

REVIEW

Vector transport of microplastics bound potentially toxic elements (PTEs) in water systems

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Summary: Microplastics can act as a vector to transport various organic and inorganic contaminants. Hydrophobic and hydrophilic organic contaminants tend to bind to microplastics due to their hydrophobicity and high surface area to volume ratio. Recent studies have focused their attention on evaluating the ability of microplastics to bind potentially toxic elements (PTEs). The co-occurrence of microplastics and PTEs may be facilitated by the ubiquitous presence of both in the environment. The metal adsorption of different microplastics has been investigated under different environmental factors and polymer properties to reveal possible interactions. The environmental factors such as solution pH, dissolved organics, dissolution media, and ionic strength have been studied the most and recognized as factors governing the adsorption of PTEs. Degree of aging and polymer type have been highlighted as the key polymer properties which influence the adsorption of PTEs. However, the effects may differ with different PTEs and environmental conditions. Though sorption capacities and mechanisms have been extensively studied, critical analysis of their behaviour in co-existence with other ions in aqueous media remains unexplored. This review focuses on critically assessing the partition coefficients between different microplastics and water for PTEs in the presence of various factors that influence the metal adsorption. Besides, postulated interactions for the adsorption of PTEs in the presence of dissolved organics, competitive ions, and different pH values are overviewed. Moreover, the associated health risks on biota and humans, when they are exposed to microplastics bound PTEs are also discussed.

Keywords: Ecotoxicity, heavy metals, microplastics, partition coefficient.

INTRODUCTION

The ubiquitous presence and overuse of plastics have become a huge environmental problem up to date, as plastics are recognized as an emerging contaminating pollutant (Alimi *et al.*, 2018; Atugoda *et al.*, 2020). Due to the uncontrolled usage of plastics stuff, the release of plastics into the environment has distinctly increased. The main pathways to transport plastics into the environment are disposal effluents of wastewater treatment plants, compost, and bio-solids, landfills, treated sewage sludge, and atmospheric deposition (Kershaw, 2016; Kilponen 2016; Rochman, 2018). Any type of plastic nurdles, fragments, or fibres with a diameter ranging from 100 nm to < 5 mm are defined as microplastics (Alimi *et al.*, 2018). Based on the formation pathways, microplastics can be categorized into two types as primary microplastics and secondary microplastics (Bradney *et al.*, 2019). Synthetically produced microplastics, including plastic nurdles, fibres, and powders for the manufacturing of textiles, plastic products, personal care products, cosmetics, and pharmaceuticals are considered as primary microplastics (Duis & Coors, 2016). Secondary microplastics are formed from the breakdown of the larger plastic debris. As illustrated in Figure 1, microplastics act as leading sources of contaminants harmful to the terrestrial and aquatic environments and life forms.

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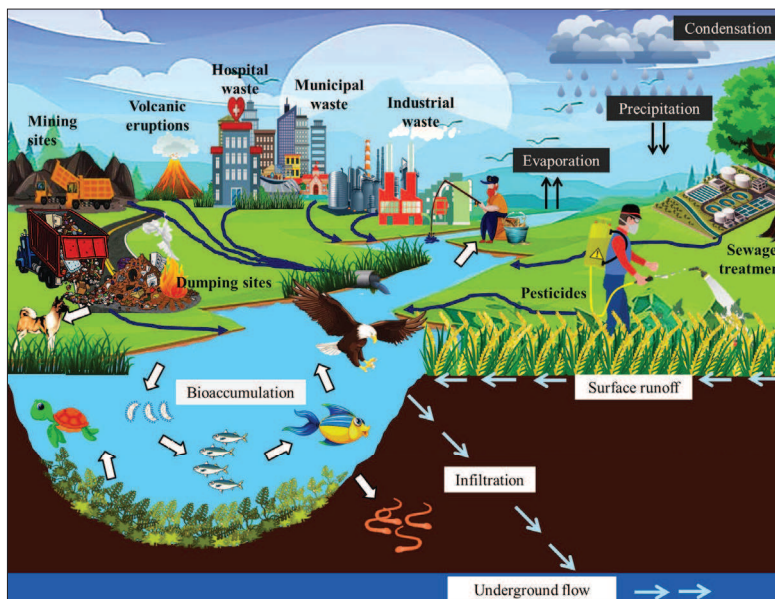


Figure 1: Sources of microplastics and potentially toxic elements (PTEs) and their potential environmental impact during the microplastic cycle in the ecosystem

Due to high hydrophobicity and high surface area to volume ratio of the microplastics, they can greatly facilitate the migration of contaminants through water, acting as a fine vehicle for them (Teuten *et al.*, 2007; Rochman, 2018; Tang *et al.*, 2020). Microplastics have shown an affinity to both hydrophobic organic pollutants and hydrophilic inorganics such as potentially toxic elements (PTEs). Moreover, plastics can also act as a vector for PTEs influencing their transport through the different environmental systems (Godoy *et al.*, 2019; Purwiyanto *et al.*, 2020; Fu *et al.*, 2021). The surface characteristics of microplastics, such as surface area to volume ratio, porosity, fragment shape, particle size, and reactivity support this vector transportation (Holmes *et al.*, 2012; Bakir *et al.*, 2014; Brennecke *et al.*, 2016). The microplastics bound contaminants may also be an immense threat to the ecosystem due to their combined toxic effects (Banaee *et al.*, 2019).

