl and hydroxide radicals in the surface of aged PS and PVC MPs was seen as a result of their UV-accelerated oxidation (Liu et al. 2019a). It is expected that longer aging times will be reflected in a higher amount of oxygen functional groups formed on the surface of MPs (Liu et al. 2020a). As a consequence, other interactions such as electrostatic interactions and chemical bonds interaction (e.g., formation of H-bonding, ion exchange, and complexation) may be involved in the sorption/desorption processes, affecting the sorption/desorption behavior of MPs (Xu et al. 2018b; Liu et al. 2020b). For instance, Liu et al. (2020a) showed that the adsorption of pharmaceuticals in pristine PS MPs relied on hydrophobic and π-π interactions, while in aged PS MPs, the adsorption was controlled by electrostatic interactions and hydrogen bonding. This could be justified by a decrease in the content of benzene rings in the PS backbone due to chain scission provoked by weathering processes (Liu et al. 2020b). An increase in the hydrophilicity of aged PS MPs was also reported (Liu et al. 2020a). It was shown that the adsorption of amlodipine on aged PS was mainly attributed to electrostatic interaction, while the adsorption of atorvastatin was dominated by hydrogen bonding (Liu et al. 2020a). In fact, hydrogen bonding is an important adsorption mechanism on aged MPs concerning hydrophilic and polar organic contaminants such as pharmaceuticals (Liu et al. 2020b).

The presence of natural factors such as humic acid (HA) during the aging process or the colonization of MPs surface by microorganisms, such as diatoms, filamentous algae, or development of a biofilm layer, can also interfere with the adsorption capacity of MPs. HA is able to adsorb on the MPs surface, affecting their adsorption capacity either by reducing the number of adsorption sites available for the adsorption of pharmaceuticals or by changing the charge characteristics of the surface of MPs due to the negatively charged groups of HA (Shen et al. 2018). On the other hand, the formation of biofilms on the MPs surface may potentiate the adsorption capacity of MPs by decreasing their hydrophobicity, slowing down the mass transfer of contaminants from the surrounding water column to the MPs, and increasing their surface area and heterogeneity, since biofilms may act as additional sorbents (Wang et al. 2016; Elizalde-Velázquez et al. 2020; Liu et al. 2020b). Wang et al. (2020b) revealed that the adsorption of the antibiotic tetracycline was enhanced on biofilm-developed PE MPs. This could be due to a complexation of tetracycline with components of the biofilm. The authors also showed that the adsorption of tetracycline on biofilm-developed PE MPs was more stable than on virgin PE MPs, which could prevent the desorption of the antibiotic from the aged MPs surface. On the other hand, Shen et al. (2018) showed that the adsorption capacity of the antibiotic tetracycline and the antiepileptic carbamazepine on HA-aged PE MPs was considerably lower than in pristine PE, probably due to changes in the MPs surface.

Overall, aging/weathering processes may completely change the properties of aged MPs relatively to those that were discharged into the environment, influencing their sorption capacity as well as the type of contaminants that may sorb to aged MPs (Li et al. 2018; Elizalde-Velázquez et al. 2020; Liu et al. 2020b). Table 3 summarizes the sorption kinetics data (equilibrium time and adsorption capacity) of

Table 2 Sorption isotherms data of pharmaceuticals on pristine microplastics

Pharmaceutical	Microplastic	MP particle size	Isotherm type	Isotherm constant (L/Kg)	Matrix	Experimental conditions	Reference
Antibiotics							
β-lactam antibiotics							
Amoxicillin	PE	75–180 μm	Linear	$K_d = 8.40 \pm 0.675$	Freshwater	4.0 g/L MPs, 0.5–15 mg/L AMX, 25 °C, pH 6.7–7.1	Li et al. (2018)
	PP		Langmuir	$K_L = 0.376$ L/mg			
	PA		Langmuir	$K_L = 0.0361$ L/mg			
	PVC		Linear	$K_d = 24.7 \pm 1.20$			
Fluoroquinolones							
Ciprofloxacin	PE	75–180 μm	Freundlich	$K_F = 222 \pm 6.59$; $n = 0.393$	Freshwater	4.0 g/L MPs, 0.5–15 mg/L CIP, 25 °C, pH 6.7–7.1	Li et al. (2018)
	PS		Freundlich	$K_F = 205 \pm 17.0$; $n = 0.316$			
	PP		Langmuir	$K_L = 0.844$ L/mg			
	PA		Langmuir	$K_L = 0.0740$ L/mg			
	PVC		Freundlich	$K_F = 184 \pm 6.19$; $n = 0.371$			
Ciprofloxacin	PS	~75 μm	Langmuir	$K_L = 0.959$	Ultrapure water	0.4 g/L MPs, 2–25 mg/L CIP, room temperature	Liu et al. (2019a)
	PVC	~75 μm	Freundlich	$K_F = 0.550$; $n = 1.380$			
Ciprofloxacin	PS	50.4 ± 11.9 μm	Langmuir	$K_L = 0.109$ L/mg	Ultrapure water	0.15 g MPs, 0.15 mg CIP, 25 °C	Liu et al. (2019b)
	HDPE	45.5 ± 12.9 μm	Langmuir	$K_L = 0.089$ L/mg			

