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Review article

Interactions between microplastics, pharmaceuticals and personal care products: Implications for vector transport

Thilakshani Atugoda [a], Meththika Vithanage [a,*], Hasintha Wijesekara [b], Nanthi Bolan [c], Ajit K. Sarmah [d], Michael S. Bank [e], Siming You [f], Yong Sik Ok [g,*]

[a] Ecosphere Resilience Research Center, Faculty of Applied Sciences, University of Sri Jayewardenepura, Nugegoda 10250, Sri Lanka
[b] Department of Natural Resources, Faculty of Applied Sciences, Sabaragamuwa University of Sri Lanka, Belihuloya 70140, Sri Lanka
[c] Global Centre for Environmental Remediation (GCER), The University of Newcastle (UON), Callaghan, NSW 2308, Australia
[d] Department of Civil and Environmental Engineering, Faculty of Engineering, The University of Auckland, Private Bag 92019, Auckland 1142, New Zealand
[e] Institute of Marine Research, Bergen, Norway
[f] James Watt School of Engineering, James Watt South Building, University of Glasgow, Glasgow G12 8QQ, UK
[g] Korea Biochar Research Center, APRU Sustainable Waste Management Program & Division of Environmental Science and Ecological Engineering, Korea University, Seoul 02841, South Korea

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ABSTRACT

Microplastics are well known for vector transport of hydrophobic organic contaminants, and there are growing concerns regarding their potential adverse effects on ecosystems and human health. However, recent studies focussing on hydrophilic compounds, such as pharmaceuticals and personal care products (PPCPs), have shown that the compounds ability to be adsorbed onto plastic surfaces. The extensive use of PPCPs has led to their ubiquitous presence in the environment resulting in their cooccurrence with microplastics. The partitioning between plastics and PPCPs and their fate through vector transport are determined by various physicochemical characteristics and environmental conditions of specific matrices. Although the sorption capacities of microplastics for different PPCP compounds have been investigated extensively, these findings have not yet been synthesized and analyzed critically. The specific objectives of this review were to synthesize and critically assess the various factors that affect the adsorption of hydrophilic compounds such as PPCPs on microplastic surfaces and their fate and transport in the environment. The review also focuses on environmental factors such as pH, salinity, and dissolved organics, and properties of polymers and PPCP compounds, and the relationships with sorption dynamics and mechanisms. Furthermore, the ecotoxicological effects of PPCP-sorbed microplastics on biota and human health are also discussed.

1. Introduction

The recent discovery of microplastics present in the environment as a ubiquitous pollutant has become a global concern and has resulted in intensive research in this field. Microplastics are a source of terrestrial and aquatic contamination in the environment, and may be found in soils (Huang et al., 2020; Ng et al., 2018), surface waters (Fischer et al., 2016; Peng et al., 2018), lagoons and estuaries (Bakir et al., 2014a), coastal shorelines (Jeyasanta et al., 2020), pelagic and benthic regions of the sea (Bollmann et al., 2019; Cincinelli et al., 2019), Arctic freshwaters (González-Pleiter et al., 2020), ice (Obbard et al., 2014), and the ocean (Fang et al., 2018; Kanhai et al., 2019). Microplastics have been defined as any type of plastic fragments, fibers, or beads with diameter ranging from 100 nm to < 5 mm; particles with sizes < 100 nm are

Abbreviations: PPCPs, pharmaceuticals and personal care products; HOCs, hydrophobic organic contaminants; PAHs, polyaromatic hydrocarbons; PCBs, polychlorinated biphenyls; PBDs, polybrominated diethers; PCB77, 3,3′,4,4′-tetrachlorobiphenyl, polyfluorinated alkyl substances, PFAS; DOM, dissolved organic matter; HA, humic acid; FA, fulvic acid; PE, polyethylene; PP, polypropylene; PS, polystyrene; PVC, polyvinyl chloride; PA, polyamide; PET, polyethylene terephthalate; PMMA, poly(methyl methacrylate); AMWPE, average-molecular weight medium-density polyethylene; MDPE, medium-density polyethylene; LDPE, low-density polyethylene; LLDPE, linear low-density polyethylene; HDPE, high-density polyethylene; UHMWPE, ultra-high-molecular weight polyethylene; K_d, Langmuir partition coefficient; K_f, Freundlich partition coefficient; K_{ow}, octanol-water partition coefficient; D_{ow}, octanol-water distribution ratio.

* Corresponding authors.
E-mail addresses: meththika@sjp.ac.lk (M. Vithanage), yongsikok@korea.ac.kr (Y.S. Ok).

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referred to as nanoplastics (Fig. 1) (Alimi et al., 2018; Michielssen et al., 2016).

Based on their formation pathway, microplastics can be categorized as primary and secondary particles (Bradney et al., 2019; Duis and Coors, 2016). Synthetically manufactured plastic pellets, beads, nurdles, fibers, and powders for commercial purposes are known as primary microplastics. These are widely used as feedstock for plastic production (e.g., resin pellets) or appliance manufacturing, textile fibers in clothing (e.g., acrylic fibers), industrial abrasives (e.g., air blasting), and exfoliants incorporated in personal care and cosmetic products (e.g., microbeads in hand and facial cleansers) (Cincinelli et al., 2019; Cole et al., 2011; Duis and Coors, 2016; Li et al., 2019). For example, cosmetic formulations may contain 0.5%–5% primary microplastics, and in a single use, and may release approximately 4500–94,500 microbeads (Prata, 2018). Secondary microplastic particles are formed by the weathering and degradation of macroplastic and mesoplastic debris in the environment. These are considered to be the main sources of most microplastics found in marine ecosystems, although we have limited knowledge regarding the actual contributions of inputs from several sources including landfills, coastal littering, ports, fishing gear, and shipping (Kane et al., 2020; Kazour et al., 2019; Law and Thompson, 2014; Waldschläger et al., 2020). Weathering and degradation cause physical and chemical fragmentation of polymeric materials, thereby reducing their size and making them brittle, eventually turning them into powdery fragments (Andrady, 2011).

The hydrophobic nature and high surface area-to-volume ratio of microplastics facilitate the accumulation of organic contaminants, including polyaromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs), perfluorinated alkyl substances (PFAS), polybrominated diethers (PBDs), and pharmaceuticals and personal care products (PPCPs) (Bakir et al., 2012; Li et al., 2018a; Mato et al., 2001; Teuten et al., 2009), as well as trace metal contaminants (Ag, Cd, Co, Cr, Cu, Hg, Ni, Pb, and Zn) (Brennecke et al., 2016; Guo et al., 2020; Guzzetti et al., 2018) on the surface of microplastics. Hydrophobic organic contaminants (HOCs) have a high tendency to get adsorbed on non-polar surfaces, including sediments and organic matter; however, they often have a greater affinity for plastic surfaces (Carbery et al., 2018). Microplastics provide a large solid surface especially within aqueous environments, and the amounts of organic contaminants accumulated on the plastic surface can be several orders of magnitude higher than that in the surrounding waters. Laboratory studies have shown that the HOC sorption capacity of microplastics range from 1 to 10,000 ng g^{-1} (Hartmann et al., 2017). In Japan, the amount of accumulated PCBs and PBDs on plastic pellets has been reported to be 10^5–10^6 times higher than those in the surrounding seawater (Mato et al., 2001). In simulated seawater, the maximum sorption capacity of polypropylene microplastics for 3,3′,4,4′-tetrachlorobiphenyl (PCB77) was estimated to be approximately 350 μg g^{-1} (Zhan et al., 2016).

Over the past decade, synthetic organic chemicals have become an indispensable component in modern human societies owing to their widespread application especially in PPCPs. The extensive use of these compounds has led to their ubiquity in aquatic environments (López-Serna et al., 2013; Oberg and Leopold, 2019). PPCPs consist of complex organic compounds, which possibly transform into harmful secondary byproducts that are persistent and have extremely long residence times in the environment (Yin et al., 2017). In surface waters and wastewaters,

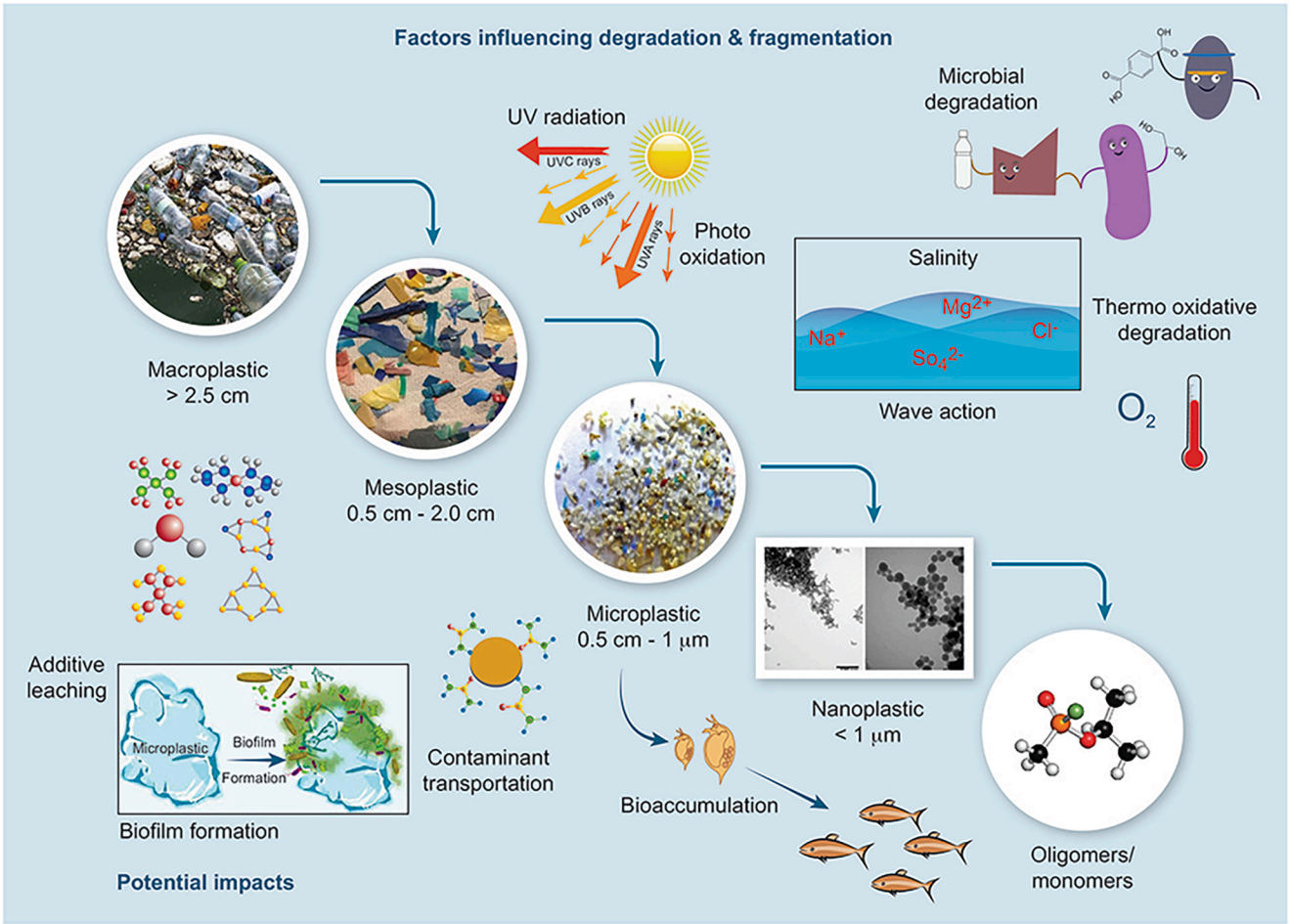

Fig. 1. Pathways to microplastic formation in the environment and their potential impact (adapted from Jahnke (2017), Rummel et al. (2017)).

approximately 160 pharmaceuticals belonging to commonly used medication groups, such as antibiotics, anti-inflammatory drugs, and medicines for heart diseases, have been detected (Kümmerer, 2009). The cooccurrence of microplastics and PPCPs in an aquatic ecosystem leads to their mutual interaction and the subsequent sorption of contaminants on microplastic surfaces. Recently, the sorption capabilities of plastic substances for commonly used antibiotic compounds have also been reported (Guo et al., 2018; Li et al., 2018a). In general, PPCPs are highly hydrophilic and polar with a low octanol–water partition coefficients (K_{ow}) and low volatility (Caliman and Gavrilescu, 2009; Ebele et al., 2017). The partitioning of an organic compound to plastic is usually governed by its hydrophobicity, which is related to its K_{ow}; for instance, HOCs with a high K_{ow} have a high affinity for non-polar polymers, and thus, a high accumulation capability on these polymers (Hüffer and Hofmann, 2016; O'Connor et al., 2016). However, hydrophobicity alone does not affect the sorption behavior which; is also affected by polymer type, degree of crystallinity, and environmental conditions (e.g., pH and salinity) (Guo et al., 2018; Li et al., 2018a).