The presence of both PTEs and microplastics in water systems may accelerate the transport of microplastics bound trace metals in water (Guo *et al.*, 2020; Aghilinasrollahabadi *et al.*, 2021). Further, environmental factors in the adsorption matrix such as solution pH, ionic strength, dissolved organic matter, and temperature of the solution readily facilitate the vector transport of microplastics in water systems (Ahechti *et al.*, 2020; Tu *et al.*, 2020). Several studies have investigated the implications for the microplastics bound

vector transport of PTEs, highlighting its severe effects on living organisms, including humans. Recent studies have demonstrated the ingestion and bioaccumulation of microplastics bound PTEs in the gut and stomach of various species of fish (Oliveira *et al.*, 2013; Carbery *et al.*, 2018). Moreover, microplastics have been detected in gills and mantle of oysters [4.53 items g^{-1} (wet weight)] while PTEs were found in excessive concentrations (30.5, 4.4, 0.4 and 181.0 $\mu\text{g g}^{-1}$ dry weight of Cr, Cd, Pb and Cu, respectively).

Recently, the interactions and possible mechanisms between PTEs and microplastics have been critically discussed highlighting the effect of environmental factors (Bradney *et al.*, 2019; Liu *et al.*, 2021). Besides, the combined toxic effects of PTEs and microplastics on organisms have also been reviewed in a few studies (Cao *et al.*, 2021; Huang *et al.*, 2021). Even though the vector transportation of microplastics bound PTEs has been explored extensively, still their adsorption based on partition coefficients (PCs) and adsorption capacities have not been reviewed critically. As PCs of PTEs for different microplastics are unique for each combination, they need to be comparatively assessed. Therefore, to overcome this knowledge gap, a detailed and critical overview on PCs between microplastics and PTEs existing in water is provided from this review, to tackle issues regarding outstanding microplastics vectors and readily adsorptive PTEs on their surfaces.

The main objectives of this review are to compare and evaluate the existing information on the adsorption of PTEs on microplastics, to have a summarized account on (1) microplastics-PTEs interactions based on (a) pH, dissolved organic matter, ionic strength, and temperature in the solution, and (b) the physicochemical properties of microplastics; (2) partition coefficients between a

variety of microplastics and water for different PTEs; (3) the potential environmental impact and health risk of microplastics bound to PTEs on both humans and animals; and (4) major future perspectives to overcome existing research deficits regarding the transportation of microplastics bound to PTEs.

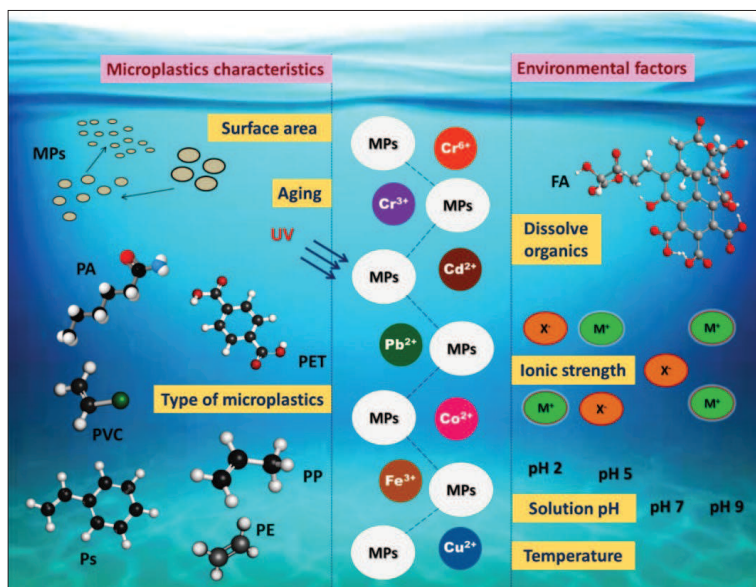


Figure 2: Different environmental factors and characteristics of microplastics influencing the PTEs adsorption on microplastics in water systems

INFLUENCING FACTORS FOR THE MICROPLASTICS BOUND PTEs TRANSPORT

Adsorption of PTEs on to microplastics depends upon either the characteristics of microplastics or environmental factors (Figure 2) (Saeedi *et al.*, 2018; Mao *et al.*, 2020; Tang *et al.*, 2020). Several environmental factors such as pH, dissolved organics, ionic strength, and the temperature influence the transport of microplastics bound PTEs, while the properties of microplastics, such as polymer type, surface characteristics, and degree of aging influence the affinity of microplastics for PTEs (Saeedi *et al.*, 2018; Richard *et al.*, 2019; Mao *et al.*, 2020). Those factors may accelerate or interrupt the adsorption process, having positive and negative effects on the binding mechanism.