Macrolides							
Tylosin	PE	<74 μm	Linear	$K_d = 62.75$	Ultrapure water containing 0.001 M NaN_3	0.005–0.03 g MPs, 5 mg/L TYL, 25 °C	Guo et al. (2018)
	PP		Freundlich	$K_F = 183.46$[b]; $n = 0.788$			
	PS		Linear	$K_d = 134.10$			
	PVC		Linear	$K_d = 155.27$			
Sulfonamides							
Sulfamethoxazole	PA	100–150 μm	Freundlich	$K_F = 205$; $n = 0.847$	Ultrapure water	2 g/L MPs, 0.5–12 mg/L SMX, 25 °C	Guo et al. (2019a)
	PE		Freundlich	$K_F = 61.3$; $n = 1.48$			
	PET		Linear	$K_d = 22.2$			
	PS		Linear	$K_d = 29.7$			
	PVC		Linear	$K_d = 28.2$			
	PP		Linear	$K_d = 30.9$			
Sulfamethoxazole	PE	150 μm	Linear	$K_d = 591.7 \pm 24.1$	Synthetic freshwater (ultrapure water containing 0.01M $CaCl_2$ and 200 mg/L NaN_3)	5 g/L MPs, 0.2-5 mg/L SMX, 25 °C	Xu et al. (2018b)
Sulfamethoxazole	UHMWPE	45–48 μm	Freundlich	$K_F = 3.09 \pm 0.66$; $1/n = 0.54 \pm 0.05$	Ultrapure water containing 0.01M $CaCl_2$ and 0.02% (w/v) NaN_3	50, 200 or 500 mg/L MPs, 1–100 μg/L SMX, 24 °C	Razanajatovo et al. (2018)

(continued)

Table 2 (continued)

Pharmaceutical	Microplastic	MP particle size	Isotherm type	Isotherm constant (L/Kg)	Matrix	Experimental conditions	Reference
Sulfamethazine	PA	100–150 μm	Linear	$K_d = 38.7$	Ultrapure water	2 g/L MPs, 0.5–12 mg/L SMT, 25 °C	Guo et al. (2019b)
	PE		Freundlich	$K_F = 20.9$; $n = 1.20$			
	PET		Linear	$K_d = 22.6$			
	PP		Linear	$K_d = 15.1$			
	PS		Linear	$K_d = 21.0$			
	PVC		Linear	$K_d = 18.6$			
Sulfadiazine	PE	75–180 μm	Linear	$K_d = 6.19 \pm 0.238$	Freshwater	4.0 g/L MPs, 0.5–15 mg/L SDZ, 25 °C, pH 6.7–7.1	Li et al. (2018)
	PS		Linear	$K_d = 7.39 \pm 0.308$			
	PP		Linear	$K_d = 7.85 \pm 0.679$			
	PA		Linear	$K_d = 7.36 \pm 0.257$			
	PVC		Linear	$K_d = 6.61 \pm 0.549$			
Sulfadiazine	PE	75–180 μm	Linear	$K_d = 6.26 \pm 0.630$	Seawater	4.0 g/L MPs, 0.5–15 mg/L SDZ, 25 °C, pH 8.0	Li et al. (2018)
	PS		Linear	$K_d = 6.80 \pm 0.352$			
	PP		Linear	$K_d = 7.13 \pm 0.952$			
	PA		Freundlich	$K_F = 2.53 \pm 0.226$; $n = 1.38$			
	PVC		Freundlich	$K_F = 0.850 \pm 0.308$; $n = 1.73$			

Tetracyclines							
Tetracycline	PE	60–150 μm	Freundlich	$K_F = 15.6$[a]; $n = 0.987$	Ultrapure water containing 0.01 M $NaNO_3$, 0.003 M NaN_3	5 g/L MPs, 0–15 mg/L TC, 25 °C	Wang et al. (2020b)
Tetracycline	PE	150 μm	Langmuir	$K_L = 0.126 \pm 0.020$	Synthetic freshwater (ultrapure water containing 0.01 M $CaCl_2$ and 200 mg/L NaN_3)	5 g/L MPs, 0.2–5 mg/L TC, 20 °C	Xu et al. (2018a)
	PP	<280 μm	Langmuir	$K_L = 0.039 \pm 0.010$			
	PS	<280 μm	Langmuir	$K_L = 0.076 \pm 0.016$			
Tetracycline	PA	75–180 μm	Langmuir	$K_L = 0.189$ L/mg	Freshwater	4.0 g/L MPs, 0.5–15 mg/L TC, 25 °C, pH 6.7–7.1	Li et al. (2018)
Oxytetracycline	PS (foam)	0.45–1 mm	Freundlich	$K_F = 425 \pm 46$[c]; $n = 1.380$	Ultrapure water containing 0.01 M NaCl and 25 mg/L NaN_3	1.7 g/L MPs, 2–50 mg/L OTC, 25 °C	Zhang et al. (2018)

(continued)

Table 2 (continued)

Pharmaceutical	Microplastic	MP particle size	Isotherm type	Isotherm constant (L/Kg)	Matrix	Experimental conditions	Reference
Other antibiotics							
Trimethoprim	PE	75–180 μm	Freundlich	$K_F = 22.0 \pm 2.59$; $n = 0.560$	Freshwater	4.0 g/L MPs, 0.5–15 mg/L TMP, 25 °C, pH 6.7–7.1	Li et al. (2018)
	PS		Freundlich	$K_F = 32.1 \pm 2.48$; $n = 0.507$			
	PP		Freundlich	$K_F = 32.3 \pm 4.01$; $n = 0.450$			
	PA		Freundlich	$K_F = 36.0 \pm 6.15$; $n = 0.696$			
	PVC		Langmuir	$K_L = 0.0259$ L/mg			
Trimethoprim	PE	75–180 μm	Langmuir	$K_L = 0.469$ L/mg	Seawater	4.0 g/L MPs, 0.5–15 mg/L TMP, 25 °C, pH 8.0	Li et al. (2018)
	PS		Linear	$K_d = 7.30 \pm 0.912$			
	PP		Langmuir	$K_L = 0.551$ L/mg			
	PA		Freundlich	$K_F = 10.0 \pm 1.64$; $n = 0.560$			
	PVC		Linear	$K_d = 5.45 \pm 0.492$			