In recent, there have been significant concern about microplastics as drivers for antibiotics resistance genes as result of the interaction between microplastics and antibiotic (Laganà et al., 2019). The exposure of antibiotics to microbial communities may likely induce mutagenic transformation to develop genetic resistance towards a particular antibiotic, and there onwards transfer the genes through propagation across other bacterial strains (Rizzo et al., 2013). Combined effect of microplastics and antibiotics can enhance the emergence of antibiotic resistance genes (Ma et al., 2020). These resistance genes are transported across the environmental matrices as these microbes colonize on the particle surface and drift away through soil and water (Bank et al., 2020).

The sorption behavior of microplastics for HOCs has been extensively studied; however, comprehensive reviews on microplastic–PPCP interactions are lacking (Koelmans et al., 2016; Verla et al., 2019). The interaction of microplastics with pharmaceuticals along with other emerging contaminants has been recently discussed by Vieira et al. (2021). However, to date, no literature reviews have been reported that are devoted entirely to PPCP-microplastic interactions and the associated molecular level mechanism. The mechanism of sorption of hydrophilic compounds by microplastics is unique and the sorption dynamics of microplastic-hydrophobic compound interaction require further assessment (Wu et al., 2016). Therefore, the goal of this review is to critically analyze and summarize the existing knowledge and research findings on the interactions between microplastics and PPCP compounds. The factors affecting microplastic–PPCP interactions, vector-assisted transportation, and the sources, fate, transport and impact of PPCP-sorbed microplastics on the environment and ecosystems are highlighted in this review. The specific objectives of this literature review were to evaluate and synthesize information pertaining to: (1) microplastic–PPCP interactions based on (a) equilibrium time and initial concentration, (b) solution chemistry of the medium such as pH, dissolved organics, and salinity, and biofouling (c) physicochemical properties of plastics and PPCP compounds; (2) the plausible mechanisms of the sorption of PPCPs on microplastics; and (3) the potential risks and hazards of microplastic–PPCP interaction on aquatic biota and human health.

2. Interactions between microplastics and PPCPs

The interactions between microplastics and PPCP compounds have been widely studied. The adsorption capacity depends on the characteristics of PPCP compounds, polymeric properties of microplastics, and matrix effects (salinity, pH, and dissolved organics) (Daugherty, 2016). Most PPCP compounds, especially several groups of antibiotics, are polar and ionizable, and their presence or speciation is often pH dependent. Therefore, cationic, anionic, and zwitterionic species interact differently with the plastic surface depending on the surface charge and functional groups. The inorganic and organic solute ions including dissolved organic matter (DOM) present in the matrix interfere with the sorption mechanism occurring between the plastic surface and the compound. Sometimes, organic matter and ions compete with PPCPs to interact with the active sites, resulting in the suppression of the sorption process (Guo and Wang, 2019a; Liu et al., 2019b). In contrast, this can have synergistic effects in increasing the overall affinity by rendering the surface more hydrophobic. The initial concentration of PPCPs is crucial because it determines the maximum sorption capacity. At low concentrations, sorption occurs rapidly as there is a high probability for molecules to attach to binding sites. However, with increasing concentrations, the active sites become saturated and sorption rapidly reaches a steady-state equilibrium (Zhang et al., 2018b).

2.1. Kinetics of microplastic–PPCP interaction

Most kinetic studies based on microplastics have mainly focused on antibiotics followed by a few PPCP compounds, such as non-steroidal anti-inflammatory drugs (NSAIDs), steroidal hormones, antimicrobial agents, and synthetic musks (Table 1). Kinetic equilibrium studies have been conducted on polyethylene (PE), polystyrene (PS), polyvinyl chloride (PVC), and polypropylene (PP) (Guo et al., 2018; Razanajatovo et al., 2018; Yu et al., 2020; Zhang et al., 2018a). For example, in 50% of the adsorption studies, equilibrium was reached within 24 h, whereas in a few cases, equilibrium was reached in 4 days (Guo et al., 2018; Guo and Wang, 2019a; Liu et al., 2019b; Zhang et al., 2018b). At a high ratio of initial PPCP concentration to microplastic dosage, the sorption equilibrium was rapidly established. Moreover, at the same initial contaminant concentration (triclosan and tetracycline) of 5 mg L^{-1}, the sorption equilibrium was reached at 72 h and 33 h when the dosages of microplastic were 2.5 g L^{-1} (Li et al., 2019) and 0.4 g L^{-1} (Fan et al., 2021), respectively. However, this is likely only applicable for one type of microplastic and PPCP when the compound is assessed under similar experimental conditions. In all the studies listed in Table 1, the sorption equilibrium time ranged from 8 h to 4 days.

2.2. The initial concentration of sorbate

2.2.1. Antibiotics

Most sorption isotherm studies based on microplastics have been performed with antibiotics, as summarized in Table 2. The concentration of antibiotics used varied from 1 μg L^{-1} to 500 mg L^{-1}. Although the environmental concentrations of antibiotics generally range from ng L^{-1} to μg L^{-1}, higher concentrations were used to assess the maximum adsorption capacity of microplastics. The sorption capacity as a function of initial concentration was studied mostly with antibiotic compounds including sulfonamides, tetracyclines, and fluoroquinolones. In some cases, very high sorption capacities were observed; for example, the sorption capacities of aged PS and PVC microplastics for ciprofloxacin were 54.8 and 15.5 mg g^{-1}, as derived using the Langmuir model (Liu et al., 2019b). Additionally, Li et al. (2018a) reported a high Langmuir sorption coefficient (K_d) of 22.7 mg g^{-1} for the sorption of amoxicillin on polyamide (PA) microplastics (Li et al., 2018a). In most studies, PE, PS, PP, and PVC microplastics were used for the isotherm studies of antibiotic adsorption (Guo et al., 2018; Liu et al., 2019b; Zhang et al., 2018a).

The effects of other factors, such as dissolved organic matter (DOM) and ionic strength, on the binding of antibiotics to microplastics are not well studied (Atugoda et al., 2020; Elizalde-Velázquez et al., 2020; Li et al., 2018a; Zhang et al., 2018a). For example, Zhang et al. (2018) studied the sorption of oxytetracycline across a wide range of humic acid (HA) and fulvic acid (FA) concentrations and found that the adsorption of oxytetracycline increased with increasing DOM content (Zhang et al., 2018a). With an increase in HA and FA concentrations to 100 mg L^{-1}, the Freundlich partition coefficient (K_f) increased from 894 $mg^{(1/1-N)}$ kg^{-1} $L^{1/N}$ to 3,940 and 1,980 $mg^{(1/1-N)}$ kg^{-1} $L^{1/N}$ for beached PS foams,

Table 1
Experimental kinetic studies on PPCP sorption by microplastics.

PPCP contaminant	Microplastic type	Initial concentration/ (mg L^{-1})	Dosage (g L^{-1})	Time to reach equilibrium (h)	PPCP sorption capacity (μg g^{-1})		Other fitting models	Reference
					Pseudo-first order	Pseudo-second order		
Oxytetracycline	PS Aged PS*	20	1.66	-	-	-	Intraparticle diffusion	(Zhang et al., 2018a)
Ciprofloxacin	PS UV aged PS PVC UV aged PVC	10	0.4	24	2,580 4,860 2,790 3,070	3,170 5,480 3,410 3,280	-	(Liu et al., 2019b)
Ciprofloxacin	PE	25	2.0	3	-	-	Parabolic diffusion Elovich	(Atugoda et al., 2020)
Tylosin	PE PP PS PVC	5	-	36	319 516 446 647	667 833 1,429 1,667	-	(Guo et al., 2018)
Tetracycline	PE	5	0.5	80	-	116	-	(Yu et al., 2020)
Sulfamethoxazole	PE	1	5	24	102	108	-	(Xu et al., 2018a)
Sulfamethoxazole	PA PE PET PS PVC PP	2.4	2	16	-	-	External mass transfer	(Guo et al., 2019a)
Sulfamethoxazole **Sulfamethazine** **Cephalosporin-C**	Aged PS** Aged PE**	2	2	16	-	-	Mixed order model	(Guo and Wang., 2019a)
Tetracycline	PLA Aged PLA PVC Aged PVC	5	0.4	33	460 1,280 430 870	900 1,970 750 1,360	-	(Fan et al., 2021)
Ciprofloxacin	PLA Aged PLA PVC Aged PVC			50	390 690 340 440	910 1,190 610 770		
Sulfamethoxazole **Propranolol** **Sertraline**	PE	0.06 (pH = 6.85)	0.2	96	45.37 60.37 81.58	46.09 64.38 88.8	-	(Razanajatovo et al., 2018)
17β-Estradiol **17α-Ethynylestradiol**	PVC	0.01	0.05	48	-	33.56 40.16	-	(Lu et al., 2020)
Tonalide **Musk xylene** **Musk ketone**	PP	0.005	2	10	1.4 1.3 1.8	-	-	(Zhang et al., 2018b)
Triclosan	PS-60 PS-100 PS-150 PS-200	5.0	2.5	72	370 480 420 390	1000 890 780 720	-	(Li et al., 2019b)
Triclosan	PVC-small PVC-large	10	0.4	24	11,700 8,100	12,700 8,980	Elovich Webber–Morris	(Ma et al., 2019)
Triclosan	PE PS	5.8	4	0.5 96	299.1 292.3	1247.8 1033.24	Intra particle diffusion	(Chen et al., 2021)

*Microplastics from the coastal beach of North China; **microplastics from the Coast of East China Sea and Yellow Sea.
Note: pharmaceutical and personal care product (PPCP), polyethylene (PE), polypropylene (PP), polystyrene (PS), polyvinyl chloride (PVC), polyamide (PA), and polyethylene terephthalate (PET), Polylactic acid (PLA).

Table 2
Summary of available literature data on isotherm studies of PPCP sorption by microplastics.

PPCP contaminant	Microplastic type	PPCP concentration range ($mg\ L^{-1}$) and pH, salinity and DOM condition	Allowed time to reach equilibrium (h)	PPCP sorption capacity (Q_{max})/coefficient derived (K) from the best-fitting isotherm models			Reference
				Langmuir Q_{max} ($mg\ g^{-1}$)	Linear K_d ($L\ kg^{-1}$)	Freundlich K_f ($(mg^{1-N}\ kg^{-1}\ L^N)$)	
Oxytetracycline	PS	2–50	54	1.52	41.7	425	(Zhang et al., 2018a)
	Aged PS*	2–50		27.5	428	894[c]	
		2–50 (HA 10 $mg\ L^{-1}$)		-	-	2,420	
		2–50 (HA 100 $mg\ L^{-1}$)		-	-	3,940	
		2–50 (FA 10 $mg\ L^{-1}$)		-	-	1,580	
		2–50 (FA 100 $mg\ L^{-1}$)		-	-	1,980	
Ciprofloxacin	PS	2–25	48	10.2	210	580[c]	(Liu et al., 2019b)
	UV aged PS			54.8	318	540	
	PVC			11.7	215	550	
	UV aged PVC			15.5	251	740	
Ciprofloxacin	PE	30-500 (pH 6.5-7.5)	3	5.85	-	-	(Atugoda et al., 2020
		30-500 (pH 6.5-7.5, HA 2.5 $mg\ L^{-1}$)		4.52			
Tylosin	PE	1–30	36	1.67	62.75	131.71	(Guo et al., 2018)
	PP			3.33	94.13	183.46	
Tetracycline	PS	0.2–5 (pH 6.8)	24	3.33	134.10	284.43	(Xu et al., 2018b)
	PVC			3.33	155.27	362.06	
	PE			0.109		85.9	
	PP			0.113	-	101	
	PS			0.167		138	
Tetracycline	Nano PS	0–10	48	44.77	-	9,180	(Wan et al., 2019)
	Nano PS (HA, pH 6)			50.99		13,700	
	Nano PS ($MgCl_2$, pH 6)			37.50		7,060	
Tetracycline	PE	1–35	36	-	67	231	(Yu et al., 2020)
	PS				58	55	
	PVC				55	16	
Ciprofloxacin	PP	0–15 (a) Freshwater (b) Seawater	96	(a) 0.615	(a) 57.1	(a) 252	
	PS			0.416	51.5	205	
	PVC			0.453	41.5	184	
	PE			0.200	55.1	222	
	PA			2.2	96.5	170	