Environmental factors affecting metal sorption

Effect of pH

Solution pH is an important property that would influence the adsorption of PTEs to microplastics, as it can affect the total chemical reaction, biological toxicity, and equilibrium conditions altering the chemical and biological characteristics of the solution (Marion *et al.*, 2011). Additionally, pH can also affect the adsorption of PTEs by influencing metal ion speciation (Yang *et al.*, 2019; Dong *et al.*, 2020). As an example, Cr may appear at low pH values as HCrO_4^- , CrOH^{2+} , and Cr^{3+} and at high pH values it appears as CrO_4^{2-} , $\text{Cr}(\text{OH})_3$, $\text{Cr}(\text{OH})_4^-$ (McNeill *et al.*, 2012). Therefore, adsorption of PTEs may drop down due to the formation of non-

targeted differently charged metal species that repulse the microplastic surfaces. For instance, at higher pH conditions, Cu is no longer involved in the adsorption process as it appears in the solution as various ionic species (CuOH^+ , $\text{Cu}_2(\text{OH})_2^{2+}$) and precipitated $\text{Cu}(\text{OH})_2$ (Yang *et al.*, 2019). Similarly, a reduced adsorption was observed due to the speciation of As from H_3AsO_3 to AsO_2^- ions above pH 4 (Sun *et al.*, 2010; Dong *et al.*, 2020). Consequently, solution pH can greatly effect the adsorption of PTEs by altering their ionic nature. As well, the surface charge of microplastics is highly dependent on the pH of the solution based on its point of zero charge pH (pH_{pzc}) (Tan *et al.*, 2008; Xu *et al.*, 2008).

Generally, pH_{pzc} of all types of microplastics is less than 7 (Xu *et al.*, 2018). When, the solution pH is higher than the pH_{pzc} , the surface of microplastics acquires negatively charged binding sites (Li *et al.*, 2019). Consequently, at the environmentally relevant pH values and above, microplastics surfaces are negatively charged, which enhances metal adsorption to the surface through electrostatic interactions. Several studies have evaluated the pH_{pzc} and revealed that sorption is mainly governed by the negatively or positively charged surfaces of microplastics (Atugoda *et al.*, 2020; Tan *et al.*, 2008). Accordingly, with a slight change of the solution pH, the sorption capacity can be conspicuously influenced by altering the surface charge. However, when the adsorption is not predominately governed by the electrostatic interactions, there is no significant influence from pH.

Effect of temperature in the solution

Depending on the enthalpy change of the sorption mechanism, the adsorption of PTEs on microplastics varies at different temperatures. If the sorption process is endothermic, at higher temperatures microplastics tend to bind PTEs favourably. On the other hand, exothermic sorption processes are interrupted at higher temperatures. Studies on the effect of temperature on the adsorption of PTEs onto different microplastics and documented data are very recent and limited. A few shreds of evidence that highlighted the influence of temperature on adsorptive behaviour can be found in the reviewed pool of literature.

In most studies, adsorption of different PTEs on microplastics has been amplified at higher temperatures, implying that most adsorptions of PTEs are endothermic processes. For instance, Tang *et al.* (2020) showed an increased chemisorption of Pb onto aged nylon microplastics at 313 K, compared to 300 and 288 K. Similarly, polyvinyl chloride (PVC) and polystyrene (PS)

achieved their maximum Pb adsorption capacity when the temperature increased from 288 K to 308 K, whereas polyethylene (PE) demolished that trend showing its highest maximum adsorption at 298 K (Lin *et al.*, 2021). Comparably, when the temperature increased from 288 K to 308 K, the adsorption capacity of Cu ions onto aged polyethylene terephthalate (PET) microplastics was boosted from 119.4 to 200.5 $\mu\text{g g}^{-1}$ while that of Zn was raised from 74.8 to 153.6 $\mu\text{g g}^{-1}$ (Zhang *et al.*, 2020). In contrast, As adsorption on PS microplastics was decreased while increasing the temperature, indicating that the adsorption process is favourable at lower temperatures (298 K) (Dong *et al.*, 2020). Consequently, it is clear that the temperature of the system can greatly influence the heavy metal sorption onto the microplastics surface. The reported experimental data support prediction of the favourability of the sorption and possible mechanisms, given the thermodynamic nature of the sorption process, to some extent.

Effect of dissolved organic matter (DOM)

Organic matter present in the sorption media is an important factor that can adversely affect the adsorption of PTEs on to the microplastics through complex interactions (Baken *et al.*, 2011; Refaey *et al.*, 2014; Xu *et al.*, 2016; Wijesekara *et al.*, 2018). Obviously, the positively charged metal ions have a greater binding affinity to DOM, forming electrostatic interactions or dative bonds with the lone pairs in carbonyl, carboxyl functional groups, and hydroxyl groups existing in fulvic acid (FA) and electron rich aromatic rings and fatty acids moieties contained in humic acid (HA) (Wang *et al.*, 2019; Wang *et al.*, 2021; Wei *et al.*, 2019). The electrostatically or datively bound DOM-metal ion complex facilitates the sorption, retention, and redistribution of metal ions considerably between the solution and the microplastic surface (Gao & Pedersen 2010; Zhao *et al.*, 2011). As an example, at higher HA concentrations, adsorption has been increased due the interactions of HA with Pb and Cd ions (Guo *et al.*, 2020; Fu *et al.*, 2021). Without limiting their binding to PTEs, DOM further interacts with microplastics accommodating more PTEs.