Psychiatric drugs							
Carbamazepine	LDPE	250–280 μm	Linear	$K_d = 191 \pm 6.4$	Synthetic freshwater (ultrapure water containing 0.01 M $CaCl_2$ and 0.01% (*w/v*) NaN_3)	0.2 or 1.0 g/L MPs, 10–200 μg/L CBZ	Wu et al. (2016)
Sertraline	UHMWPE	45–48 μm	Freundlich	$K_F = 4.36 \pm 0.88$; $1/n = 0.89 \pm 0.05$	Ultrapure water containing 0.01 M $CaCl_2$ and 0.02% (w/v) NaN_3	50, 200 or 500 mg/L MPs, 1–100 μg/L SRT, 24 °C	Razanajatovo et al. (2018)
Cardiovascular drugs							
Propranolol	UHMWPE	45–48 μm	Linear	$K_d = 2.30 \pm 2.79$	Ultrapure water containing 0.01 M $CaCl_2$ and 0.02% (w/v) NaN_3	50, 200 or 500 mg/L MPs, 1–100 μg/L PRP, 24 °C	Razanajatovo et al. (2018)
Amlodipine	PS	50.4 ± 11.9 μm	Langmuir	$K_L = 0.16$ L/mg	Ultrapure water containing 10 mM NaCl	5 g/L MPs, 10 mg/L AML, 25 °C, pH 6.5	Liu et al. (2020a)

(continued)

Table 2 (continued)

Pharmaceutical	Microplastic	MP particle size	Isotherm type	Isotherm constant (L/Kg)	Matrix	Experimental conditions	Reference
Nonsteroidal anti-inflammatory drugs (NSAIDs)							
Ibuprofen	UHMWPE	2–10 μm	Linear	$K_d = 16.03 \pm 0.68$	Ultrapure water containing 0.01 M $CaCl_2$ – freshwater conditions	10 g/L MPs, 5–350 μg/L each NSAID 25 °C, pH 6.9	Elizalde-Velázquez et al. (2020)
	AMWPE	300–400 μm	Linear	$K_d = 13.26 \pm 0.25$			
	PS	600–800 μm	Linear	$K_d = 7.77 \pm 0.22$			
	PP	~1 mm	Linear	$K_d = 2.48 \pm 0.39$			
Diclofenac	UHMWPE	2–10 μm	Linear	$K_d = 32.30 \pm 0.63$			
	AMWPE	300–400 μm	Linear	$K_d = 26.22 \pm 0.86$			
	PS	600–800 μm	Linear	$K_d = 27.87 \pm 1.08$			
	PP	~1 mm	Linear	$K_d = 12.33 \pm 0.50$			
Naproxen	UHMWPE	2–10 μm	Linear	$K_d = 4.47 \pm 0.44$			
	AMWPE	300–400 μm	Linear	$K_d = 4.29 \pm 0.49$			
	PS	600–800 μm	Linear	$K_d = 2.36 \pm 0.10$			
	PP	~1 mm	Linear	$K_d = 1.86 \pm 0.16$			
Ibuprofen	UHMWPE	2–10 μm	Linear	$K_d = 7.54 \pm 0.91$	Synthetic seawater	10 g/L MPs, 5–350 μg/L each NSAID, 25 °C, pH 8.1	
	AMWPE	300–400 μm	Linear	$K_d = 8.84 \pm 0.32$			
	PS	600–800 μm	Linear	$K_d = 5.49 \pm 0.97$			
	PP	~1 mm	Linear	$K_d = 1.68 \pm 0.39$			
Diclofenac	UHMWPE	2–10 μm	Linear	$K_d = 6.86 \pm 0.22$			
	AMWPE	300–400 μm	Linear	$K_d = 2.48 \pm 0.08$			
	PS	600–800 μm	Linear	$K_d = 2.48 \pm 0.02$			
	PP	~1 mm	Linear	$K_d = 1.63 \pm 0.22$			
Naproxen	UHMWPE	2–10 μm	Linear	$K_d = 1.56 \pm 0.15$			
	AMWPE	300–400 μm	Linear	$K_d = 0.64 \pm 0.15$			
	PS	600–800 μm	Linear	$K_d = 1.61 \pm 0.01$			
	PP	~1 mm	Linear	$K_d = 0.69 \pm 0.05$			

Lipid regulators							
Atorvastatin	PS	50.4 ± 11.9 μm	Langmuir	$K_L = 0.58$ L/mg	Ultrapure water containing 10 mM NaCl	5 g/L MPs, 10 mg/L ATV, 25 °C, pH 6.5	Liu et al. (2020a)
Sex hormones							
17α-Ethinylestradiol	LDPE	250–280 μm	Linear	$K_d = 312 \pm 21.5$	Synthetic freshwater (ultrapure water containing 0.01 M $CaCl_2$ and 0.01% (*w/v*) NaN_3)	0.2 or 1.0 g/L MPs, 10–200 μg/L EE2	Wu et al. (2016)

PE polyethylene, *PP* polypropylene, *PA* polyamide, *PVC* polyvinylchloride, *PS* polystyrene, *HDPE* high-density polyethylene, *PET* polyethylene terephthalate, *UHMWPE* ultrahigh molecular-weight polyethylene, *LDPE* light-density polyethylene, *AMWPE* average molecular-weight medium-density polyethylene

[a] K_F expressed in $((\mu g/g)(mg/L)^{-n})$

[b] K_F expressed in $((mg/kg)(mg/L)^{n})$

[c] K_F expressed in $((mg/kg)(mg/L)1/^{n})$

Table 3 Adsorption kinetics data of pharmaceuticals into aged/weathered microplastics