(*continued on next page*)

Table 2 (*continued*)

Trimethoprim	PP			(a) 0.102	(a) 9.7 (b) 3.9	(a) 32.3	(Li et al., 2018a)
	PS			0.174	9.5 7.3	32.1	
	PVC			0.481	8.4 5.5	13.4	
	PE			0.154	8.9 6.5	22.0	
	PA			0.468	17.1 5.9	36.0	
Sulfadiazine	PP			-	(a) 7.9 (b) 7.1	(a) 8.0 (b) 6.5	
	PS				7.4 6.8	4.1 5.7	
	PVC				6.6 5.4	3.2 0.9	
	PE				6.2 6.3	2.2 3.0	
	PA				7.4 6.6	1.1 2.5	
Amoxicillin	PP			(a) 0.294	(a) 17.5	-	
	PVC			0.523	24.7		
	PE			0.131	8.4		
Tetracycline	PA			22.7	756.0		
	PA			(a) 3.84	(a) 356 (b) 4.4	(a)588 (b) 12.4	
Sulfamethoxazole	PE	0.2–5	24	-	591.7	665	(Xu et al., 2018a)
Sulfamethoxazole	PA	0.5–12	24	96.4	284.0	205.0[c]	(Guo et al., 2019a)
	PE			0.66	30.0	61.3	
	PET			0.71	29.7	51.5	
	PS			114.0	22.2	24.7	
	PVC			2.80	28.2	14.2	
	PP			6.9	30.9	5.84	
Sulfamethoxazole	Aged PS**	0.5–10 (a) Freshwater (b) Seawater	overnight	-	(a) 32.8	(a) 14.4 [c]	(Guo and Wang., 2019a)
	Aged PE**				-	-	
Sulfamethazine	Aged PS**			-	(a) 29.3	(a) 5.7 [c]	
	Aged PE**				38.3	110	
Cephalosporin-C	Aged PS **			(b) 0.71	(a) 23.6 (b)72.0	(a) 5 (b) 5,690 [c]	
	Aged PE**			0.72	37.0 56.7	100 1,280	
Ciprofloxacin	PLA	0.5-10	48	3.19	-	210.6 [c]	(Fan et al., 2021)
	Aged PLA			3.77		251.8	
	PVC			0.67		457.2	
	Aged PVC			0.85		544.1	
Tetracycline	PLA			2.51	-	182.0	
	Aged PLA			5.49		285.5	
	PVC			0.96		600.3	
	Aged PVC			1.57		866.5	
Sulfamethoxazole **Propranolol**	PE	0.001–0.100 (pH 6.85)	96	-	700 2,300	3,090[c] 2,240	(Razanajatovo et al., 2018)

(*continued on next page*)

Table 2 (*continued*)

Sertraline					3,330	4,360	
Ibuprofen	AMWPE	0.05–0.35	120	-	(a) 8.8 (b) 13.3	-	(Elizalde-Velázquez et al., 2020)
	UHMWPE				7.5 16.0		
	PS				5.5 7.8		
	PP				1.7 2.5		
Diclofenac	AMWPE	(a) Synthetic seawater (pH 8.1) (b) Freshwater (pH 6.9)			(a) 2.5 (b) 26.2	-	
	UHMWPE				6.9 32.3		
	PS				2.9 27.9		
	PP				1.6 12.3		
Naproxen	AMWPE				(a) 0.6 (b) 4.3	-	
	UHMWPE				1.5 4.5		
	PS				1.6 2.4		
	PP				0.7 1.9		
Carbamazepine	PE	0.010–0.20	120	-	191	-	(Wu et al., 2016)
4-Methylbenzylidene camphor					312		
Triclosan					5,140		
17α-Ethinylestradiol					53,225		
17α-Ethynylestradiol	PVC	0.005–0.1	-	1.65	-	-	(Lu et al., 2020)
Triclosan	PP	0.1–10	-	-	-	2,090[c]	(Wu et al., 2020)
	12 days aged PP					2,910	
	20 days aged PP					4,310	
	40 days aged PP					5,930	
Triclosan	PS-100	1–12 (15 °C)	24	-	150	-	(Li et al., 2019b)
		1–12 (25 °C)			160		
		1–12 (35 °C)			180		
		1–12 (45 °C)			170		
Triclosan	PVC-small	2–20	24	20,400[γ] 42,100	1,350	-	(Ma et al., 2019)
	PVC-large			17,100[γ] 40,200	1,050		
Triclosan	PE	1.8-11	96	6.11	3810[α]	910[c]	(Chen et al., 2021)
	PS			-	1030[α]	140	

*Microplastics from the coastal beach of North China; **microplastics from the Coast of East China Sea and Yellow Sea.
Note: pharmaceutical and personal care product (PPCP), polyethylene (PE), polypropylene (PP), polystyrene (PS), polyvinyl chloride (PVC), polyamide (PA), polyethylene terephthalate (PET), poly(methyl methacrylate) (PMMA), average-molecular weight medium-density polyethylene (AMWPE), medium-density polyethylene (MDPE), low-density polyethylene (LDPE), linear low-density polyethylene (LLDPE), high-density polyethylene (HDPE), ultra-high-molecular weight polyethylene (UHMWPE), and medium-density polyethylene (MDPE), Polylactic acid (PLA).
[γ]Dubinin–Radushkevich model, [α] Temkin model.

respectively. Recently, the isotherm behaviors of ciprofloxacin, trimethoprim, sulfadiazine, amoxicillin, tetracycline, sulfamethoxazole, sulfamethazine, and cephalosporin in freshwater and seawater were also studied (Guo and Wang, 2019b; Li et al., 2018a). To achieve maximum sorption, the authors allowed the equilibration to occur for > 1 day. The best-fitting isotherm models for microplastic-antibiotic interactions were found to be linear (Guo et al., 2019a; Guo and Wang, 2019a; Xu et al., 2018a; Yu et al., 2020), Langmuir (Guo et al., 2018; Liu et al., 2019b; Wan et al., 2019; Xu et al., 2018b), and Freundlich (Guo et al., 2018; Liu et al., 2019b; Razanajatovo et al., 2018; Xu et al., 2018a) models, which best described the majority of cases. Guo and Wang (2019a) and Zhang et al. (2018a) studied the effect of initial antibiotic concentrations on sorption behavior of naturally beached microplastics using microplastics from field samples. Zhang et al. (2018a) found that the K_f of oxytetracycline (894 mg^{1-N} kg^{-1} L^N) for beached PS samples was two-fold greater than that for pristine PS. Liu et al. (2019b) studied the sorption isotherm of antibiotics for artificially aged microplastics. The maximum ciprofloxacin sorption capacity increased from 10.2 and 11.7 mg g^{-1} for pristine PS and PVC to 54.8 and 15.5 mg g^{-1} for aged PS and PVC, respectively (Liu et al., 2019b). Research on the sorption capacities of aged and field microplastics and microplastics made of polar polymers (PA and polyacrylics) as a function of antibiotic concentration

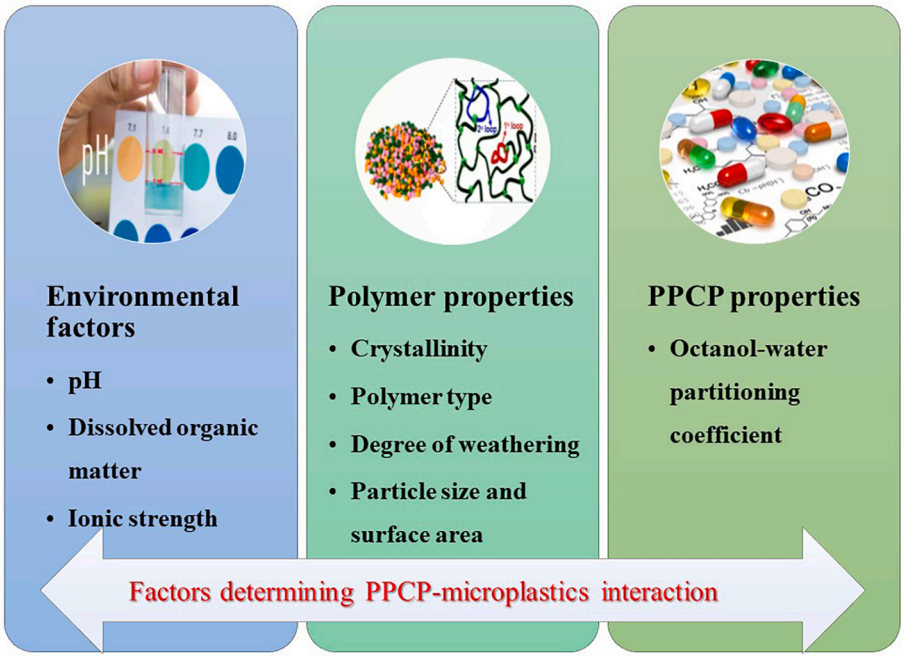

Fig. 2. Factors affecting the sorption of contaminants by microplastics.

are either limited or nonexistent.

2.2.2. *β-Blockers, antidepressants, and anticonvulsants*

The sorption isotherms of propranolol (β-blocker) and sertraline (antidepressant) on PE microplastics were investigated by Razanajatovo et al. (2018) with initial concentrations < 1 mg L^{-1}. The maximum sorption coefficients (K_d) of propranolol and sertraline were 2,300 and 3,300 L kg^{-1}, respectively, as derived by the linear isotherm model. Similarly, Wu et al. (2016) studied the sorption of carbamazepine (anticonvulsant) on PE for solution concentrations in the range of μg L^{-1}. However, the partitioning of carbamazepine to PE microplastics was not as strong as what was observed for propranolol and sertraline.

2.2.3. *Non-steroidal anti-inflammatory drugs*

The effect of initial concentration was examined for several NSAIDs by Elizalde-Velázquez et al. (2020). The sorption of ibuprofen, diclofenac, and naproxen on PP, PS, average-molecular weight PE (AMWPE), and ultra-high-molecular weight (UHMWPE) microplastics was investigated in seawater and freshwater with an initial concentration range of < 1 mg L^{-1}, which is environmentally relevant. The partitioning was significantly stronger in freshwater than that in seawater due to the high salinity and DOM content of seawater. In seawater, ibuprofen, diclofenac, and naproxen exhibited the highest partitioning to AMWPE, UHMWPE, and PS. However, in freshwater, ibuprofen and naproxen exhibited the highest partitioning to UHMWPE with K_d values of 16.0 and 4.5 L kg^{-1}, respectively, while diclofenac exhibited the highest partitioning to AMWPE with a K_d value of 32.3 L kg^{-1}, as derived from linear modeling. Although NSAIDs are a widely used class of drugs, the assessment of their interaction with microplastics was limited to only ibuprofen, diclofenac, and naproxen. In addition, the effects of environmental conditions and sorption affinities of aged microplastic samples remain unknown for this class of compounds.

2.2.4. *Steroidal hormones*

Steroidal hormones are one of the pharmaceutical classes whose microplastic sorption has not yet been thoroughly investigated. Two steroidal hormones, 17β-estradiol and 17α-ethynylestradiol, with initial concentrations ranging from ng L^{-1} to μg L^{-1}, were studied by Liu et al. (2019c), Lu et al. (2020), and Wu et al. (2016). Liu et al. (2019c) examined the sorption of 17β-estradiol on a wide range of plastic types, i.e., linear low-density polyethylene (LLDPE), UHMWPE, low-density polyethylene (LDPE), high-density polyethylene (HDPE), medium-density polyethylene (MDPE), PP, poly(methyl methacrylate) (PMMA), PVC, PA, and PS, which was best fitted by the Freundlich model. For 17α-ethynylestradiol, Lu et al. (2020) derived a maximum sorption capacity of 1.65 mg g^{-1} for sorption on PVC by the Langmuir model, whereas Wu et al. (2016) derived a K_d value of 5,141 L kg^{-1} for sorption on PE by the linear model.