Some microplastics which have π electron containing functional groups (carbonyl, double bonds, aromatic rings) are capable of complexing with HA and FA, forming π - π conjugation and hydrophobic interactions (Guo *et al.*, 2015; Chen *et al.*, 2018). For instance, PS nano plastics interacted with DOM through π - π conjugation, confirming the interactions between DOM and microplastics (Chen *et al.*, 2018). When both DOM and PTEs are treated with microplastics, PTEs have to

compete with DOM to cohere to the adsorption sites of the microplastics, as DOM interacts first with the surface of the microplastics through hydrophobic complexation (Refaey *et al.*, 2014; Saeedi *et al.*, 2018; Wijesekara *et al.*, 2018). However, either the combined effect of HA-metal complexation and HA-microplastics complexation or individual effects may positively support the adsorption of PTEs on the surface of microplastics. Apart from positive effects, DOM may also influence the adsorption of PTEs negatively.

At higher DOM concentrations, the complexation of HA with microplastics and HA with PTEs readily inhibits the adsorption of PTEs due to the huge hindrance between the microplastics surface and the metal ions. The complexed PTEs are no longer able to attach onto the microplastic surface (Xu *et al.*, 2006; Tan *et al.*, 2008). As an example, Pb ions have exhibited a decreased adsorption onto nylon microplastics in the presence of higher FA concentrations, indicating the obstructive effects of DOM on the metal adsorption (Tang *et al.*, 2020). Further, the formation of MP-metal-DOM ternary complexes can also be accelerated (Gu & Karthikeyan, 2008; Guo *et al.*, 2020). Consequently, DOM in the sorption media can influence the adsorption of PTEs on to microplastics surface negatively or positively. Finally, it can be concluded that the DOM bound to microplastics or DOM in the sorption medium can directly influence the adsorption of PTEs. However, depending on the microplastics type, DOM concentration, and the sequence in which the DOM or PTEs reach the surface of the microplastics, the ways of influence may differ.

Effect of ionic strength

Different types of salt substances except PTEs and DOM, existing in the water system, are one of the most critical influencing factors which determine the extent of the adsorption process. More importantly, the effects of ionic strength on the adsorption can vary depending on factors such as the type of polymer, the distribution coefficient, and speciation of PTEs (Holmes *et al.*, 2014; Wang *et al.*, 2018; Yu *et al.*, 2019). The addition of positively charged ions would alter the electrostatic interactions between PTEs and microplastics by inverting the surface charge and reducing the activity (Mattigod *et al.*, 1979; Xu *et al.*, 2018). In addition, PTEs have to compete with the positively charged ions for the activated sorption sites (Wang *et al.*, 2010; Haas *et al.*, 2019; Guo *et al.*, 2020). As an example, when cations were added to the solution, Cr^{3+} and Co^{2+} showed reduced adsorption onto PE and PS surfaces due to the competition between the cations for the negatively charged adsorption sites (Godoy *et al.*,

2019). The presence of salt ions can disrupt the metal-microplastics interactions by acting as an electrostatic screen between the microplastics surface and the metal ions (Vermöhlen *et al.*, 2000; Torres *et al.*, 2007). For instance, Cd adsorption on PVC, PE, PS, and PP and Pb adsorption on PVC, PS, and PE have been reduced at higher ionic strengths (Guo *et al.*, 2020; Lin *et al.*, 2021). Also, in the presence of higher ionic strengths, the number of activated adsorption sites can be reduced as the adsorbent particles start to agglomerate and aggregate (Eren & Afsin 2007; Godoy *et al.*, 2020). However, the impact of ionic concentration is not limited to adsorbates and adsorbent; it can further lead to changing the electric potential surrounding the microplastics surface.

Salt ions often commence penetrating an electric double layer on the microplastic surface forming a denser aggregation structure close to the surface (Yin *et al.*, 2019). Hence, adsorption of PTEs diminishes at higher ionic strength as the thickness of the electrical double layer begins to decrease, resulting in a reduced electrostatic potential and total free energy (Tang *et al.*, 2020). For instance, Tang *et al.* (2020) explained the inhibiting effect of NaCl for Pb adsorption on the nylon microplastics employing the total free energy deduction. However, at higher temperatures, this phenomenon would be changed, because the energy available for the adsorption increases. Moreover, the effects of ionic strength are not same for the adsorption of different PTEs on the same microplastic. As an example, Cd, Co, Ni, and Pb showed significantly reduced adsorption on PE pellets at higher salinity while Cr exhibited enhanced adsorption (Holmes *et al.*, 2014). Extrusion effects and salting-out effects in the solution are key factors which can also have an impact on the adsorption of PTEs, for instance, where the extrusion effect is stronger than the salting-out effect in the solution. Consequently, it can be noted that ionic species in the sorption matrix have an effect on the adsorption of PTEs in different ways, with either positive, negative, or zero effects depending on the type of microplastics.