Pharmaceutical	Microplastic	MP particle size	Aging/weathering process	Adsorption capacity (*qe*)	Equilibrium time	Matrix	Experimental conditions	Reference
Antibiotics								
β-lactam antibiotics (Cephalosporins)								
Cephalosporin C	PS	0.5–1 mm	Naturally aged (obtained from coast of East China Sea and Yellow Sea (China))	58 mg/g	16 h	Freshwater	2 g/L MPs, 2 mg/L CEP-C, 25 °C	Guo and Wang (2019)
	PE (fibers)	0.1–0.2 mm (diameter); 0.5–2 mm (length)		42 mg/g				
	PS	0.5–1 mm		400 mg/g	24 h	Simulated seawater (ultrapure containing 3.5% (w/w) NaCl)		
	PE (fibers)	0.1–0.2 mm (diameter); 0.5–2 mm (length)		255 mg/g				
Fluoroquinolones								
Ciprofloxacin	PS	~75 μm	UV light for 96 h	5.480 mg/g	24 h[a]	Ultrapure water	0.4 g/L MPs, 2–25 mg/L CIP, room temperature	Liu et al. (2019a)
	PVC	~75 μm		3.280 mg/g				
Ciprofloxacin	PS	<10–20 μm	Heat-activated $K_2S_2O_8$	349.45–552.89 mg/g	–[b]	Ultrapure water	0.15 g MPs, 0.15 mg CIP, 25 °C	Liu et al. (2019b)
		<10–30 μm	Fenton reaction	292.64–469.50 mg/g				
	HDPE	<10–20 μm	Heat-activated $K_2S_2O_8$	310.86–533.41 mg/g				
		<10–30 μm	Fenton reaction	244.27–387.13 mg/g				

Sulfonamides								
Sulfamethoxazole	PS	0.5–1 mm	Naturally aged (obtained from coast of East China Sea and Yellow Sea (China))	78 mg/g	16 h	Freshwater	2 g/L MPs, 2 mg/L SMX, 25 °C	Guo and Wang (2019)
	PE (fibers)	0.1–0.2 mm (diameter); 0.5–2 mm (length)		51 mg/g				
Sulfamethazine	PS	0.5–1 mm	Naturally aged (obtained from coast of East China Sea and Yellow Sea (China))	60 mg/g	8 h	Freshwater	2 g/L MPs, 2 mg/L SMT, 25 °C	Guo and Wang (2019)
Tetracyclines								
Tetracycline	PE	60–150 μm	Biofilm developed on MP	762.2 μg/g	–[b]	Ultrapure water containing 0.01 M $NaNO_3$, 0.003 M NaN_3	5 g/L MPs, 5 mg/L TC, 25 °C	Wang et al. (2020b)
Tetracycline	PE	150–250 μm	pH (4, 7, 10), 30 days	91.74–114.94 mg/g	10 days	Ultrapure water	10 g/L MPs, 10 mg/L TC, 20 °C	Shen et al. (2018)
			Temperature (5, 20, 30 °C), 30 days	91.74–125 mg/g				
			Ionic strength (0.01, 0.1, 1 M NaCl), 30 days	99.01–104.17 mg/g				
			Humic acid (2, 10, or 50 mg/L), 30 days	48.1–60.25 μg/g				

(continued)

Table 3 (continued)

Pharmaceutical	Microplastic	MP particle size	Aging/weathering process	Adsorption capacity (*qe*)	Equilibrium time	Matrix	Experimental conditions	Reference
Oxytetracycline	PS (foam)	0.45–1 mm	Naturally aged (plastic debris collected from the coastal beaches of North China). The weathered external surfaces were peeled off and grounded.	–[a]	54 h	Ultrapure water containing 0.01 M NaCl and 25 mg/L NaN_3	1.7 g/L MPs, 20 mg/L OTC, 25 °C	Zhang et al. (2018)
Psychiatric drugs								
Carbamazepine	PE	150–250 μm	Humic acid (2, 10, or 50 mg/L), 30 days	31.4–46.4 μg/g	10 days	Ultrapure water	10 g/L MPs, 20 °C	Shen et al. (2018)
Cardiovascular drugs								
Amlodipine	PS	20.0–35.3 μm	Photo-Fenton reaction	0.47–1.10 mg/g	48h	Ultrapure water containing 10 mM NaCl	5 g/L MPs, 10 mg/L AML, 25 °C, pH 6.5	Liu et al. (2020a)
Lipid regulators								
Atorvastatin	PS	20.0–35.3 μm	Photo-Fenton reaction	0.59–1.33 mg/g	48 h	Ultrapure water containing 10 mM NaCl	5 g/L MPs, 10 mg/L ATV, 25 °C, pH 6.5	Liu et al. (2020a)

PS polystyrene, *PE* polyethylene, *PVC* polyvinylchloride, *HDPE* high density polyethylene
[a]Extrapolated from kinetics graphics
[b]Not indicated

pharmaceuticals on aged/weathered MPs, while Table 4 sums up the sorption equilibrium data of pharmaceuticals on aged/weathered MPs. Just a few studies evaluated the influence of aging/weathering of MPs on sorption processes, and most of them focused on the sorption behavior of antibiotics. Equilibrium time of pharmaceuticals on aged MPs can be reached between 8 h (e.g., sulfamethazine on aged PS) and 10 days (e.g., tetracycline and carbamazepine on aged PE) (Table 3). In general, the sorption equilibrium data of pharmaceuticals on aged/weathered MPs better fits the linear and Langmuir models (Table 4).