2.2.5. *Antimicrobials and ultraviolet screening agents*

Among the compounds used in personal care products, the sorption behaviors of triclosan (antimicrobial agent) and 4-methylbenzylidene camphor (a ultraviolet light blocker used in sunscreens) have been exclusively studied (Li et al., 2019b; Wu et al., 2020). For example, Wu et al. (2020) studied the sorption of triclosan on aged PS (over 40 days) and found that with aging, the sorption capacity of PS for triclosan increased with a high K_f value of 5,930 mg^{1-N} kg^{-1} L^{N}. Li et al. (2019b) investigated sorption as a function of temperature and found that sorption increased with temperature. In another study, Wu et al. (2016b) investigated the sorption of both triclosan and 4-methylbenzylidene camphor on PE and reported a relatively low K_d value of 312 L kg^{-1} for 4-methylbenzylidene camphor and a relatively high K_d value of 5,140 L kg^{-1} for triclosan. The initial concentrations used by Wu et al. (2016) were below 1 mg L^{-1}, whereas Wu et al. (2020) and Li et al. (2019) conducted their studies using a concentration range of 1–20 mg L^{-1}. The studies based on the sorption of personal care compounds especially regarding the type of microplastics and environmental conditions are limited. Since microplastics are incorporated into these personal care products, their uptake at relevant concentration levels should also be considered.

3. Environmental factors affecting sorption

3.1. *Solution pH*

Solution pH could alter the biochemical and physicochemical properties of a compound, which consequently affects its chemical reactivity, equilibrium condition, and toxicity (Marion et al., 2011). Approximately

Table 3
Summary of PPCP sorption capacities of microplastics under different conditions.

PPCP contaminant	Microplastic type	PPCP initial concentration (mg L^{-1})	Dosage (g L^{-1})	pH	pH impacted PPCP sorption capacity (μg g^{-1})	Salinity/Ionic strength of water source	Salinity impacted PPCP sorption capacity (μg g^{-1})	DOM	DOM impacted sorption capacity (μg g^{-1})	Reference
Oxytetracycline	PS	20	1.66	5	2,000	$CaCl_2$ (0.016–0.15) NaCl (0.016–0.15) Na_2SO_4 (0.016–0.15)	2,150-1,342 840-495 1,445-480	-	-	(Zhang et al., 2018a)
	PS aged*				6,000	$CaCl_2$ (0.016–0.15) NaCl (0.016–0.15) Na_2SO_4 (0.016–0.15)	2,368-1,850 2,340-1,719 2,077-1,742	-	-	
Ciprofloxacin	PS UV aged PS PVC UV aged PVC	10	0.4	5 8 9 9	5,725 5,063 4,805 5,255	NaCl 35%	1,583 1,168 383 340	-	-	(Liu et al., 2019b)
Ciprofloxacin	PE	10	2.0	6.5-7.5	2,900	NaCl 0.001M NaCl 0.1M	2,620 2,340	HA 0.5 mg L^{-1} 1.5 mg L^{-1} 2.5 mg L^{-1}	1,100 1,000 670	(Atugoda et al., 2020)
Tetracycline	PE PP PS	1	5	6	81.70 104.70 140.00	NaCl 2% (pH 6.8)	61 57 84	FA 1–20 mg L^{-1} (pH 6.8)	100-2.7 96.2-4.4 102-7.3	(Xu et al., 2018b)
Tetracycline	Nano PS	10	0.2	6	3,336	$MgCl_2$ 10 mmol L^{-1} (pH 6)	2,406	HA 10 mg L^{-1} pH 6	3,697	(Wan et al., 2019)
Tetracycline	Pristine PE Aged PE (HA) Aged PE (pH 10) Aged PE (pH 4)	10	10	7 7 10 4	91.7 120.5 114.9 111.1	-	-	HA 2 mg L^{-1}	120.5	(Shen et al., 2018)
Sulfamethoxazole	PA PE PET PS PVC PP	2.4	2	3	327 150 178 200 181 176	Salinity 35%	67 39 45 30 47 31	-	-	(Guo et al., 2019a)
Ciprofloxacin	PVC LDPE HDPE PP	100	0.5	2-4 12 12 12	19.44[α] 22.92 13.23 12.57	-	-	-	-	(Puckowski et al., 2021)
Propranolol	PP			12	10.3 [α]					
17β-estradiol **17α-ethynylestradiol**	PVC	0.010	0.05	8	40.36 44.30	Salinity 32 g L^{-1}	33.56 40.16	HA 20 mg L^{-1}	80.96 87.94	(Lu et al., 2020)
Triclosan	PS-100	5	2.5	3–6	900	NaCl (0.001–0.5 M)	900	-	-	(Li et al., 2019b)

*Microplastics from the coastal beach of North China; [α]K_d values (L kg^{-1}).
Note: pharmaceutical and personal care product (PPCP), polyethylene (PE), polypropylene (PP), polystyrene (PS), polyvinyl chloride (PVC), polyamide (PA), polyacrylate (PAC), polyethylene terephthalate (PET).

85%–95% of active pharmaceutical ingredients are ionizable compounds that are present in the form of weak acids or weak bases (Karlsson et al., 2017). These compounds contain multiple functional groups and dissociate in the water matrix, generating different ionic species depending on pH levels; therefore, their sorption behavior varies with changing pH (Fig. 2). Although the pH of most natural water sources range from 6 to 9 (Boyd, 2020), water sources can have extreme pH conditions ranging from 2 to 10. This broadens the scope of environmental conditions that govern the fate and behavior of PPCP contaminants (Bundschuh et al., 2016; Karlsson et al., 2017). Furthermore, surface charge of microplastics varies significantly and is dependent on the pH of the solution. The point of zero charge (pH_{PZC}) of microplastics is generally less than pH 7; therefore, at environmentally relevant pH and above, the surface is negatively charged. Xu et al. (2018a) studied

the sorption of tetracycline on microplastics and reported that the pH_{PZC} of PE, PP, and PS microplastics were 4.30, 4.26, and 3.96, respectively. Tetracycline dissociates into cations at low pHs, anions at high pHs, and zwitterions in the pH range of 5–7. The electrostatic attractions between zwitterionic tetracycline and microplastic were the strongest; thus, the maximum sorption capacity was observed at a pH level of 6 (Table 3). With an increase in pH, sorption capacity decreased as both tetracycline and microplastics became increasingly negatively charged, which resulted in a repulsive interaction. The findings of Wan et al. (2019) support the study on the sorption of tetracycline on PS nanoparticles, which reported high and low adsorption at neutral and extreme pH, respectively. The antibiotic sulfamethoxazole exhibited a high adsorption by all types of microplastics in acidic pH (Guo et al., 2019a). In the pH range of 3–6, sulfamethoxazole is mainly neutral and exhibits hydrophobic interactions with the non-polar plastic surface. In contrast, at $pH > 6$, sulfamethoxazole becomes anionic and the electronegativity of microplastics increases; therefore, adsorption is suppressed due to repulsive interaction. A similar behavior has been observed for two other types of antibiotics, i.e., sulfamethazine (pK_{a1}: 2.28; pK_{a2}: 7.42) (Guo et al., 2019b) and tylosin (pK_b: 7.1) (Guo et al., 2018), which show high adsorption in a low pH environment. Electrostatic and hydrophobic attractions played a significant role in the massive uptake of antibiotics as they became positively charged in acidic pH, whereas electrostatic interaction became less significant at alkaline pH as the antibiotics became negatively charged. While the pH-dependent sorption of antibiotics has been extensively studied, there is a paucity of data on personal care products. The sorption behavior of triclosan was studied by Li et al. (2019) and Ma et al. (2019) for PS and PVC microplastics, respectively. The adsorption of triclosan was maximum at $pH < pK_a$ (8.14) since triclosan remained mainly undissociated in this pH range. Once the pH exceeded pK_a, triclosan dissociated into anions, which resulted in the weakening of the hydrophobic forces, while the electrostatic repulsions became more significant and the adsorption by PS and PVC decreased (Behera et al., 2010; Ma et al., 2019). However, contrary to these findings, Xu et al. (2018a) observed that pH only played a minor role in the sorption dynamics of sulfamethoxazole by PE microplastics. The pH-independent sorption behavior has also been demonstrated for non-ionic organic molecules, such as perfluorooctanesulfonamide (FOSA) (Wang et al., 2015) and phthalate (Liu et al., 2019a) compounds for PE, PS, and PVC microplastics. Such a behavior, especially in the case of non-ionic organic molecules, can be explained by the combined effects of hydrophobic interactions and van der Waals forces rather than electrostatic interactions (Llorca et al., 2018; Zhang et al., 2020).

Generally, PPCP compounds display pH-dependent sorption on different types of microplastics. Based on the research-derived data on the role of pH in microplastic interaction with PPCPs, the following conclusions can be drawn: (a) at low pH, PPCP compounds form cationic species, which enhances their electrostatic attraction with the microplastic surface, and thus, sorption affinity; (b) zwitterionic or neutral PPCP species interact with non-polar plastic surfaces through hydrophobic and van der Waals forces, which promotes sorption (c) at a high pH, PPCP compounds form anionic species, which enhances their electrostatic repulsive interaction with the plastic surface, resulting in a decrease in their sorption.

3.2. Ionic strength and salinity

The effect of ionic strength on contaminant sorption dynamics has been studied by simulating the ionic strength of natural seawater by dissolving salts such as NaCl, $CaCl_2$, $MgCl_2$, and Na_2SO_4 in water, or by using synthetic seawater or natural seawater. The ciprofloxacin adsorption efficiency of PS microplastics decreased with the addition of NaCl; the efficiency further decreased when the NaCl concentration was increased from 8.8% to 35%, while the sorption on PVC decreased steadily (Liu et al., 2019b) (Table 3). Similar trends were observed for the sorption of sulfamethoxazole (Guo et al., 2019a) and sulfamethazine (Guo et al., 2019b) on all types of microplastics (PE, PS, PA, PCV, PP, PET) using 10%, 20%, and 35% NaCl concentrations. It has been demonstrated that the sorption capacity of antibiotics in seawater is lower than their sorption capacity in freshwater (Elizalde-Velázquez et al., 2020; Li et al., 2018a). Since microplastic particle surfaces are negatively charged, the cations (e.g., Na^+ and Ca^{2+}) in the medium can bind electrostatically to the binding sites, thereby disturbing the charge equilibrium of the surface. The dissolution of salt increases the viscosity and density of the matrix, hindering the mass transfer of molecules to the surface, whereas ionic species easily access the surface binding sites than the molecular species; thus, the electrolytes compete with organic molecules for the adsorption sites (Guo et al., 2019b; Wu et al., 2018).

Ionic strength likely affects the aggregation of microplastics, especially in the case of nanoplastics (Fig. 2). Wan et al. (2019) reported that tetracycline sorption was drastically inhibited in the presence of $MgCl_2$ due to the severe aggregation of PS nanospheres. The increase in ionic strength promotes the aggregation of PS nanoplastics through compression or elimination of the electrical double layer on the surface, thereby reducing the repulsive forces between the particles (Wu et al., 2019). Divalent electrolytes accelerate the aggregation of microplastics more than monovalent electrolytes (Li et al., 2018b). Therefore, the active sites on the plastic surface become inaccessible to the PPCP compound molecules, which in turn decreases the overall sorption (Liu et al., 2019a).

Interestingly, in some cases, a high ionic strength is known to promote the sorption of PPCP compounds. Ma et al. (2019) reported that with an increase of 35% in salinity (NaCl), the sorption amount of triclosan increased by 43.8% and 73.4% for small and large PVC particles, respectively. Wu et al. (2016) also reported that the sorption of triclosan by PE debris increased with an increase in salinity. Similarly, the sorption of 17β-estradiol and 17α-ethynylestradiol (steroid hormones) increased three-fold when salinity of seawater was doubled (Lu et al., 2020). The sorption enhancement by ionic species can be mainly attributed to the salting-out effect and ionic complexation (Ma et al., 2019; Soubaneh et al., 2014).