Effect of the properties of microplastics

Physical properties

Surface properties of the microplastics such as surface area to volume ratio, porosity, fragment shape, and particle size can play a vital role in influencing the metal adsorption on to the microplastics surface (Bakir *et al.*, 2014; Holmes *et al.*, 2014; Brennecke *et al.*, 2016). Depending on the shape of microplastics, virgin PS beads have exhibited a decreased adsorption due

to their smaller surface area to volume ratio than aged PVC fragments having an irregular shape with a higher surface area (Brennecke *et al.*, 2016). In addition, the adsorption of As was decreased when increasing the particle size of PS microplastics indicating that smaller particle sizes enhance As adsorption (Dong *et al.*, 2020). According to the specific surface area of the PE, PP, PS, and PVC microplastics, the sorption rate of Cd ions onto their surfaces was increased in sequence PE < PP < PS < PVC (Guo *et al.*, 2018; 2020). By contrast, surface properties of microplastics are not always the main factor for capturing Pb. As an example, though the order of surface area per unit of mass was PS > PP > PE > PVC > PET, the adsorption performances were changed without any order indicating that the adsorption was probably not depended on all surface chemical and physical properties (Godoy *et al.*, 2019). Therefore, the sorption behaviour of metals to microplastics would be expected to differ with the change of surface area, shape, and hydrophobicity of each type of microplastics (Teuten *et al.*, 2007; Karapanagioti & Klontza, 2008).

Degree of aging

During the aging process, the surface of microplastics begins to change the surface uniformity, becoming rough and forming pores. Further, aging leads to change in particle size and degree of folding of microplastics (Lang *et al.*, 2020; Luo *et al.*, 2020). Therefore, these possible surface changes increase the surface area of microplastics promoting the vector transport of metal pollutants (Paul-Pont *et al.*, 2016; Zhang *et al.*, 2020). Aged PVC fragments showed comparatively higher Cu adsorption capacities than virgin PS beads (Brennecke *et al.*, 2016). Moreover, the generated wrinkles and cracks during the aging process led to increasing the number of activated sites for metal adsorption, and hence accelerate the adsorption through a pore filling mechanism (Lang *et al.*, 2020; Mao *et al.*, 2020). Aging time and aging environment also influence or enhance metal adsorption (Luo *et al.*, 2020). Illustratively, degree of aging of PS microplastics in different environments were UV air > UV seawater > UV pure water (Mao *et al.*, 2020). Aging also eases the adsorption by oxidizing the functional groups in the microplastics, forming carbonyl and carboxylic groups (Fotopoulou & Karapanagioti, 2012; Liu *et al.*, 2019; Mao *et al.*, 2020). Further, processes such as photo-oxidative weathering, accumulation of biofilms, and chemical hydrogenous precipitates can also influence the metal adsorption on microplastics (Morét-Ferguson *et al.*, 2010; Holmes *et al.*, 2012). Additionally, newly formed polar moieties like carbonyl and carboxylic groups containing lone pairs in aged

microplastics preferentially attract metal ions through electrostatic interactions or dative bonds (Cao *et al.*, 2021). Accordingly, it is clear that aging promotes the adsorption of PTEs by changing the physical and chemical characteristics of microplastics surface.

Type of microplastics

The metal sorption capacity may be influenced by the type of microplastics. The specific functional groups in microplastics, such as, chlorine, amide, hydroxyl, carboxyl, and aromatic enhance the metal adsorption through electrostatic and hydrophobic interactions by increasing the polarity of the polymer surface. Moreover, the affinity of positively charged metal ions to polar functional group makes the adsorption easier through electrostatic or dative bonds (Cao *et al.*, 2021; Liu *et al.*, 2021). Interestingly, most common plastics such as PS, PA, and PET have at least one π -electron rich or polar region which is able to attract metal ions. The hydroxyl and carboxyl groups introduced to the polymer structure of PS microplastics during the ball milling process have collectively provided positive electrostatic potential for As adsorption (Dong *et al.*, 2020). Furthermore, for the Pb adsorption, PS microplastics have exhibited the maximum sorption rate, while PE and PVC showed a smaller sorption rate (Lin *et al.*, 2021). Due to the presence of a more polar chlorine group in PVC chain, it has adsorbed a higher amount of Cu than PS microplastics (Brennecke *et al.*, 2016). Therefore, it can be concluded that the type of microplastics can crucially affect the adsorption of PTEs.

ADSORPTION ABILITY OF MICROPLASTICS TO PTEs

Partition coefficient (PC) value is a more important parameter to understand the real performance of adsorbent in binding to the contaminants, than the adsorption capacity and the removal efficiency. The equilibrium PC for a given system under constant pressure, temperature, and matrix composition such as pH, ionic strength, and DOM, is independent of both adsorbate and adsorbent concentrations (Zhao & Hou, 2012). Hence, the bias in both adsorption capacity and removal efficiency can be reduced by using PC values to discuss the potential of adsorbents (Ramanayaka *et al.*, 2019; Vikrant & Kim 2019; Ashiq *et al.*, 2021). Therefore, PC values of the reviewed microplastics are listed in Table 1 to assess the true adsorption ability of microplastics with respect to PTEs. Higher PC values indicate the magnified adsorption potential of microplastics.