Generally, pharmaceuticals are considered hydrophilic compounds, thus it is expected that they may have a high possibility to adsorb to aged MPs, due to their hydrophilicity and to the presence of oxygen-containing functional groups in their surface (Liu et al. 2019a, 2020a). Aged PS and PVC showed a higher sorption capacity for the antibiotic ciprofloxacin than the pristine ones, due to the introduction of oxygen-containing functional groups in their surface, which reduced their hydrophobicity (Liu et al. 2019a). On the other hand, Shen et al. (2018) aged PE for 30 days at different conditions of pH, temperature, and ionic strength, showing that these aging processes only have a slight effect on the low-adsorption capacity of the antibiotic tetracycline on PE. In fact, virgin PE MPs showed a higher adsorption capacity (120.5 μg/g) than aged PE (91.7 μg/g). The authors attributed these results to the lack of functional groups on the surface of PE, which showed a stable chemical structure. An increase in oxidized functional groups in the surface of beached PS foams was reported, showing that weathered PS had more carboxyl and ester carbonyl groups, which could facilitate the adsorption of the antibiotic oxytetracycline, mainly by H-bonding mechanisms (Zhang et al. 2018).

Besides the changes in the physical-chemical properties suffered by MPs, during the aging/weathering processes, plastic additives and MP-derived intermediates, such as oligomers and oxygenated intermediates, may also be released, due to cleavage and oxidation of the polymer chain (Liu et al. 2020a; Liu et al. 2020b). The release of plastic additives is mainly controlled by weathering conditions, such as UV light, oxygen, water pH and temperature, MPs properties, and the additives themselves (Liu et al. 2020b). Plastic additives and intermediates released during the aging/weathering processes may affect the adsorption capacity of pharmaceuticals as was proved by Liu et al. (2020a). The authors observed that the intermediates released from the aging process of PS MPs significantly decreased the adsorption of atorvastatin, but increased the adsorption of amlodipine. These opposite behaviors might be justified due to the competition of the released low-molecular weight intermediates for the adsorption sites of pharmaceuticals, and because most of the released intermediates were negatively charged, therefore the adsorption of the intermediates on MPs could increase the number of negative charges on the surface of aged PS. In this way, the electrostatic attraction of aged PS with positively charged pharmaceuticals, such as amlodipine, would be enhanced, while negatively charged pharmaceuticals, such atorvastatin, would suffer electrostatic repulsion, reducing its adsorption capacity (Liu et al. 2020a). Although the aging intermediates may affect the adsorption behavior of MPs, there is still a gap of knowledge on their effects in the adsorption process between pharmaceuticals and aged MPs. Nevertheless, it

Table 4 Sorption isotherms data of pharmaceuticals on aged/weathered microplastics

Pharmaceutical	Microplastic	MP particle size	Aging /weathering process	Isotherm type	Isotherm constant (L/Kg)	Matrix	Experimental conditions	Reference
Antibiotics								
β-lactam antibiotics (Cephalosporins)								
Cephalosporin C	PS	0.5–1 mm	Naturally aged (obtained from coast of East China Sea and Yellow Sea (China))	Linear	$K_d = 0.0236$ L/g	Freshwater	2 g/L MPs, 0–10 mg/L CEP-C, 25 °C	Guo and Wang (2019)
	PE (fibers)	0.1–0.2 mm (diameter); 0.5–2 mm (length)		Linear	$K_d = 0.0370$ L/g			
	PS	0.5–1 mm		Langmuir	$K_L = 0.000366$ L/μg	Simulated seawater (ultrapure containing 3.5% (w/w) NaCl)		
	PE (fibers)	0.1–0.2 mm (diameter); 0.5–2 mm (length)		Linear	$K_d = 0.0567$ L/g			
Fluoroquinolones								
Ciprofloxacin	PS	~75 μm	UV light for 96 h	Linear	$K_d = 0.318$	Ultrapure water	0.4 g/L MPs, 2–25 mg/L CIP, room temperature	Liu et al. (2019a)
	PVC	~75 μm		Linear	$K_d = 0.251$			
Ciprofloxacin	PS	<10–20 μm	Heat-activated $K_2S_2O_8$	Langmuir	$K_L = 0.302$ L/mg	Ultrapure water	0.15 g MPs, 0.15 mg CIP, 25 °C	Liu et al. (2019b)
		<10–30 μm	Fenton reaction	Langmuir	$K_L = 0.241$ L/mg			
	HDPE	<10–20 μm	Heat-activated $K_2S_2O_8$	Langmuir	$K_L = 0.173$ L/mg			
		<10–30 μm	Fenton reaction	Langmuir	$K_L = 0.123$ L/mg			

Sulfonamides								
Sulfamethoxazole	PS	0.5–1 mm	Naturally aged (obtained from coast of East China Sea and Yellow Sea (China))	Linear	$K_d = 0.0328$ L/g	Freshwater	2 g/L MPs, 0–10 mg/L SMX, 25 °C	Guo and Wang (2019)
	PE (fibers)	0.1–0.2 mm (diameter); 0.5–2 mm (length)		Linear	$K_d = 0.0383$ L/g			
Sulfamethazine	PS	0.5–1 mm	Naturally aged (obtained from coast of East China Sea and Yellow Sea (China))	Linear	$K_d = 0.0293$ L/g	Freshwater	2 g/L MPs, 0–10 mg/L SMT, 25 °C	Guo and Wang (2019)
Tetracyclines								
Tetracycline	PE	60–150 μm	Biofilm developed on MP	Freundlich	$K_F = 56.0$[a]; $n = 2.20$	Ultrapure water containing 0.01 M $NaNO_3$, 0.003 M NaN_3	5 g/L MPs, 0–15 mg/L TC, 25 °C	Wang et al. (2020b)
Oxytetracycline	PS (foam)	0.45–1 mm	Naturally aged (plastic debris collected from the coastal beaches of North China). The weathered external surfaces were peeled off and grounded	Freundlich	$K_F = 894 \pm 84$[b]; $1/n = 0.75 \pm 0.03$	Ultrapure water containing 0.01 M NaCl and 25 mg/L NaN_3	1.7 g/L MPs, 2–50 mg/L OTC, 25 °C	Zhang et al. (2018)