The salting-out effect affects the sorption dynamics of organic compounds in the presence of ions. This phenomenon occurs when the solubility of a non-electrolyte compound decreases with the increase of salt concentration (Poole, 2020). Inorganic salts can shift the sorption equilibrium of organic compounds toward the organic phase by decreasing the solubility of non-polar and weakly polar organic compounds in the aqueous medium (Alimi et al., 2018). The effect of salt on partition coefficients of a compound can be determined by Setschenow constants, which showed that the solubility of pharmaceuticals decreases with increasing NaCl concentrations (Liu et al., 2014). Additionally, higher salinity improved the adsorption capacity of HOCs by clay, soil, and sediments. Therefore, the salting-out effect enhances the hydrophobic interactions of organic contaminants with microplastics (Liu et al., 2019a). The presence of ions also promotes cationic bridging, especially with multivalent ions. Multivalent ions facilitate the formation of ternary complexes through the bonding of the pharmaceuticals with the functional groups of the adsorbent, which enhances the sorption properties (MacKay and Canterbury, 2005). Zhang et al. (2018a) reported that the presence of Ca^{2+} facilitated sorption by promoting surface complexation between an adsorbent and adsorbate. Moreover, the partitioning of cephalosporin-C in simulated seawater increased with a decrease in solubility and complexation with the adsorbent surface (Guo and Wang, 2019a).

However, some studies have reported that ionic strength has a negligible effect on the adsorption of some PPCP compounds on microplastics. Xu et al. (2018a, 2018b) observed that for a NaCl concentration range of 0.05%–3.5%, the difference in the sorption's of tetracycline and sulfamethoxazole was insignificant; this implied that electrostatic interactions did not play a significant role in the sorption mechanism. The effect of NaCl (0.001–0.1 M) was negligible for

triclosan as sorption occurred predominantly via hydrophobic interactions with uncharged molecular species, and electrolytes played a negligible role in this case (Li et al., 2019b).

The solution pH influences the effect of salinity. The bridging effect of Ca^{2+} was evident for the ionic form of 4-butyric acid at pH > 7 with a high partitioning to PE and decreased at a low pH in its non-ionic form (Hüffer et al., 2019). A similar mechanism was observed in soil at a low pH, where the preferential binding of Cd^{2+} occurred due to a change in the surface charge of soil, which promoted organic ligand binding at a low pH (Bolan et al., 1999). Moreover, the sorption of tetracycline decreased at pH > 5 due to the severe aggregation of PS nanospheres (Wan et al., 2019). These studies indicate that the effect of ionic strength on the sorption of contaminants may vary depending on the type of adsorbent, adsorbate, electrolyte, and solution chemistry.

3.3. Dissolved organic matter

DOM (such as FA and HA) can either inhibit or enhance sorption of organic molecules on the microplastic surface depending on adsorbent and adsorbate properties (Bolan et al., 2011). The sorption of tetracycline on PS, PP, and PE drastically decreased by 93%, 95%, and 97%, respectively, when the FA concentration was increased to 20 mg L^{-1} (Xu et al., 2018b) (Table 3). Wu et al. (2016) also reported a decrease in the sorption capacity of PE microplastics for PPCP compounds, 17 α-ethinylestradiol, 4-methylbenzylidene camphor, and triclosan, with an increase in HA concentration (0–20 mg L^{-1}); however, the effect of HA concentration on the highly polar carbamazepine was the least. The presence of DOM possibly decreases the bioavailability of hydrophobic chemicals through complexation with the hydrophobic HA parts, which changes the partitioning between the solid surface and water (Burkhard, 2000; Seidensticker et al., 2017a). However, in the case of PPCP compounds, both the hydrophobic and hydrophilic compartments of HA may undergo complexation with heterogenic functional groups (carboxyl and amino groups) of the PPCP compounds (Aristilde and Sposito, 2010; Gu et al., 2007). Liu et al. (2017) reported more significant partitioning of sulfamethoxazole and ciprofloxacin on HA due to hydrogen bonding. Antibiotics may undergo preferential complexation with HA, which would reduce their sorption on microplastics. Antibiotics can also interact with inorganic metal cations and humic substances in the environment to form stable ternary complexes (HA–metal–antibiotic), as suggested by the molecular dynamics studies based on the interaction of ciprofloxacin (Aristilde and Sposito, 2010), tetracycline (Gu et al., 2007), and oxytetracycline (MacKay and Canterbury, 2005) with humic substances. Such complexation can reduce the surface mobility of antibiotics, thereby reducing the possibility of sorption by microplastics.

As the sorption of organics preferentially occurs in the amorphous region of polymers and is confined to some regions of the polymer. HA and organic pollutants compete for the limited sorption sites on the microplastic surface (Teuten et al., 2009). Humic molecules may replace the organic pollutants occupying the sorption sites by desorbing the organic pollutants from microplastics (Daugherty, 2016). Since DOMs are generally large molecules, they can block the pores on the microplastic surface, thereby preventing further entry of organic pollutants (Velzeboer et al., 2014).

To date, only a few studies have investigated the promotion of the sorption process by DOM. A high HA concentration (0–20 mg L^{-1}) was found to enhance the sorption of hormonal steroid compounds by microplastics, which was attributed to HA-microplastic complexation (Lu et al., 2020). Additionally, the sorption of oxytetracycline on beached PS foams increased with increasing FA concentration. The carbonyl functional groups of DOM can interact with the aromatic structure of PS through π–π conjugation to form a copolymer, DOM-microplastic, having a high electron density (Chen et al., 2018). Thus, the positively charged molecular species bind to this copolymer through electrostatic attraction, resulting in improved sorption. Besides, FA was found to have a negligible effect on sulfamethoxazole sorption, possibly due to the higher affinity of the antibiotic toward microplastics (Xu et al., 2018a).

In contrast, HA may adsorb onto a microplastic surface and increase the hydrophobicity, enhancing sorption, or occupy the limited sorption sites available to the organic pollutant molecules (Daugherty, 2016). However, some studies have shown that the interaction between HA and microplastics is negligible, possibly because of the hydrophilic properties of HA (Wu et al., 2016). This was further confirmed by Seidensticker et al. (2017) using fluorescence measurements of HA and PE (Seidensticker et al., 2017b). The coexistence of microplastics and DOM over prolonged periods in the environment can induce structural changes in plastic (e.g., formation of biofilm and aggregation), thereby altering the surface morphology and buoyancy of microplastics and hence the fate and transport of the hydrophobic contaminants (Chen et al., 2018; Wang et al., 2020e). In contrast, some studies have shown the independent behavior of DOM with respect to microplastic and PPCP interactions. For example, Xu et al. (2018b) found that DOM does not affect the sorption of sulfamethoxazole by PE microplastics, and similar observations were reported by Liu et al. (2019a) for sorption of phthalate compounds on PA microplastics. In this case, it can be assumed that microplastics have a high affinity for PPCPs than for DOM. Therefore, the overall effect of DOM is the cumulative balance of all these interactions and factors, which are often governed by solution chemistry.

3.4. Other competing sorbents

The natural environmental matrices are heterogenous and quite complicated in composition. Therefore, partitioning of molecules merely onto microplastics can be considered unless otherwise other naturally or artificially occurring sorbents are present. Sorbents can arise from both inorganic and organic materials such as activated carbon particles, engineered nano materials, suspended natural clay and mineral oxides that could interfere with microplastic partitioning in soil and water. According to Koelmans et al. (2016) distribution of the HOC over other media is a crucial factor to determine the relative importance of microplastics as a vector and correlated with the hydrophobicity of the phase. In the view of PPCP since they are ionizable compounds pH and ionic strength plays key roles for adoption on natural organic sorbents (Kah et al., 2017). Many studies have evaluated various carbonaceous materials such as activated carbon (Mailler et al., 2015), carbon nanotubes (Jung et al., 2015; Wang et al., 2017), graphene (Liu et al., 2014; Rostamian and Behnejad, 2016) and biochar (Ahmad et al., 2014) for their performance and efficiency in PPCP removal in wastewater. Also, PPCPs have shown greater partitioning to sediments, clay and mineral oxides especially in soil (Figueroa and MacKay, 2005; Martínez-Hernández et al., 2014; Xu et al., 2020). However, studies on PPCP partitioning to microplastics with regard to other co-existing sorbents have been over looked rarely. Microplastics can as well alter sorption onto other sorbents vice versa. In the study of Liu et al. (2020a) increased microplastics ratio accelerated sorption equilibrium of 17β-estradiol by magnetic biochar. Also, in another study by Li et al. (2020) microplastics reduced the sorption of oxytetracycline in sandy loamy soil enhancing bioavailability and mobility. Therefore, taking into account of partitioning onto other competitive sorbents including microplastics is important in assessing the interactions as well as the desorption in a particular environment (Hartmann et al., 2017; Koelmans et al., 2016). In terms of HOC, microplastics have been found to have negligible impact in comparison to natural exposure pathways, but may have specific scenarios for hydrophilic compounds such as PPCPs .

3.5. Biofouling

Biofouling is a form of biotic mediated degradation process that occurs on the microplastics surface which alters morphology and physicochemical properties of the particle. The process takes place when

microbial cells physically adhere on the microplastic surface through extracellular polymeric substances (EPS) to colonize on the surface (Glaser, 2020). As a result, microplastic becomes a particle with a polymer core and an outer shell organic layer, hence any molecule in the ambient aqueous media has to surpass the organic barrier. The overall kinetics is dependent on the rate of mass transfer from water into the biofilm layer and then to the polymer material (Endo and Koelmans, 2019). By this way biofilm layer can slowdown the mass transfer process acting as a barrier for the molecule migration (Liu et al., 2020c). Organic contaminants interact via hydrophobic partitioning into the biopolymer or through binding onto the sorption sites of the heterogenic EPS (Writer et al., 2011). The produced EPS by the biofilm comprise of ionizable functional groups such as carboxyl, phosphoryl, amino, and hydroxyl arising from the organic compounds (carbohydrate, protein and lipids) (Guan et al., 2020; Writer et al., 2011). The surface polarity induced by biofilms enhanced sorption capacities of metal ions (e.g. Cs, Sr, Co, Ni, Cu, Zn, Cd, Ag, Cr, Pb, Al, K, Mg, and U) by microplastic as reported by the previous studies (Guan et al., 2020; Johansen et al., 2018; Richard et al., 2019). The sorption of tetracycline was improved with biofilm formed microplastics according to the study by Wang et al. (2020d) and the presence of Cu ions had synergistic effects on the sorption by forming antibiotic-Cu-EPS complex through ionic bridging effect. Biofilm development also increases the specific surface area to volume ratio of the microplastic to interact with organic molecules (Menéndez-Pedriza and Jaumot, 2020; Wang et al., 2020e). However, the relative effects of biofilm formation for microplastic-PPCP interaction, their fate and transport still remains vastly unexplored.

4. Polymer properties of microplastics affecting PPCP sorption

4.1. Crystallinity

The degree of crystallinity determines the hardness, density, and mechanical, thermal and diffusion properties of a polymer (Yang et al., 2018). Therefore, the permeability of gasses and liquids is restricted in densely arranged crystalline areas compared to amorphous regions. The mobility of hydrocarbon chains in polymer structures is restricted in the crystalline region, and thus, high amounts of energy are required to disrupt the polymer chain arrangement. In contrast, amorphous (rubbery) regions are flexible, with randomly oriented polymer chains (Karapanagioti and Werner, 2019; Teuten et al., 2009). Hence, the physical structure plays an important role in the interaction of organic contaminants with plastics (Velez et al., 2018). The rate of transfer of solute species through polymer matrix is governed by its solubility in the matrix, molecular size, and the interstices within the polymer (McKeen, 2017).

Since PPCP compounds are hydrophilic and polar, their sorption mechanism may differ from that of hydrophobic compounds. The sorption of sulfamethazine and sulfamethoxazole was less on PE, despite the amorphous nature of PE, whereas the sorption on glassy PA microplastics was maximum (Guo et al., 2019b, 2019a). Li et al. (2018a) reported that sorption for antibiotics on five different types of microplastics (PE, PP, PA, PS, and PVC) was not related to the degree of polymer crystallinity. The degree of crystallinity of the polymers decrease in the order of PE > PP > PA = PS > PVC. The sorption of all antibiotics was most favorable on crystalline PA, lowest on PVC, and intermediate on PE. Tetracycline displayed a similar behavior, exhibiting a high sorption on glassy PS than that on rubbery PE. Although PS is crystalline, the space between the adjacent polymeric benzene chains facilitate the diffusion of tetracycline (Xu et al., 2018b). Furthermore, the aging and weathering of microplastics destroyed the crystalline regions of the polymer, which resulted in a high uptake of triclosan by aged PS. X-ray diffraction analysis revealed a reduction in the crystallinity of PS after aging, which resulted in the formation of more rubbery domains for sorption (Wu et al., 2020). Thus, crystallinity alone is not a determining factor for contaminant sorption by microplastics, and other intrinsic factors of the polymer, such as the polarity, size, and weathering, play an important role in the sorption process.

