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Review

Risk associated with microplastics in urban aquatic environments: A critical review

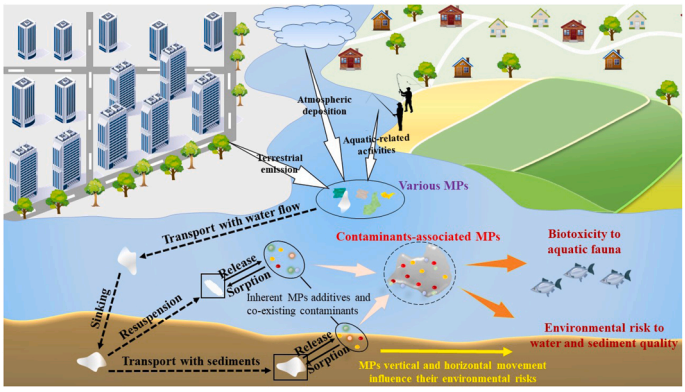
Beibei He [a,b], An Liu [a,*], Huabo Duan [b], Buddhi Wijesiri [c], Ashantha Goonetilleke [c]

[a] College of Chemistry and Environmental Engineering, Shenzhen University, Shenzhen 518060, China
[b] College of Civil and Transportation Engineering, Shenzhen University, Shenzhen 518060, China
[c] School of Civil and Environmental Engineering, Faculty of Engineering, Queensland University of Technology (QUT), P.O. Box 2434, Brisbane, Qld 4001, Australia

HIGHLIGHTS

- MPs' occurrence, sources and movement behaviour in urban waterways were reviewed.
- MPs movement can influence their environmental risks in freshwater systems.
- Physicochemical properties of MPs affect release/sorption of other contaminants.
- Risk assessment of contaminants associated with MPs needs significant more research.
- Current MPs transport modelling primarily based on virgin plastics rather than aged.

GRAPHICAL ABSTRACT



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ABSTRACT

The presence of microplastics (MPs) has been recognized as a significant environmental threat due to adverse effects spanning from molecular level, organism health, ecosystem services to human health and well-being. MPs are complex environmental contaminants as they bind to a wide range of other contaminants. MPs associated contaminants include toxic chemical substances that are used as additives during the plastic manufacturing process and adsorbed contaminants that co-exist with MPs in aquatic environments. With the transfer between the water column and sediments, and the migration within aquatic systems, such contaminants associated MPs potentially pose high risk to aquatic systems. However, only limited research has been undertaken currently to link the environmental risk associated with MPs occurrence and movement behaviour in aquatic systems. Given the significant environmental risk and current knowledge gaps, this review focuses on the role played by the abundance of different MP species in water and sediment compartments as well as provides the context for assessing and quantifying the multiple risks associated with the occurrence and movement behaviour of different MP types. Based on the review of past literature, it is found that the physicochemical properties of MPs influence the release/sorption of other contaminants and current MPs transport modelling studies have primarily focused on virgin plastics rather than aged plastics. Additionally, risk assessment of contaminants-associated MPs needs

* Corresponding author.
E-mail address: liuan@szu.edu.cn (A. Liu).

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significantly more research. This paper consolidates the current state-of-the art knowledge on the source to sink movement behaviour of MPs and methodologies for assessing the risk of different MP species. Moreover, knowledge gaps and emerging trends in the field are also identified for future research endeavours.

1. Introduction

The presence of microplastics (MPs) in world's aquatic environments is a critical issue of concern because of the potential adverse impacts on human and ecosystem health (Rochman et al., 2013). Reports on the occurrence of MP particles in aquatic systems originated from the early 1970 s (Carpenter et al., 1972), primarily focusing on the sources, spatial and temporal distribution, and influential factors (Hasan Anik et al., 2021; Weiss et al., 2021; Zhang et al., 2022). As a result of rising awareness of the detrimental effects such as the lethality of MPs on aquatic fauna (Gao et al., 2022; Lei et al., 2018; Li et al., 2020b), risk assessment studies have primarily focussed on their inherent toxicity and the consequences of interactions with other contaminants in the environment (Ricardo et al., 2021).

Large quantities of MPs are generated in urban environments due to anthropologic activities. Urban aquatic systems are the first places to suffer from plastic pollution from terrestrial environments (Horton and Dixon, 2018). Further, polluted rivers, ponds, and lakes serve as sinks and transport media for plastic pollutants from terrestrial areas to oceans (Wright et al., 2013; Sharma and Chatterjee, 2017). Hence, it is important to investigate the presence of MP pollutants in relation to the abundance, transportation and ecological risks in relation to urban aquatic systems.