(continued)

Table 4 (continued)

Pharmaceutical	Microplastic	MP particle size	Aging /weathering process	Isotherm type	Isotherm constant (L/Kg)	Matrix	Experimental conditions	Reference
Cardiovascular drugs								
Amlodipine	PS	20.0–35.3 μm	Photo-Fenton reaction	Freundlich (PS-12, PS-48)	$K_F = 0.31–0.42$; $n = 0.25–0.26$	Ultrapure water containing 10 mM NaCl	5 g/L MPs, 10 mg/L AML, 25 °C, pH 6.5	Liu et al. (2020a)
				Langmuir (PS-108)	$K_L = 0.57$ L/mg			
Lipid regulators								
Atorvastatin	PS	20.0–35.3 μm	Photo-Fenton reaction	Langmuir	$K_L = 0.28–0.41$ L/mg	Ultrapure water containing 10 mM NaCl	5 g/L MPs, 10 mg/L ATV, 25 °C, pH 6.5	Liu et al. (2020a)

PS polystyrene, *PE* polyethylene, *PVC* polyvinylchloride, *HDPE* high density polyethylene

[a] K_F expressed in $((\mu g/g)(mg/L)^{-n})$

[b] K_F expressed in $((mg/kg)(mg/L)1/^{n})$

should be in mind that intermediates may interact either with aged MPs or pharmaceuticals, and their effects will be dependent of the degree of aging of MPs and the properties of pharmaceuticals (Liu et al. 2020a).

Influence of Natural Factors (e.g., pH, salinity, DOM, etc.) in the Sorption Processes

Natural environments are complex systems that have many factors that could influence in the sorption processes between MPs and pharmaceuticals. Natural factors such as pH, salinity, dissolved organic matter (DOM), and temperature, among others, should be taken into account, since they have the ability to interfere in the sorption of pharmaceuticals on MPs in different ways, and are integral part of natural environments. In general, pH, salinity, and the presence of DOM are among the natural factors that mostly affect the sorption capacity of pharmaceuticals on MPs.

Dissolved Organic Matter (DOM)

Dissolved organic matter (DOM) can interfere in the sorption processes of pharmaceuticals on MPs in different ways, namely: (1) DOM has various functional groups that can directly compete with contaminants for the sorption sites of MPs, decreasing their sorption; (2) DOM can interact with MPs through its abundant oxygen functional groups, changing their polarity and surface charge; and (3) DOM can interact with hydrophilic and polar contaminants by complexation and hydrogen bonding, affecting the sorption of contaminants on MPs. Additionally, DOM can firstly adsorb to MPs and then interact with pharmaceuticals too (Wu et al. 2016; Liu et al. 2020b).

Shen et al. (2018) showed that humic acid (HA) significantly decreased the adsorptive capacity of MPs for the antibiotic tetracycline. This might be due to the high number of functional groups on HA that could change the surface charge of MPs, reducing the adsorption affinity of tetracycline, or by a competition between HA and tetracycline for the adsorption sites on the surface of MPs. A significant decrease (more than 90%) in the sorption capacity of tetracycline on PS, PP, and PE in the presence of DOM was also shown by Xu et al. (2018a). But in this case, the authors suggested that the decrease of the adsorption of tetracycline was due to the complexation of this antibiotic with the carboxylic functional groups of DOM, instead of a change on the surface of MPs. An increase in DOM concentration also reduced the adsorption of 17α-ethinylestradiol on PE MPs, due to the interaction of this pharmaceutical to DOM (Wu et al. 2016). Oppositely, the adsorption of oxytetracycline on naturally aged PS foams was enhanced in the presence of DOM (Zhang et al. 2018). This might be justified by the complexation of DOM with MPs surface, which worked as a bridge between aged PS MPs and the antibiotic oxytetracycline. However, not all pharmaceuticals would be affected by the presence of DOM. A negligible effect in the adsorption of sulfamethoxazole (Xu et al. 2018b) or carbamazepine (Wu et al. 2016) on PE MPs was observed. This was attributed to a greater affinity of these pharmaceuticals to PE MPs than to DOM. All in all, DOM

has the ability to interfere with the sorption capacity of pharmaceuticals on MPs by interacting with both MPs and pharmaceuticals.

Furthermore, the presence of DOM in the aquatic environment may interfere with the sorption processes of pharmaceuticals on MPs by enhancing the desorption of pharmaceuticals such as tetracycline from MPs (Xu et al. 2018a), affecting the fate and transport of pharmaceuticals by MPs.