4.2. Polymer types

Microplastics are derived from a wide variety of polymer resins having distinct physical and chemical characteristics. Therefore, the surface charge, functional groups, polarity, and molecular arrangement need to be studied comprehensively to understand the sorption process of organic contaminants. In general, aliphatic polymers containing monomers with C and H atoms are considered as non-polar. The strong C–C and C–H covalent bonds in PE impart a high structural strength and chemical resistance. The incorporation of different functional groups in the C, H backbone, or side chains enhances the chemical reactivity of the polymer, and therefore increases the water accessibility to the surface. Plastics such as PA, PS, PC, PVC, and PP comprising polar functional groups (CO–NH–, benzene, –COOH, –Cl, and $–CH_3$, respectively) are polar polymers and are thus capable of interacting with water and reactive compounds (Guo et al., 2019b; Liufei et al., 2019a).

The differences in the sorption of perfluorooctanesulfonate (PFOS) and FOSA by PE, PS, and PVC microplastics were ascribed to differences in polarity and polymer characteristics of the plastics (Wang et al., 2015). The polarity of polymers decreases in the order of PVC > PS > PE. Non-ionic FOSA, which is less polar, exhibited the highest partitioning (K_d) to PE, while polar PFOS exhibited the highest partitioning to PVC and the least partitioning for PS. The differences in the sorption behavior of PS and PVC and of PE can be attributed to the substitution of a hydrogen atom in the PE monomer by a chlorine atom in the case of PVC and a benzene molecule in the case of PS. However, the steric hindrance in PS creates less space, which restricts the sorption of molecules compared with PE and PVC (Wang et al., 2015). The sorption capacity of PA was the highest for sulfamethazine and sulfamethoxazole, whereas the sorption capacities of the other polymers did not correlate with the polarity order. Since pharmaceuticals such as antibiotics are hydrophilic compounds, they tend to interact more with polar polymers such as PA and less with non-polar polymers such as PE (Li et al., 2018a; Tourinho et al., 2019). Conversely, HOCs exhibit more affinity toward non-polar polymers and less affinity toward polar polymers; for instance, the partitioning of hydrophobic phenanthrene and pyrene to polar nylon and PVC was less than that to non-polar PE (Wang and Wang, 2018; Wang et al., 2018). Furthermore, non-polar polymers, such as HDPE, LDPE, and PP, accumulate greater amounts of hydrophobic contaminants than polar polymers, such as PET and PVC (Frias et al., 2010). Therefore, the surface polarity of the plastic determines the binding affinity of a particular compound, which is found to be lesser on hydrophobic surfaces than on hydrophilic surfaces for PPCP compounds.

4.3. Degree of weathering

The oxidation process during the weathering of microplastics yields oxygen-containing functional groups (carboxyl, hydroxyl, ketone, and ester) to the polymer structure introducing hydrophilic properties, which in turn affect the partitioning of HOCs (Barnes et al., 2009). Zhang et al. (2018a) studied oxytetracycline sorption on PS in the coastal beaches of North China and observed that the adsorption capacity of beached microplastics was twice of that of virgin PS pellets. Correspondently, ciprofloxacin (Liu et al., 2019b) and triclosan (Wang et al., 2015) exhibited the same trend for PS, PVC, and PP aged under laboratory conditions (UV and heat oxidation) with respect to pristine polymers. The presence of oxygen-containing bonds, as determined by Fourier transform infrared spectroscopic analysis, revealed that the extent of surface oxidation was significant (Liu et al., 2019b). In addition to the affinity of microplastics to water, the surface area of the particle can also increase due to weathering (Wang et al., 2020a). Hence, weathering and consequent aging processes can render the microplastic hydrophilic, thereby increasing the sorption of hydrophilic compounds.

Liu et al. (2020) conducted a desorption study on pristine and photo-Fenton-treated PS to investigate the release of pharmaceuticals atorvastatin and amlodipine and found that desorption was suppressed for treated PS. Aging decreased the hydrophobicity and increased the electrostatic interactions between the aged microplastics and pharmaceutical compounds, resulting in their retention on the polymer. Simultaneously, Hüffer et al. (2018) demonstrated the effect of UV-induced aging for 21 different HOCs. The results were contrary to those of PPCP compounds, with the sorption capacities decreasing by an order of magnitude for the analyzed HOCs on the aged microplastics compared with pristine microplastics due to a reduction in surface hydrophobicity.

4.4. Particle size and surface area

Although particle size plays a minor role than other physical parameters, it can affect the adsorption/desorption rate, rate of equilibrium establishment, and adsorption capacity (Hartmann et al., 2017; Tourinho et al., 2019). Polyethylene has the highest surface area followed by PS, PVC, and PP (Liu et al., 2019a; Wang and Wang, 2018). Elizalde-Velázquez et al. (2020) studied the sorption of NSAIDs on microplastics of several size ranges, PP (~1 mm), PS (600–800 μm), AMWPE (300–400 μm), and UHMWPE (2–10 μm), and found that the sorption capacities of all the particles followed the size trend despite the dissimilarity in polymer composition. A similar behavior was reported for PVC, wherein a high adsorption of triclosan was observed for the smaller particles (Ma et al., 2019). However, in some cases, a large surface area might not be the governing factor, as reported by Xu et al. (2018a) who observed the least sorption of tetracycline by PE.

The study on the adsorption of phenanthrene and nitrobenzene by Wang et al. (2019) provides an excellent example for particle aggregation with micron-, submicron-, and nano-sized PS microplastics (diameters of 170, 102, 50, and 30 μm and 800, 235, and 50 nm). They observed that with a decrease in PS size from 170 μm to 50 nm, *log* K_d gradually increased from 3.07 to 4.20 L kg^{-1} and then decreased from 4.20 to 4.07 L kg^{-1}, with the maximum sorption obtained for 235 nm size. The authors suggested that as the particle size decreased to the nanoscale, aggregation occurred, resulting in a decrease in sorption, which was evident from the lowest sorption obtained for 50 nm particles. Furthermore, the variation in particle size affects the time required to reach equilibrium. Non-aggregated small particles can increase the diffusion rate, thereby facilitating the faster establishment of steady-state equilibrium (Bakir et al., 2014b; Wang et al., 2019). For example, Hüffer et al. (2018) reported that milled PP particles reached equilibrium faster than pellets for difenoconazole. Therefore, particle size and surface area play an important role in the sorption of organic contaminants onto a variety of microplastics.

5. Chemical properties of PPCP influencing sorption: Octanol-water partition coefficient (Kow)

Sorbent properties are often related to hydrophobicity parameters in terms of K_{ow} or aqueous solubility (Eq. (1)). Compounds with high positive K_{ow} values are highly hydrophobic, whereas compounds with negative and low K_{ow} values are weakly hydrophobic or polar (Moldoveanu and David, 2015).

$$K_{ow} = \frac{[solute]_{octanol}}{[solute]_{water}} \quad (1)$$

Since polar compounds dissociate and exist as multiple species depending on the pH, the octanol-water distribution ratio (D_{ow}) of the species is considered to avoid variations in K_{ow} (Eq. (2)). However, this does not rely on hydrophobicity alone but also on the distribution of solute species in the two phases at a certain pH. In the absence of ionization, $K_{ow} = D_{ow}$, and as ionization occurs at any given pH, $K_{ow} > D_{ow}$. For amphoteric compounds such as antibiotics, when the neutral molecules are in their zwitterionic form, $D_{ow} < K_{ow}$ (Wegst-Uhrich et al., 2014). Based on K_{ow} values, contaminants are categorized as weakly hydrophobic or hydrophilic (log $K_{ow} < 3$), moderately hydrophobic ($4 \leq \log K_{ow} \geq 5$), and strongly hydrophobic (log $K_{ow} > 5$) (Gao et al., 2019; Kaur et al., 2018).

$$D_{ow} = \frac{[non\ ionized + ionized\ species]_{octanol}}{[non\ ionized + ionized\ species]_{water}}$$

PPCP compounds display a wide range of hydrophobicity; however, majority of compounds are generally moderately or weakly hydrophobic. Antibiotics, which are ubiquitous in the environment, generally exhibit log $K_{ow} < 1$ and are thus considered hydrophilic or polar compounds (Wang and Wang, 2016). Studies on the sorption capacities of contaminants on a single type of microplastic have shown correlation with the log K_{ow} value of the contaminants. For instance, Guo and Wang (2019a) observed that the sorption capacities of PS microplastics for three antibiotics are correlated with log K_{ow} values, and accordingly, they decrease in the order of sulfamethoxazole (log K_{ow} 0.89) > sulfamethazine (log K_{ow} 0.14) > cephalosporin-C (log K_{ow} 2.4). Similarly, ciprofloxacin, trimethoprim, sulfadiazine, amoxicillin, and tetracycline exhibited a positive correlation with log K_{ow} and their K_d values for PP, PE, PS, and PVC microplastics (Li et al., 2018a). In another study, the sorption of several PPCP compounds, carbamazepine, 4-methylbenzylidene camphor, triclosan, and 17α-ethinylestradiol on PE exhibited a similar trend (Wu et al., 2016). Furthermore, K_{ow} determines the desorption of PPCP compounds, with a high desorption rate observed for increased hydrophobicity, which likely increases the risk of bioaccumulation in the environment (Razanajatovo et al., 2018). Compared with neutral compounds, polar compounds exhibit a much lower affinity toward microplastics under environmentally relevant concentrations. However, in this case, the sorption behavior of PPCP compounds is not solely determined by hydrophobicity (log K_{ow}), and other factors might have to be considered (Seidensticker et al., 2018).

6. Mechanism of microplastic–PPCP compound interactions

Microplastics and PPCP compounds interact mainly through hydrophobic and electrostatic interactions, pore-filling mechanism, and π–π interactions, which are largely governed by the nature of polymer of the microplastic, chemical properties of the PPCP compound, and environmental conditions, such as the pH and ionic strength, of the media (Wang et al., 2020d).

6.1. Hydrophobic interactions

Hydrophobic interactions are non-covalent forces that cause the aggregation of non-polar moieties in a polar medium such as water (Thomas et al., 2014). The neutral molecules of PPCP compounds tend to bind to surfaces mainly through hydrophobic interactions (Wang et al., 2020d). Non-polar (PE and PP) and weakly polar (PS) plastics, which are homogenous and highly hydrophobic in nature, interact through non-polar or neutral molecules. Antibiotic compounds have been reported to show high partitioning to PE through hydrophobic attractions (Li et al., 2018a; Razanajatovo et al., 2018; Xu et al., 2018a; Yu et al., 2020). The zwitterions of PPCP compounds have positive and negative charges with a net zero charge resembling the molecular form. Therefore, zwitterions interact with neutral or non-polar adsorbent surfaces through hydrophobic interactions (Guo et al., 2019a; Wang et al., 2020d). Neutral triclosan molecules exhibit a high adsorption affinity to PS microplastics, mainly due to hydrophobic mechanisms (Li et al., 2019b). Similarly, tetracycline exhibit maximum adsorption on all types of non-polar microplastics in its zwitterionic state (Xu et al., 2018b).

For rubbery polymers, sorption occurs mainly through the diffusion

of contaminants into the highly flexible polymer bulk, referred to as partition or absorption (Teuten et al., 2009; Vinet and Zhedanov, 2011). The plastic-water partitioning coefficient (log K_{pw}) and log K_{ow} are crucial for understanding the sorption and desorption behavior of a chemical on plastics. There is a linear correlation between log K_{pw} and log K_{ow} of organic contaminants for a wide variety of microplastics, which suggests partitioning into plastics driven by hydrophobic interactions, similar to partitioning into animal lipid (Rist and Hartmann, 2018). PE, a semi-crystalline polymer, consists of loosely arranged rubbery segments that facilitate the penetration of contaminants into the porous polymer bulk. In these cases, the sorption through hydrophobic interaction is related to log K_{ow} of the PPCP compounds. The strength of hydrophobic interactions significantly depends on the chemical nature of the contaminants and the morphological properties of the surface of the plastic. Generally, the chemical and physical properties of the adsorbent and adsorbate are often affected by the solution chemistry (e.g., pH, salinity, and DOM).