Numerous studies have reported that Pb and Cr have a higher affinity to adsorb onto pristine or aged microplastics such as PP, PS, PE, PVC, PET, and nylon (Tan *et al.*, 2008; Li *et al.*, 2020; Tang *et al.*, 2020). The high adsorption ability of all microplastics varieties is likely supported by the large amount of dissolved organics in the adsorption

matrix to bind more Pb (Godoy *et al.*, 2019). When the sorption media was sea water, aged PE pellets have showed remarkable adsorption performance for both Pb and Cr, compared to river water, highlighting stronger adsorption than virgin PE pellets (Holmes *et al.*, 2012; 2014; Lin *et al.*, 2021). The enhanced adsorption may be

Table 1: Adsorption capacities and partition coefficients of reviewed microplastics to different potentially toxic elements (PTEs)

Type of microplastic	PTE	Adsorption capacity ($\mu\text{g g}^{-1}$)		Partition coefficient (L g^{-1})		Sorption medium	Reference
		Pristine	Aged	Pristine	Aged		
PE Pellets	Cr	29.70×10^{-2}	44.10×10^{-2}	5.50×10^{-5}	22.10×10^{-2}	Sea water	(Holmes <i>et al.</i> , 2012)
	Co	1.80×10^{-2}	3.80×10^{-2}	1.00×10^{-5}	4.03×10^{-3}		
	Ni	0.80×10^{-2}	7.00×10^{-2}	1.40×10^{-5}	8.87×10^{-3}		
	Cu	26.10×10^{-2}	-	3.30×10^{-5}	4.52×10^{-2}		
	Cd	0.04×10^{-2}	1.00×10^{-2}	2.00×10^{-9}	7.94×10^{-3}		
	Pb	-	71.60×10^{-2}	2.00×10^{-8}	0.15		
PE pellets	Cd	1.00×10^{-2}	0.25	2.01×10^{-3}	5.23×10^{-2}	River water	(Holmes <i>et al.</i> , 2014)
	Co	6.89×10^{-2}	7.96×10^{-2}	1.40×10^{-2}	1.62×10^{-2}		
	Cr	-	4.21×10^{-2}	-	1.90×10^{-2}		
	Cu	10.04×10^{-2}	-	2.01×10^{-2}	-		
	Ni	1.66×10^{-2}	15.14×10^{-2}	3.32×10^{-3}	3.46×10^{-2}		
	Pb	19.10×10^{-2}	273.50×10^{-2}	3.97×10^{-2}	1.21		
PE pellets	Cd	0.43×10^{-4}	1.00×10^{-2}	8.60×10^{-6}	2.04×10^{-3}	Sea water	(Turner & Holmes, 2015)
	Co	1.76×10^{-2}	4.21×10^{-2}	3.54×10^{-3}	8.49×10^{-3}		
	Cr	29.74×10^{-2}	44.09×10^{-2}	6.32×10^{-2}	9.67×10^{-2}		
	Cu	26.12×10^{-2}	-	5.51×10^{-2}	-		
	Ni	0.76×10^{-2}	7.34×10^{-2}	1.52×10^{-3}	1.49×10^{-2}		
	Pb	-	68.38×10^{-2}	-	0.16		
PE pellets	Ag	1.28×10^{-2}	1.07	1.81×10^{-3}	9.63×10^{-2}	River water	(Farhan <i>et al.</i> , 2018)
	Cd	1.01×10^{-2}	0.25	3.47×10^{-4}	-		
	Co	6.92×10^{-2}	0.08	3.04×10^{-4}	-		
	Cr	-	0.09	1.01×10^{-3}	4.60×10^{-3}		
	Cu	10.00×10^{-2}	-	3.62×10^{-3}	6.10×10^{-2}		
	Hg	17.00×10^{-2}	2.78	5.84×10^{-3}	3.18×10^{-2}		
	Ni	1.66×10^{-2}	0.15	4.91×10^{-4}	-		
	Pb	19.10×10^{-2}	2.74	5.80×10^{-3}	0.22		
	Zn	-	-	-	-		
LDPE	Cr	1.70	-	1.73	-	Sea water	(Farhan <i>et al.</i> , 2018)
Mixture of PP, PE, PES, PVC, and nylon	Pb	-	0.47	-	-	River water	(Purwiyanto <i>et al.</i> , 2020)
	Cu	-	0.09	-	-		
PE	Al	6.20	-	-	-	After suspended in harbor sea water for 8 weeks	(Ashton <i>et al.</i> , 2010)
	Fe	17.98	-	-	-		
	Mn	2.61	-	-	-		
	Cu	0.28	-	-	-		
	Pb	1.72	-	-	-		
	Zn	0.25	-	-	-		
	Ag	2.47×10^{-2}	-	-	-		