pH

The environmental pH may influence the sorption capacity of pharmaceuticals on MPs, especially in what concern to ionizable compounds. Both the p*Ka* of pharmaceuticals and the pH point of zero charge (pH_{pzc}) of MPs will directly be influenced by the water pH, since the former will define the speciation of pharmaceuticals and the latter the surface charge of MPs. Thus, a higher sorption capacity will be expected for neutral species, while, in general, a weaker sorption will occur for charged species (Li et al. 2018; Elizalde-Velázquez et al. 2020). Xu et al. (2018a) showed that electrostatic repulsion determines the sorption process of tetracycline on three MPs (PS, PP, and PE) in acidic and alkaline aqueous environments, because of the same positive or negative charges between the antibiotic and the MPs, inhibiting the sorption. The same behavior was observed by Wang et al. (2020b) for the adsorption of tetracycline on PE MPs. Nevertheless, when the authors considered the adsorption of this antibiotic on biofilm-developed PE MPs, the same trend was not exhibited, probably due to the complexation effects of biofilm. On the other hand, in freshwater systems (pH between 6.5 and 8.5), the sorption capacity of tetracycline on PS, PP, and PE was higher. On PE and PP, the sorption of tetracycline occurred mainly through hydrophobic interactions, thus a pH increase or decrease would inhibit the adsorption of this antibiotic by electrostatic repulsion, since more tetracycline in the ionizable form would be present in the water. In the case of PS, besides the aforementioned mechanisms, polar interactions and π-π interactions could also occur, which increased its sorption affinity for tetracycline (Xu et al. 2018a). The pH of the medium also influences in the adsorption of NSAIDs (ibuprofen, diclofenac, and naproxen) on four MPs (UHMWPE, AMWPE, PP, and PS). The highest adsorption behavior was seen at pH 2, because under acidic conditions, the NSAIDs were predominantly in the nonionized form and there was no electrostatic repulsion with the positively charged surface of MPs. Nevertheless, at pH 6.9 and 10, both NSAIDs and MPs were negatively charged, reducing the sorption capacity due to electrostatic repulsion (Elizalde-Velázquez et al. 2020). This behavior contributes for the low-adsorption capacity of NSAIDs on MPs in freshwater (pH = 6.9) and seawater (pH = 7.5–8.4), since the electrostatic repulsion between the negatively charged surface of MPs and the ionic species of NSAIDs is promoted (Elizalde-Velázquez et al. 2020). The sorption of the antibiotics tylosin (Guo et al. 2018), sulfamethoxazole (Guo et al. 2019a), and sulfamethazine (Guo et al. 2019b) on MPs also decreased with the increasing of pH, mainly because at alkaline pH MPs are always negatively charged and antibiotics can be in their anionic form, such as sulfamethoxazole or sulfamethazine, increasing the electrostatic repulsion between antibiotic and MPs, or in their molecular form, such as

tylosin, preventing the occurrence of hydrophobic interactions between antibiotic and negatively charged MPs. In conclusion, changes in the environmental pH will mostly influence the sorption of pharmaceuticals on MPs for which electrostatic interactions are the main mechanisms involved in their sorption process.

Attending to this behavior, for pH-dependent compounds such as antibiotics or NSAIDs, changes in the medium pH might also influence the desorption of compounds loaded on MPs due to electrostatic repulsion, which would contribute for the transference of pharmaceuticals from MPs into the environment (Xu et al. 2018a).

Nevertheless, there are also pharmaceuticals whose adsorption on MPs is not affected by changes in the pH as it is the case of oxytetracycline. Zhang et al. (2018) showed that the adsorption of this antibiotic on PS MPs was not influenced by the pH of the medium. Xu et al. (2018b) also did not see a significant effect of pH on the adsorption of sulfamethoxazole on PE MPs.

Salinity

Water salinity is an important factor that may significantly affect the sorption of contaminants on MPs. The effect of salinity is compound-dependent, depending on the properties of organic contaminants (e.g., water solubility) and plastic polymers as well as from the sorption mechanisms involved in the sorption process.

The salinity of the surrounding environment affects the water solubility of organic contaminants, thus influencing their adsorption behavior onto MPs. In general, an increase in the salinity is expected to decrease the solubility of nonpolar and weakly polar organic contaminants in water by an effect of salting out (Alimi et al. 2018). However, in the case of pharmaceuticals, since they are predominantly polar compounds (hydrophilic contaminants), it is expected that an increase in the salinity will increase their solubility, reducing their adsorption capacity to MPs. This behavior was observed for the antibiotic ciprofloxacin that showed a decrease in its adsorption efficiency on PS and PVC MPs due to an increase in the NaCl content (Liu et al. 2019a). Wang et al. (2020a) also reported that the adsorption of antibiotics to PE MPs seemed more favorable in freshwater (low salinity) than in seawater (high salinity).

In general, salinity has a more significant effect on the sorption mechanisms of ionic compounds than on hydrophobic partition sorption mechanisms (Xu et al. 2018b; Elizalde-Velázquez et al. 2020; Wang et al. 2020a). An increase in the salinity represents a higher ionic strength of the medium due to the presence of a higher amount of electrolytes, as happens in seawater, which could affect the electrostatic interactions, because there is a competition between electrolytes, such as the cations Na^+ and Ca^{2+}, and pharmaceuticals for the electrostatic sites present in the MPs surface (Li et al. 2018). Additionally, the presence of inorganic exchangeable cations (e.g., Na^+) can also replace the hydrogen ions of the acidic groups present in the MPs surface, thus inhibiting the formation of H-binding (Li et al. 2018). Li et al. (2018) showed differences in the adsorption of antibiotics (sulfadiazine, amoxicillin, tetracycline, ciprofloxacin, and trimethoprim) on MPs (PE, PS, PP, PA, and PVC) between freshwater and seawater systems. For instance, in the seawater system, there was not adsorption of ciprofloxacin and amoxicillin, while