MPs refer to plastic particles with a diameter smaller than 5 mm without a specific shape (Barboza and Gimenez, 2015), which can either originate from the manufacturing process referred to as primary MPs (Duis and Coors, 2016) or derived from the fragmentation and degradation processes of large plastics which is referred to as secondary MPs (Li et al., 2016). The presence of MPs in the water column and sediments has been widely reported in riverine, estuarine, lacustrine, marshy, and reservoir systems worldwide with considerable concentrations and diverse characteristics (Szymańska and Obolewski, 2020). MPs are known for being associated with a range of toxic compounds that are either additives included during the manufacturing process or adsorbed from the surrounding environment (Groh et al., 2019; Turner and Filella, 2021). For example, toxic compounds such as polychlorinated biphenyls (PCBs) and polycyclic aromatic hydrocarbons (PAHs) are added during the production process to improve various properties including colour, strength and thermo-resilience of plastic products (Rios et al., 2007). A series of experimental studies in recent years have investigated the leaching and adsorption capacity of MPs in terms of organic and inorganic contaminants such as pesticides (PAHs, PCBs, DDTs) and metals and their ecotoxicological implications to aquatic organisms (Bour et al., 2021; Mato et al., 2001; Rai et al., 2021). To date, several reviews have focused on the spatial-temporal distributions of MPs (Barletta et al., 2019; Yuan et al., 2019), the mechanisms in relation to the interactions between MPs and toxic contaminants such as heavy metals (HMs) (Binda et al., 2021) and microbial contaminants (Mammo et al., 2020; Wang et al., 2021b), as well as the implications of MPs toxicity on aquatic organisms (Ateia et al., 2022; Rai et al., 2022; Verla et al., 2019). However, little knowledge is currently available on the risk associated with the occurrence and movement of various MPs in freshwater systems.

In this context, this review initially discusses the occurrence, sources and movement behaviour of MPs in urban aquatic systems. It then focuses on the implications of MPs as a complex contaminant as these plastic particles can leach toxic contaminants from the additives as well as adsorb other contaminants from the surrounding environment. Next, this paper reviews the toxicity of contaminants associated with MPs and current risk assessment approaches adopted for urban aquatic systems. Further, current knowledge gaps have been identified and recommendations are provided for future research from a risk assessment perspective. Fig. 1 illustrates the framework adopted for this review.

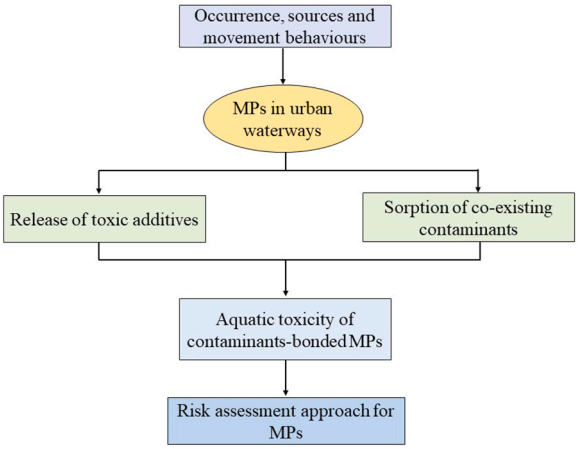

Fig. 1. Framework for the critical review undertaken.

2. Occurrence, sources and movement of MPs in freshwaters

MP pollutants in freshwater systems have been detected worldwide with uneven spatial distribution patterns. It is common knowledge that land-based discharge of plastic litter is the main contributor to MP presence in aquatic environments. However, the uneven distribution of MPs in various freshwater systems indicates that they can migrate between different environments in a variety of ways and their spatial distributions could be the result of diverse factors which include anthropogenic activities, wind, and hydraulic conditions. In general, MPs float on the water surface or water column due to the relatively lower density. However, accumulated contaminants and/or organisms on their surface will increase the weight of MPs and allow them to settle to the benthic ecosystem. Since the physicochemical characteristics of MPs and hydrodynamic parameters can largely change over time, different MP particles would have different transport mechanisms. Hence, knowledge of the migration processes of different MPs should guide the interpretation of the sources of different MPs, and contribute to strengthening management strategies.