Continued -

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Type of microplastic	PTE	Adsorption capacity ($\mu\text{g g}^{-1}$)		Partition coefficient (L g^{-1})		Sorption medium	Reference
		Pristine	Aged	Pristine	Aged		
PE	Cr	3340.00		71.67×10^{-2}		Distilled water	
		2560.00	-	47.06×10^{-2}	-	Sea water	
		7900.00		79		Urban water	
		6670.00		5.02		Irrigation water	
	Pb	1770.00		28.41×10^{-2}		Distilled water	
		3280.00	-	69.49×10^{-2}	-	Sea water	
		7630.00		20.62		Urban water	
		5690.00		2.46		Irrigation water	
PET	Pb	2310.00		40.60×10^{-2}		Distilled water	
		3730.00	-	87.35×10^{-2}	-	Sea water	
		7520.00		15.67		Urban water	
		5260.00		1.92		Irrigation water	
	Cu	1590.00		24.80×10^{-2}		Distilled water	
		2870.00	-	55.95×10^{-2}	-	Sea water	
		6240.00		3.55		Urban water	
		995.00		14.20×10^{-2}		Irrigation water	
PP	Pb	2290.00		40.11×10^{-2}		Distilled water	(Godoy <i>et al.</i> , 2019)
		3150.00	-	64.95×10^{-2}	-	Sea water	
		7660.00		22.53		Urban water	
		5170.00		1.83		Irrigation water	
	Co	535.00		7.17×10^{-2}		Distilled water	
		400.00	-	5.26×10^{-2}	-	Sea water	
		239.00		3.08×10^{-2}		Urban water	
		395.00		5.19×10^{-2}		Irrigation water	
PS	Pb	2390.00		42.60×10^{-2}		Distilled water	
		3290.00	-	69.85×10^{-2}	-	Sea water	
		7400.00		12.33		Urban water	
		5640.00		2.39		Irrigation water	
	Cr	1730.00		27.59×10^{-2}		Distilled water	
		6140.00	-	3.30	-	Sea water	
		7680.00		24.00		Urban water	
		7640.00		21.22		Irrigation water	
PVC	Pb	1100.00		0.16		Distilled water	
		3450.00	-	0.76	-	Sea water	
		7600.00		19.00		Urban water	
		5080.00		1.74		Irrigation water	
Mixture of PE and PP films and fragments (<5mm, mainly 2–3mm)	Ni		0.50 – 2.40			River sediments (Wang <i>et al.</i> , 2017)	
	Cd		2.10 – 17.60				
	Pb		38.20 – 131.10				
	Cu	-	80.90 – 500.60	-	-		
	Zn		2414.00 – 14815.00				
	Ti		13617.00 – 38823.70				
Orange, filamentous PE	Cd		3.94			Beach sand (Turner, 2017)	
	Cr	-	3.28	-	-		
	Pb		8.40				

governed through the electrostatic interaction between metal ions and PP and PE microplastics. Reviewed data clearly indicate that when the adsorption matrix rich in ions and DOM, both Cr and Pb ions have higher affinity to microplastics which contain more polar moieties.

Copper was excellently adsorbed onto virgin PET where urban water was the adsorption medium (Godoy *et al.*, 2019). At high temperatures, pristine PET can also act as a leading adsorbent for Cu (Zhang *et al.*, 2020). Besides, PS, PE (either as the virgin or aged form), and polymethylmethacrylate (PMMA) also have exhibited their ability to act as a carrier for Cu ions (Yang *et al.*, 2019; Mao *et al.*, 2020). In conclusion, it can be clearly identified that, at high temperatures, Cu has a high affinity for PMMA, PS, PVC and PET microplastics in the presence of surfactants and DOM rich sorption medium.

Adsorption of Zn is quite different to other PTEs, as a variety of microplastics, such as PS, PE, PET, PVC, and a mixture of PE and PP microplastic, has exhibited their adsorption potential towards Zn (Godoy *et al.*, 2019). Contrastingly, both virgin and aged PE pellets have shown zero adsorption of Zn in the presence of river water (Turner & Holmes, 2015). However, pristine PS microplastics have showed a higher adsorption ability for Zn than for Cu, while aged PVC microplastics showed nearly the same performance for both Cu and Zn in the absence of antifouling paint treatment, in seawater (Brennecke *et al.*, 2016). Nevertheless, when compared to the other PTEs, microplastics have demonstrated a lesser adsorption of Zn regardless of the sorption medium.

Virgin PS microplastics have showed the highest adsorption ability for Co through a significant enhanced adsorption in Milli-Q water than other types of water (Godoy *et al.*, 2019). Thus, adsorption potential of PS microplastics for Co was again higher than that of both virgin and aged PE pellets where seawater is the adsorption medium (Holmes *et al.*, 2012; Godoy *et al.*, 2019). However, with the limited data for Co adsorption the trend of its adsorption cannot be predicted precisely. Nevertheless, according to the available data, PS microplastics have showed outstanding adsorption ability for Co where the sorption matrix was not crowded with competing ions and organic matter.

Very little reported data on PC based adsorption performance of microplastics to bind Cd are found in the literature to date. Instead of PC values, most studies have investigated only the adsorption capacities

of different microplastics (Turner 2017; Wang *et al.*, 2017). Nevertheless, both aged and pristine PE pellets have showed low PC values for Cd where the sorption medium was seawater and river water (Holmes *et al.*, 2014). The study by Turner and Holmes (2015) reported very similar behaviour to that of Holmes *et al.* (2014). However, to have a clear elucidation about PC values for Cd adsorption on microplastics, the reported data is not sufficient.

The adsorption of Sr, As and Ni to different microplastics has not been studied much. However, pristine PE microplastics have shown a strongly enhanced adsorption of Sr with a 3.92 L g^{-1} PC value, compared to that for the PET microplastics. In addition, PS microplastics adsorbed As ions resulting in a 0.023 L g^{-1} PC value (Dong *et al.*, 2020). Presumably, a few studies have conducted adsorption experiments to elucidate the capability of Ni to adsorb on to the aged PE and PS microplastics compared to pristine ones. Nevertheless, microplastics bound transportation of some PTEs such as Hg, Ag, Al, Ti, and Mn, which are likely to be harmful to the environment, remain relatively unexplored. Lastly, assessment of the available PC data in the literature strongly contributed to a discernment of the adsorption of PTEs on different microplastics to some extent, elucidating the role of microplastics as sinks or sources of PTEs.