the sorption capacity of trimethoprim, sulfadiazine, and tetracycline was reduced comparatively to the freshwater system. The authors attributed the differences mainly to the variation of the ionic strength, which represented a decrease in the available adsorption sites, and to pH of the medium (freshwater: pH = 6.7–7.1 vs. seawater: pH = 8.0). Wang et al. (2020a) also evaluated the adsorption capacity of four antibiotics (sulfamerazine, tetracycline, chloramphenicol, and tylosin) on PE MPs in waters with different salinities, namely, river, estuary, and seawater, showing that MPs can concentrate more antibiotics in river freshwater than in estuary and marine waters. In fact, sulfamerazine and chloramphenicol adsorbed on PE MPs in river water (37.55 ± 1.73 μg/g and 4.69 ± 0.97 μg/g, respectively) were almost twice more than in estuary (17.17 ± 1.52 μg/g and 1.56 ± 0.21 μg/g, respectively) and seawater (12.13 ± 1.37 μg/g and 0.52 ± 0.04 μg/g, respectively). The observed differences were attributed to changes in the total salt content and chemical composition of the water. In the presence of higher content of salts, a competition for the adsorption sites against the antibiotics could happen. Sea salts might also reduce cation exchange sites, leading to a weaker adsorption of antibiotics on PE MPs in seawater (Wang et al. 2020a). A decrease in the adsorption of sulfamethazine on six MPs (PA, PE, PS, PET, PVC, and PP) with the increase of salinity was also reported, being attributed to the higher content of Na^+ ions available to interact with the anionic surface of MPs (Guo et al. 2019b). On the other hand, contradictory behaviors were registered for the adsorption of sulfamethoxazole on MPs. Although Guo et al. (2019a) reported a decrease in the adsorption capacity of this antibiotic on PA, PE, PET, PVC, and PP, due to an increase in the salinity, Xu et al. (2018b) showed that its sorption on PE MPs was not significantly affected by salinity. Negligible effects on the adsorption of tetracycline on PS, PP, and PE MPs (Xu et al. 2018a) and on the adsorption of carbamazepine and 17α-ethinylestradiol on PE MPs (Wu et al. 2016) due to an increase in the salinity were also reported.

An increase in the salinity of the medium may also induce a decrease in the adsorption efficiency of pharmaceuticals on aged MPs as was proved by Liu et al. (2019a). These authors showed a decrease in the adsorption of ciprofloxacin on aged PVC MPs, while no significant effect was seen on the adsorption behavior of the same antibiotic on aged PS MPs. On the other hand, the sorption of sulfamethazine and sulfamethoxazole on naturally aged PS and PE MPs in seawater was not observed, while the sorption of cephalosporin C on both aged MPs was enhanced (Guo and Wang 2019). This might be attributed to a decrease in the solubility of the antibiotic and to its complexation with the aged MPs surface.

All in all, adsorption of pharmaceuticals on MPs seems to be more favored in freshwater ecosystems than in seawater due to the differences in the salinity of both ecosystems. Furthermore, these changes in the sorption behavior of pharmaceuticals on MPs due to salinity reinforce the idea that MPs may act as carriers of pollutants, since they are able to release pharmaceuticals sorbed from rivers, estuaries to oceans, attending to the increase of salinity between these environmental compartments, which is favorable to the desorption of pharmaceuticals from MPs (Zhang et al. 2020).

Conclusions and Future Perspectives

Understanding the sorption behavior of pharmaceuticals on MPs is essential to better evaluate the environmental impact of these emerging contaminants that can coexist due to their great use worldwide. Therefore, some aspects and current gaps of knowledge should be considered for further studies:

- Although some studies already focus on the evaluation of the adsorption capacity of real MPs, most of them are conducted using pristine MPs, which are commercially available. However, care should be taken in the extrapolation of the results from sorption experiments made with pristine MPs to real scenarios, since environmental conditions, such as aging processes, also play an important role in the sorption behavior of pharmaceuticals on MPs.
- Besides that, real MPs have other constituents in addition to the plastic polymer as, for instance, plasticizers, stabilizers, and pigments, among others, which may interfere in their adsorption behavior. Thus, plastic additives should also be taken into account on the evaluation of the sorption behavior of pharmaceuticals on MPs.
- Since sorption processes are polymer-pharmaceutical specific, as was seen in this chapter, more studies are needed to better understand the interaction between pharmaceuticals and MPs as well as the mechanisms involved, especially in what concerns to aged MPs, which represent a more realistic scenario. In this way, a deeper knowledge of the role of MPs in the fate and transport of pharmaceuticals in the environment can be attained as well as of the environmental impact of MPs.
- Furthermore, more sorption studies using real MPs and focusing on environmental relevant concentrations of contaminants are needed for a more realistic evaluation of the interaction of pharmaceuticals with MPs in the environment.
- It was seen that natural factors, such as pH, salinity, and DOM, may influence the adsorption of pharmaceuticals on MPs. Nevertheless, most of the studies only focus on the impact of these factors on pristine MPs. Further studies should be extended to aged MPs, in order to increase the knowledge on their sorption behavior. Other natural factors, such as temperature, should also be considered, since sorption isotherms and mechanisms may vary with temperature.
- Finally, the desorption behavior between pharmaceuticals and MPs should also be evaluated. Presently, data on desorption is limited, nevertheless this might be a critical factor when contaminated MPs are ingested by aquatic organisms. MPs may act as vector of pharmaceuticals into the tissues by desorption. Thus, the desorption of pharmaceuticals from MPs under physiological conditions of pH and temperature should be assessed, given that these factors may influence the sorption behavior of pharmaceuticals on MPs. Desorption of pharmaceuticals may be relevant on their toxicity to aquatic organisms.

Acknowledgments This work was funded by the Spanish State Research Agency of the Spanish Ministry of Science, Innovation and Universities and European Fund for Regional Development (project PLAS-MED; FEDER-MCIU-AEI/CTM2017-89701-C3-2-R). Authors thank Generalitat

de Catalunya through Consolidated Research Group (ICRA-ENV 2017 SGR 1124 & 2017 SGR 1404). ICRA researchers thank funding from CERCA program. Lúcia H.M.L.M. Santos thanks the Juan de la Cierva program (IJCI-2017-32747) and Sara Rodríguez-Mozaz thanks the Ramon y Cajal program (RYC-2014-16707) from the Spanish State Research Agency of the Spanish Ministry of Science, Innovation and Universities (AEI-MCIU).

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