2.1. Occurrence and sources

Published articles on MPs in freshwater has increased dramatically over the last decades. The abundance of MPs in freshwater environments is commonly reported in units of 'particles', items', 'pieces' and 'numbers'. A limited number of researchers have used the weight of MPs to determine their abundance (Li et al., 2020a; Yang et al., 2021). As a visible contaminant, it is easy to acquire an intuitive understanding of MPs abundance when using the units mentioned above (He et al., 2020b). However, it is noteworthy that these units easily result in inaccurate assessment of the extent of MPs contamination. Field sampled MPs can be extremely fragile due to degradation, and thus easily disintegrate into smaller fragments during separation and identification processes (Alimi et al., 2022; Manzoor et al., 2022), which result in over-estimation of MPs abundance. For example, as reported by He et al. (2020b), when MPs are reported as the unit of number, Brisbane River is highly polluted compared to most freshwater environments;

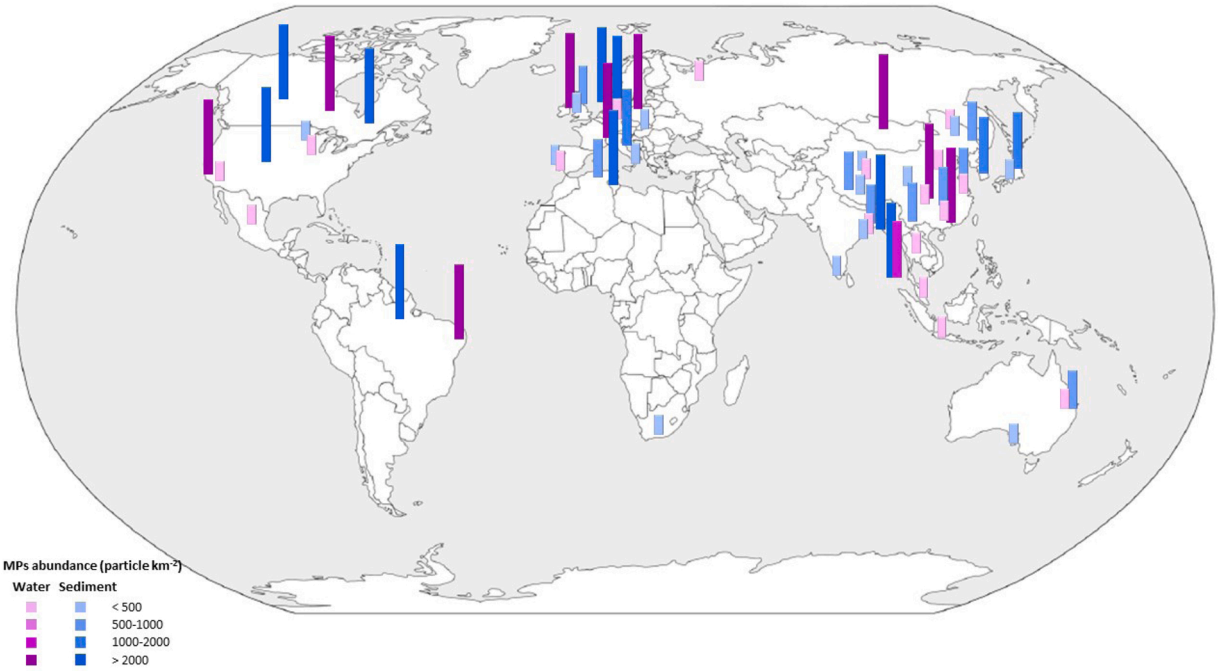

Fig. 2. Geographical distribution of MPs abundance in freshwater resources based on published studies (Abidli et al., 2017; Anderson et al., 2017; Ballent et al., 2016, 2013, 2012; Belontz et al., 2022; Bian et al., 2022; Bordos et al., 2019; Chen et al., 2021; Choong et al., 2021; Corcoran et al., 2020; Correa-Araneda et al., 2022; Costa et al., 2011; Dai et al., 2022; Devereux et al., 2022; Eibes and Gabel, 2022; Eriksen et al., 2013; Fahrenfeld et al., 2019; Faure et al., 2012; Free et al., 2014; Gerolin et al., 2020; Hayes et al., 2021; He et al., 2020b; Horton and Dixon, 2018; Hossain et al., 2022; Jiang et al., 2019; Kabir et al., 2022; Kapp and Yeatman, 2018; Karthe et al., 2017; Kiss et al., 2021; Klein et al., 2015; Liu et al., 2021; Napper et al., 2021; Nel et al., 2017; Oo et al., 2021; Peng et al., 2018; Sulistyowati et al., 2022; Tsering et al., 2021; Wu et al., 2022; Xiong et al., 2022; Yan et al., 2019; Yin et al., 2022; Yuan et al., 2022, 2019; Zhang et al., 2018; Zhdanov et al., 2022). (Note
: MPs with the unit of numbers are reported in this figure).