POTENTIAL ENVIRONMENTAL IMPACT

Due to the tiny particle sizes and highly availability in the environment, microplastics can be readily ingested by organisms mainly through food webs and water. For instance, microplastics can be easily mistaken for plankton (Egbeocha *et al.*, 2018), due to their small size and shape; hence these can be accidentally eaten by a range of small invertebrates and vertebrates that live in natural ecosystems (Mathalon & Hill, 2014; Huerta Lwanga *et al.*, 2016). Ingestion of microplastics has directly damaged organs in the digestive tract and reproductive system, leading to a decline or decrease of growth, feeding rate, nonlethal morphology and reproductive output of marine organisms (Murphy & Quinn, 2017; Curren *et al.*, 2020). Besides, when microalgae adsorb plastic particles their photosynthetic processes may drop, leading to an increase in reactive oxygen species (ROS) and having an adverse impact on biologically important molecules, including DNA, protein, and lipid, by interacting with other molecules (Liu *et al.*, 2019; Prokić *et al.*, 2019). Even so, microplastics may remain in the gut for a longer time as a result of the formation of hetero aggregates, leading to an enhanced bio-accumulation in

higher trophic levels (Carbery *et al.*, 2018; Egbeocha *et al.*, 2018). As an example, different microplastics such as PS, PE, and PP, have been discovered in commercial fish (Cho *et al.*, 2019; Robin *et al.*, 2019). However, apart from their individual effects, microplastics cause more harmful impacts on organisms during their vector transport, with combined toxic effects (Liu *et al.*, 2019).

High levels of PTEs (As, Cd, Cr, Cu, Ni, Pb, and Zn) and microplastics (PE, PET) can be found in both marine and freshwater animals (Murphy & Quinn, 2017; Zhu *et al.*, 2020; Sarkar *et al.*, 2021). However, the combined toxic effects may vary with the type of plastics, selected PTE, and test species (Liu *et al.*, 2021). The ingestion of microplastic bound PTEs have higher potentially harmful effects upon the feeding behaviour, growth, and reproduction of marine organism (Anderson *et al.*, 2016; Botterell *et al.*, 2019). For instance, when Cd and microplastics were applied to carp, their blood biochemical and immunological indicators were significantly changed (Banaee *et al.*, 2019). Also, microplastics most probably concentrate in the digestive system, influencing food intake and oxygen uptake, and later with more effect upon the absorption of nutrients (Zhu *et al.*, 2020). Moreover, desorption of microplastics bound PTEs in gut fluids is facilitated by various gut fluid components (Hodson *et al.*, 2017). In addition, the presence of microplastics in fish accelerate their intake of heavy metals, which induce chronic health issues in humans through food chains (Liu *et al.*, 2021). The impacts on humans depend upon dose, level of exposure, age, sex, and genetics (Godoy *et al.*, 2020). Accordingly, long term consumption of microplastics bound PTEs may lead to prolonged lethal effects in both human and biota.

FUTURE PERSPECTIVES

Although important and enormous findings are available to describe the sorption behaviour of metal contaminants with microplastics, considerable data gaps have arisen when interpreting their sorption. Hence, the following unexplored issue should be studied extensively. Though theoretically it is considered that the equilibrium PC is a system constant, independent of adsorbate and adsorbent concentrations, several studies have experienced that experimentally measured PC values of a particular system decreased with increasing adsorbent concentration (Hatje *et al.*, 2003; Kim *et al.*, 2003). Therefore, the PC itself has several limitations for evaluating the adsorption performance. Accordingly, future studies are needed to focus on new parameters, to explain adsorption

capacities precisely. Since non-polar and non-degradable microplastics have been widely used in previous studies, future work should be mostly focused to determine the heavy metal adsorption affinity of polar and degradable microplastics with regard to their functional groups. The precise role of aged microplastics and their effect on the adsorption of PTEs deserves to be further studied in order to identify the exact mechanisms, including the formation of new binding sites, use of weathering layers as a precipitation nucleus, etc., that account for efficient metal retention on microplastics. Most studies on the adsorption of PTEs were performed with the use of laboratory-aged or field-extracted weathered microplastics under simulated laboratory conditions. Hence, future investigations should be urgently directed towards experiments under environmentally realistic conditions. Also, future investigations ought to emphasize the solid-liquid interactions which may support the adsorption of PTEs.

Researchers should pay attention to evaluate the role of a mixture of microplastics in transporting and retaining a broader range of PTEs. Thus, it is necessary to study how the competing ions in the water systems act on the adsorption of PTEs. In addition, sorption studies under microbial exposure merits future study. Besides, enthalpy, entropy, and free energy changes of the adsorption processes should be examined to understand the thermodynamics. Further, particular attention must be paid to study the desorption of heavy metals adsorbed on microplastics in fragile ecosystems. Additional information is needed to determine the burden of plastics-bound heavy metals on wildlife, their migration to different organisms, and possible conflicting effects. Also, a realistic risk assessment is urgently required to address the possible risks associated with metal-bound microplastics to the whole ecosystem. Moreover, an exploration of the impact of heavy metal release from microplastics under different gut environments is needed.

Conflict of interest

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

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