6.2. Electrostatic interactions

Electrostatic interactions occur between positively and negatively charged solute and sorbent moieties (Bolan et al., 1999). These kinds of interactions are dominant when ionizable functional groups are present in the adsorbent and adsorbate. PPCP compounds contain ionizable functional groups and exist as different species with anionic and cationic charges. The pH_{PZC} of plastics indicate that the surfaces of most microplastics have a net negative charge under environmental conditions (Tourinho et al., 2019). Therefore, the positively charged domains of the hydrophilic molecules of solutes interact with the plastic surface through electrostatic attraction, whereas the negatively charged domains repel the plastic surface (Fig. 3). Therefore, with an increase in pH, the adsorption affinity of antibiotics to microplastics decreases as they transit from positively charged to negatively charged species with increasing pH (Li et al., 2018a). Generally, for hydrophilic compounds, pH-assisted adsorption is mainly governed by electrostatic interactions. The presence of polar groups on the adsorbent surface increases the electrostatic attraction between the hydrophilic domains of the contaminant molecules.

PPCP compounds with ionizable polar groups display higher affinity to polar polymers, such as PA and PVC polymer compared to non-polar polymers, such as PE polymer. The polar functional groups of the polymer can induce dipole–dipole or dipole-induced dipole interactions with the functional groups of the polar groups of PPCP compounds through electrostatic interactions (Kotdawala et al., 2005). Electrostatic interactions such as hydrogen bonds and van der Waals forces are weaker than covalent and ionic bonds that promote molecular binding to microplastics (Wang et al., 2020d). Hydrogen bonds are also considered as dipole–dipole interactions that exist between a hydrogen atom bonded to a strongly electronegative atom and a lone pair of electrons of another electronegative atom in the vicinity. Pharmaceuticals and hormones are regarded as H-bond donor compounds and interact with H-accepting surfaces such as PA and PAC (Endo et al., 2011). There is sufficient evidence that suggest the significant sorption of PPCP compounds on PA microplastics through hydrogen bonding (Liu et al., 2019c). Antibiotics such as ciprofloxacin, amoxicillin, tetracycline, and sulfamethoxazole, and steroidal hormones such as 17β-estradiol strongly interact with PA due to hydrogen bonding (Guo et al., 2019a; Li et al., 2018a; Liu et al., 2019c). Particularly, the weathering of

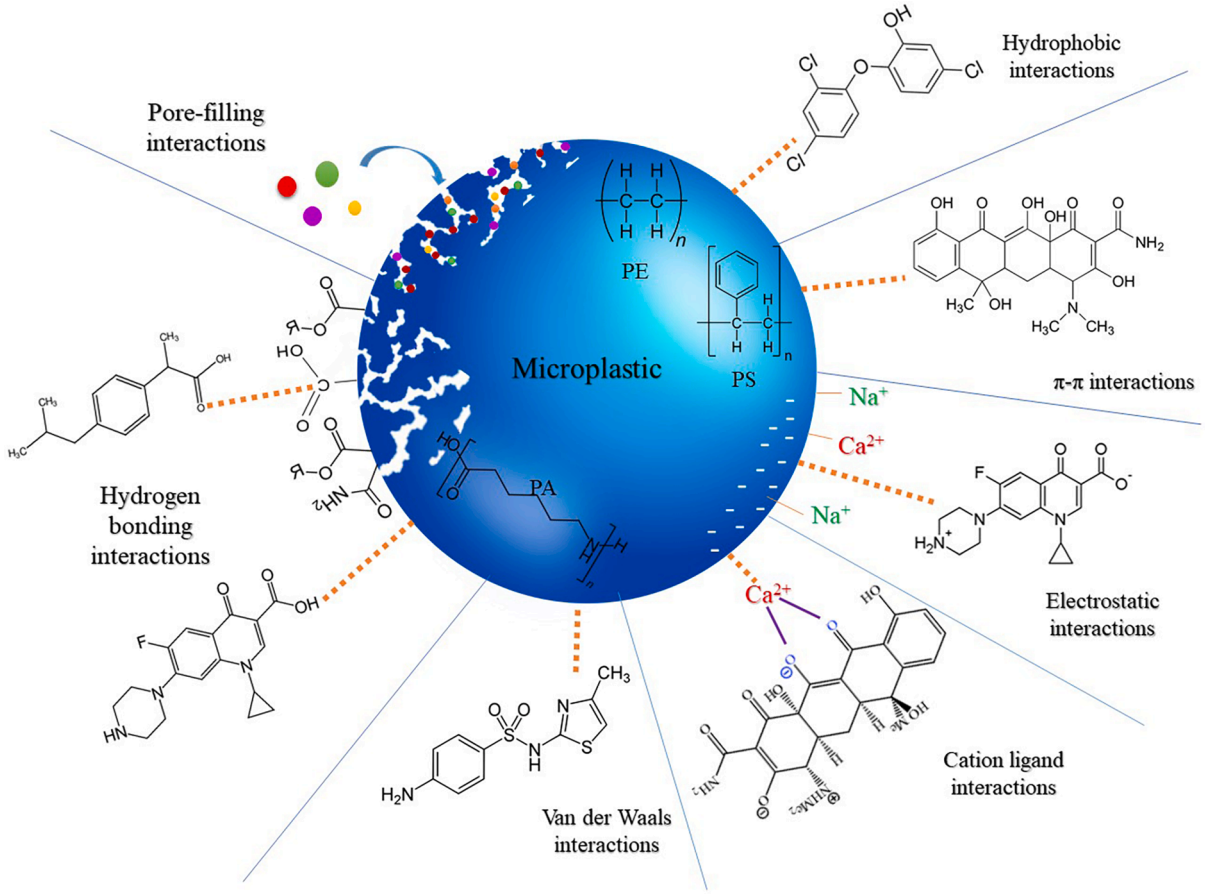

Fig. 3. Different types of interface interactions between contaminants and microplastic surfaces (adapted from Wang et al. (2020b)).

plastic surface imparts oxygen-containing groups such as carboxyl, ester, and ketone groups on the surfaces of PS, PP, and PVC, thereby facilitating hydrogen bonding with ciprofloxacin, oxytetracycline, and triclosan, which governs the mechanism of sorption of PPCP compounds on aged microplastics (Liu et al., 2019b; Wu et al., 2020; Zhang et al., 2018a). When a dipole instantaneously induces a dipole on a neighboring molecular fragment causing attraction or repulsion, the interaction is called van der Waals interactions. Van der Waals forces are non-specific, weak intermolecular forces. Typically, aliphatic polymers such as PE and PP interact with organic compounds through van der Waals forces due to the presence of non-specific functional groups (Hüffer and Hofmann, 2016; Xu et al., 2018b). The PPCP compound-microplastic interaction mechanism is reported to be governed by van der Waals forces as well. For instance, Xu et al. (2018a) mainly attributed the interaction between sulfamethoxazole and PE to van der Waals forces.

6.3. π–π interactions

The π–π interactions involve non-covalent bonds that prominently exist in aromatic compounds specifically involving π bonds. Pharmaceuticals are retained by polymers through π–π interactions, which is the basis for the development of drug-delivery systems using polymeric nanoparticles. The extent of interaction depends on the presence of aromatic pendant groups in the polymer backbone and the aromaticity of the PPCP compound (Zhuang et al., 2019). The high adsorption of PPCP compounds on aromatic polymers such as PS is attributed to the strong interaction between the conjugated π cloud of the aromatic structure of the PPCP compound and aromatic moiety of the polymer (Hüffer and Hofmann, 2016). Li et al. (2018a) reported that the sorption of ciprofloxacin, trimethoprim, and sulfadiazine on PS was higher than their sorption on PE due to the π–π interactions between the antibiotics and aromatic polymer PS. Xu et al. (2018b) and Wan et al. (2019) also reported enhanced tetracycline sorption onto PS due to an enhanced π–π conjugation between PS and tetracycline. Furthermore, π–π interactions play a major role in the higher sorption of NSAIDs (ibuprofen, diclofenac, and naproxen) on PS than on PP (Elizalde-Velázquez et al., 2020).

6.4. Pore-filling mechanism

Pore filling is an adsorption-like interaction that occurs between HOCs and glassy polymers, resulting in the filling of the nano- and micropores of the polymers by HOCs at their solubility limit (Liu et al., 2019c; Uber et al., 2019; Velez et al., 2018). Usually, the sorption isotherms of glassy polymers are non-linear, which is considered selective adsorption on the heterogeneous surface of glassy polymers (Liu et al., 2019c) (Karapanagioti and Werner, 2019). The pore-filling mechanism is the dominant mechanism for sorption by glassy polymers, such as PA, PVC, and PS (Hüffer and Hofmann, 2016). However, recently, rubbery polymers were observed to display a combination of partition and adsorption during sorption (Uber et al., 2019). The non-linear sorption behavior also suggests the pore-filling mechanism, which is similar to the adsorption of oxytetracycline onto beached PS foams reported by Zhang et al. 2018a. The high micropore area of beached PS foam facilitated pore filling, which increased the sorption capacity of beached PS foam than that of virgin foams. Pore filling is controlled by the diffusion of molecules into the pores. The adsorption mechanism of triclosan and tylosin on microplastics (PE, PP, PS, and PVC) was controlled by the particle diffusion process (Guo et al., 2018; Ma et al., 2019). The rate of diffusion of the molecules from the surface of the plastic into the pore is controlled by the size of the molecule and pore diameter. The larger pores are occupied first and the rate of sorption gradually decreases with the smaller pores remaining. Pore filling was observed to be the dominant mechanism of ciprofloxacin sorption onto PS and PVC (Liu et al., 2019b).

7. The fate of PPCP-sorbed microplastics

7.1. Ecotoxicological effects of PPCP-sorbed microplastics

Owing to their abundance and small size, microplastics have a high potential to be ingested by freshwater and marine invertebrates and vertebrates, which may lead to physical effects including physiological stress responses (Oliveira et al., 2013). Microplastics resemble plankton and food particles that are accessible to lower trophic positioned aquatic biota and many of these organisms are non-selective filter feeders that filter large quantities of water and benthic feeders that feed on sediments for nutrient uptake (Cole et al., 2013; Horton et al., 2017). Once ingested, the microplastics either are eliminated through defecation or remain in the gut or pass through the gut wall and translocate to other tissues and systems such as the circulatory system, especially in the case of nanoplastics (Browne et al., 2008; Farrell and Nelson, 2013). Similar to the harmful effects of macroplastics on larger animals, microplastics can cause the blockage of feeding appendages and intestinal tract, stop the secretion of gastric enzymes, and reduce the levels of steroid hormones and delay ovulation, thereby leading to reproductive failure and death (Carbery et al., 2018; Guzzetti et al., 2018; Wardrop et al., 2016). The accumulation of microplastics in the stomach and intestinal tract can hinder the passage of food, thus reducing the efficiency of food uptake and resulting in pseudo-satiation (Derraik, 2002; Tourinho et al., 2010). Those inhabiting oceanic gyres and deep-sea sediment are considered as microplastic reservoirs due to the higher concentrations of plastics in the deep oceanic compartment. However, even pelagic regions are recently discovered as microplastic hotspots whereas organisms in the water column do not escape of accumulating them (Carbery et al., 2018; Choy et al., 2019).

Microplastics coated with organic pollutants may transport across waters, thereby polluting otherwise pristine ecosystems, or are taken up by marine and aquatic organisms, leading to their transfer to the aquatic biota (Cole et al., 2011). Adsorbed contaminants may release when the partitioning of the contaminant to the polymer becomes low as well as the binding strength of the contaminant to the surface becomes weak. Low pH, high temperature, and low amount of DOM in the environment promote the desorption of organics (Bakir et al., 2014b). Several studies involving intra-organismal fluids (e.g., gut fluid) or the direct exposure to microplastics adhering to the interior of an organism have confirmed the desorption of adhered pollutants and their subsequent translocation to body tissues (e.g., the gut or gill walls) (Teuten et al., 2007). The rate of biotic transfer is higher in intestinal and gut fluid than that in ambient waters (Hartmann et al., 2017). Desorption behavior of pharmaceutical adsorbed microplastics in simulated gut fluid were studied by several authors. The desorption rates of PE microplastics increased in the order of sulfamethoxazole, sertraline and propranolol which correlated with the increment of the adsorption capacity and the hydrophobicity (Razanajatovo et al., 2018). A comprehensive study was done by Liu et al. (2020a) for atorvastatin (log K_{ow} 6.4) and amlodipine (log K_{ow} 2.1) in simulated gastric fluid (pH 2), intestinal fluid (pH 7) and seawater (pH 7) to examine the effect of microplastic aging for desorption. The desorption was greater for atorvastatin in intestinal fluid and for amlodipine in gastric fluid while the aging suppressed the overall desorption due to the reduction of hydrophobicity imparted by oxygen containing functional groups on the surface. However, for tetracycline and ciprofloxacin antibiotics as the binding strength was increased with aging the desorption rates decreased but on the other way the desorption capacities increased in simulated intestinal fluid (Fan et al., 2021). In addition, the studies revealed that the warm-blooded organisms are at higher risk than cold blooded organisms as desorption rates increase with higher temperatures in gut and intestinal conditions demonstrated by atorvastatin, amlodipine and tetracycline (Lin et al., 2020; Liu et al., 2020b).

More than >50% of ecotoxicological studies on microplastic-bound contaminants are focused on persistent organic pollutants (POPs) and

only a few studies have focused on PPCPs (de Sá et al., 2018). The extent and the rate at which contaminants enter the tissues or the circulatory system and the bioavailability depend on the physiological conditions and the characteristics of both the contaminant and microplastic. However, regarding pharmaceuticals, even low levels can have toxicological effects on organisms, especially during their chronic, long-term exposure to degradation products and metabolites that can accumulate in the biota. Prata et al. (2018) studied two types of antibiotics, i.e., procainamide and doxycycline, and found that compared to individual contamination, in the presence of a mixture of contaminants, microplastics have synergistic toxic effects on the marine microalga *Tetraselmis chuii*. Since organisms are often exposed to mixtures of PPCP contaminants and microplastics (co-contamination) over generations, the adverse effects on the organism may be magnified. Guilhermino et al. (2018) made a similar observation for the antimicrobial florfenicol and reported that the freshwater bivalve (*Corbicula fluminea*) was at greater risk during the combined exposure of microplastic and antimicrobial substances. Furthermore, the increase in the ambient temperature in water increased the risk for common goby juveniles (*Pomatoschistus microps*) due to contamination by both cefalexin-microplastic mixture and individual components; this demonstrates the role of environmental conditions, as discussed earlier (Fonte et al., 2016). Browne et al. (2013) discovered that the uptake of triclosan-sorbed PVC particles by lugworm (*Arenicola marina*) increased the mortality rate by over 50% of the population. The ingestion of contaminants can also cause behavioral changes in organisms. Chen et al. (2017) reported that 17α-ethynylestradiol-sorbed microplastics cause locomotory changes in zebrafish (*Danio rerio*) larvae. Moreover, the coexistence of other toxicants on the plastic surface regulates the bioavailability of a particular compound. For example, Sleight et al. (2017) found that co-contamination of microplastics by 17α-ethynylestradiol and phenanthrene reduced bioavailability more than expected when they were exposed separately.

Microplastic particles bound to toxic contaminants act as a continuous "hidden" source of contaminants as they are unintentionally taken up by organisms; therefore, they are regarded as tiny "Trojan horses" present in the environment (Cormier, 2007). With an increase in the accumulation of both microplastics and associated pollutants, the biota is continuously exposed to toxic substances over its lifetime. Since the available experimental evidence is insufficient to derive any conclusion or consensus for ecotoxicological effects, further studies are required to further understand the toxicity of PPCP-sorbed microplastics (de Sá et al., 2018).

7.2. Associated human health risk

Humans are susceptible to microplastic exposure via multiple sources, such as through the consumption of seafood and terrestrial food, drinking water, and via inhalation (Barboza et al., 2018; Eerkes-Medrano et al., 2015). Microplastics are also present in the salt derived from seawater (Seth and Shriwastav, 2018) as well as in sugar and honey (Liebezeit and Liebezeit, 2013). The main uptake is believed to take place by the ingestion of a wide variety of marine animals, including fish (mackerel, cod, and tuna), mollusks (squid and octopus), and those famed in coastal and lagoons such as bivalves (mussels and oysters) and crustaceans (prawns) (Barboza et al., 2018). Microplastics

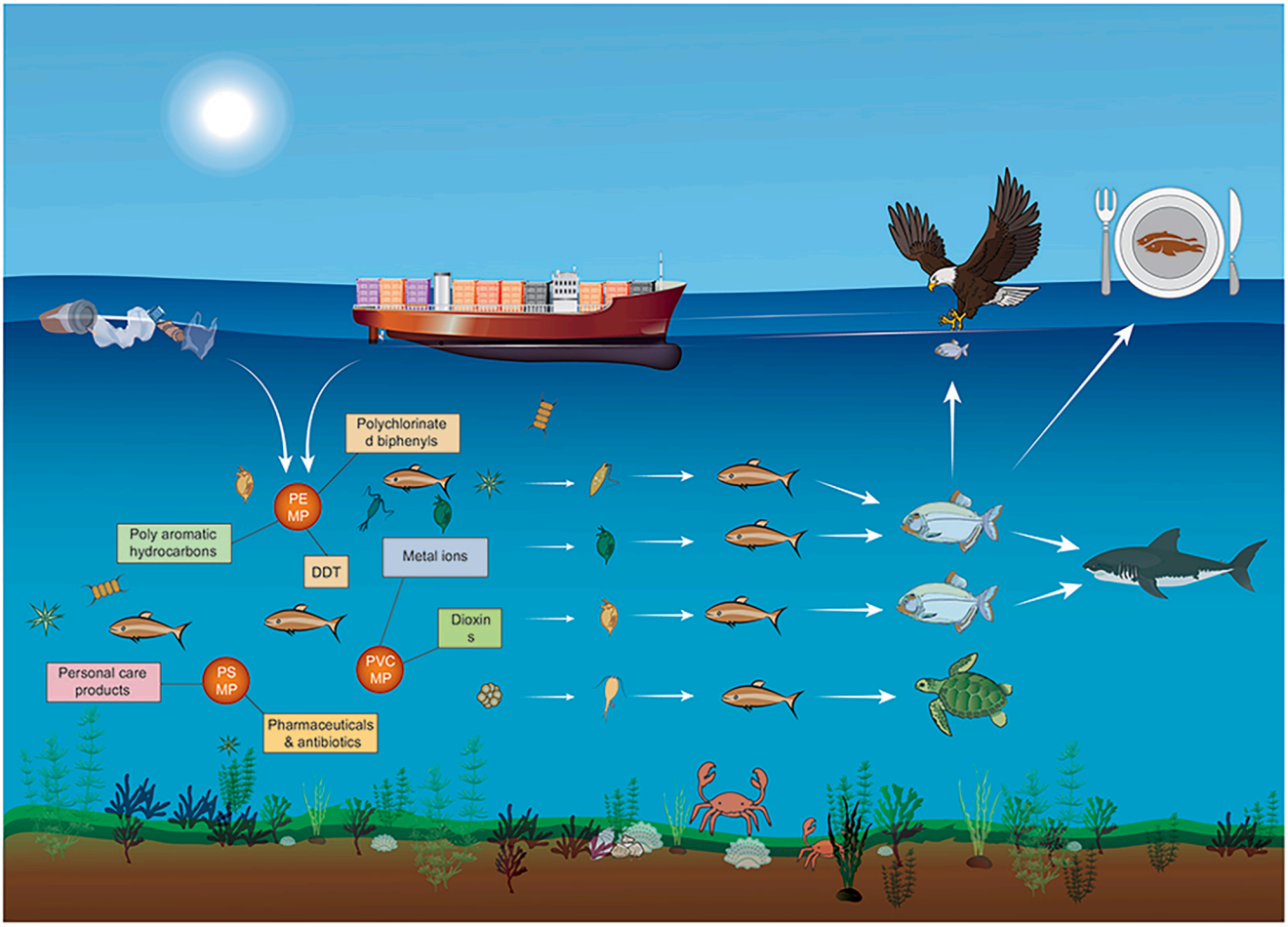

Fig. 4. Trophic transfer possibilities of microplastic-bound PPCP compounds and other contaminants in the ecosystem.

can be ingested indirectly through trophic transfer along with the food web due to the natural prey-predator relationship and may accumulate in higher trophic positioned organisms (Barboza et al., 2018; Carbery et al., 2018; Cole et al., 2013) (Fig. 4).

Since microplastics are considered to be a complex and diverse suite of contaminants (Rochman et al., 2019) including such substances such as PAHs, PCBs, phthalates, bisphenols, hormones, and pharmaceuticals, they can transfer contaminants, thus exposing humans to the physical and chemical toxicities of microplastics. Most investigations have studied trophic transfer and bioaccumulation of POPs, and studies on PPCP compounds, endocrine-disrupting chemicals, and metals are scarce (EFSA, 2016)). Humans are exposed to these chemicals either by the direct ingestion of microplastics or by secondary sources such as the consumption of organisms that bioaccumulated these contaminants by consuming contaminated prey (Carbery et al., 2018). The bioaccumulation of these contaminants in the human body can result in skin irritations, respiratory problems, cardiovascular diseases, digestive problems, and reproductive issues (Naik et al., 2019).

Moreover, as antibiotics are constantly being released into the aquatic environment at hazardous levels, there is an increasing concern about microplastics being the drivers of antibiotic resistance (Laganà et al., 2019). Microplastics assist the evolution of different microbial communities and promote gene exchange/transfer between the colonized communities on the plastic surface (Wang et al., 2020c). The exposure to antibiotics adhered on the microplastic surface may promote the emergence of antibiotic-resistant bacteria, multi-antibiotic-resistant bacteria known as "superbugs," spread of antibiotic resistance genes, and increase in the effective dose for bacterial inhibition, which may pose a significant threat to human medical treatment (Lu et al., 2019). Therefore, microplastics have the potential to serve as the vectors of chemical toxins and microbial pathogens. However, the complex interaction and toxicity of microplastics in aquatic ecosystems are still under investigation (EFSA, 2016).

8. Conclusion and future perspectives

This review discusses multiple factors that govern the microplastic-assisted vector transportation of PPCP compounds, which results in the ubiquitous presence of antibiotics, non-steroidal drugs, hormones, antimicrobial agents, and personal care products in water. Unlike HOCs, PPCP contaminants are hydrophilic and thus have different sorption mechanisms depending on plastic type. The sorption behavior is assessed in the context of environmental chemistry, physicochemical properties of the polymer, and the characteristics of PPCP compounds. The pH, ionic strength, and DOM of the matrix significantly affect the microplastic–PPCP interaction either by promoting or by suppressing the sorption. More hydrophilic compounds exhibit greater adsorption affinity toward polar and amorphous polymers. Hence, PPCP compounds tend to adsorb on weathered and aged microplastics than on pristine microplastics because of induced polarity on the plastic surface.

Both hydrophobic and electrostatic interactions mainly govern the sorption mechanism of PPCP compounds on microplastics. However, further studies are required to understand the sorption affinity to more polar plastics than to non-polar PE, PP, and PS, which have been widely used in previous studies. Since most studies are based on antibiotics, the effects of other groups of PPCP compounds remain largely unknown. Future studies should focus on the interaction of microplastics with β-blockers, antidepressants, NSAIDs, analgesics, steroidal hormones, antimicrobials, and ultraviolet screening agents. The effects of pH, salinity, and DOM on the kinetic and isotherm sorption behaviors have not been studied and need to be urgently investigated as they affect the maximum sorption capacity and the rate of equilibrium establishment when the contaminant concentration is greater than the environmental levels of contaminants. Although aged microplastics can efficiently retain hydrophilic compounds, there are limited studies on laboratory-aged microplastics or field-extracted weathered microplastics. Hence, researchers should focus on aged microplastics and their sorption capacity for PPCP compounds using environmentally realistic conditions. In addition, it is important to pay attention to personal care products especially considering that primary microplastics (i.e., microbeads) are already in contact with these compounds in exfoliation products, facial cleansers, and cosmetics. With increasing global demand of plastic products, the pollution of marine and freshwater has increased, resulting in the continuous pollution of soil and water by microplastics, which is further exacerbated by the weathering of the plastic surface that promotes the sorption of PPCP contaminants. This has led to intensive research on the interaction of microplastics with PPCP contaminants. Nevertheless, the fate of PPCP-sorbed microplastics and their ecotoxicological effects on biota, especially health risks for humans, are still not well understood. Therefore, further research is required to face the upcoming challenges of the toxicity of PPCP-sorbed microplastics to humans and higher trophic mammals.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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