whereas if the weight of MPs was considered as the unit, Brisbane River is lightly polluted by MPs compared to past studies. Additionally, in the case of MPs of the same type, size and shape, but with different aging status, the weight can vary among different particles, since the molecular weight can alter due to fragmentation, degradation, and biofilm accumulation when exposed to the field environment. In view of the above issues, the weight is recommended in future research as the unit to determine MPs contamination, particularly for investigating toxicity of MPs. However, when research studies are conducted for environmental detection of MPs, it is acceptable to use 'particles', 'items', 'pieces' or 'numbers'. In this review, in order to avoid incompatibility between results on MPs contamination in different published studies, MPs abundance is reported using numbers between the water column and sediments. As shown in Fig. 2, the concentrations of MPs in the water column are generally higher than in sediments. This is not surprising as MPs have a lower density than water, which enables them to easily float in water rather than depositing in benthic sediments. For example, in Lake Geneva, Switzerland, 48,146 items km^{-2} of MPs were detected in water, while only 9 items km^{-2} was measured in sediments (Faure et al., 2012). In Qinghai Lake, China, the loads of floating MPs were up to 757, 500 particles km^{-2}, whereas only 1292 particles km^{-2} were found in sediments (Xiong et al., 2018).

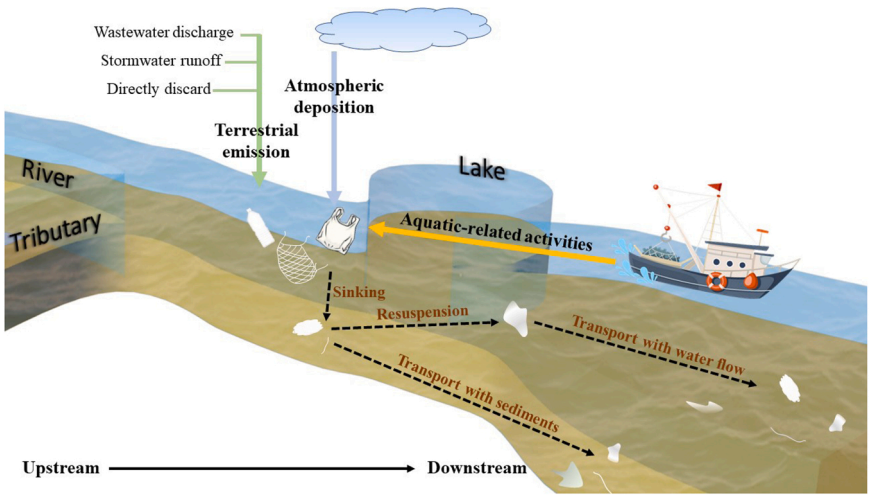

Fig. 3. Conceptual diagram illustrating the major sources of MPs to waterbodies.

Table 1
Overview of commonly used additives in plastic materials (Hahladakis et al., 2018; Hansen et al., 2013; Lithner et al., 2011).

Categories	Functionality	Example substances	Hazard effects	Mainly used in type of plastics
Functional additives	Antioxidants	–	–	For all plastic products
	Antistatic agents	–	–	For all plastic products
	Antimicrobial agents	Bis(tributyltin)oxide	Carcinogenicity, germ cell mutagenicity, reproductive toxicity, toxic/very persistent	Polyurethane (PUR)
	Curing agents	Formaldehyde	Skin sensitisation, oral/dermal/inhalation toxicity, carcinogenicity	Melamine-formaldehyde resin
	Flame retardants	Phthalic anhydride, bisphenol	Respiratory/skin sensitisation, oral/dermal/inhalation toxicity, reproductive toxicity, serious eye damage/eye irritation	Thermoplastics, Polycarbonate
	Plasticizers	Benzyl butyl phthalate, dibutyl phthalate, benzenedicarboxylic acid	Reproductive toxicity	Polyvinyl chloride (PVC), PET
	Slip agents	Lead stearate, zinc stearate	Respiratory/skin sensitisation, reproductive toxicity,	PE, PVC
	Stabilizers	Cadmium and lead compounds, nonylphenol compounds, bisphenol A	Respiratory/skin sensitisation, serious eye damage/eye irritation, reproductive toxicity, specific target organ toxicity	PVC
Colorants	Pigments	Cadmium compounds, chromium compounds, lead compounds	Germ cell mutagenicity, acute toxicity, respiratory/skin sensitisation, specific target organ toxicity, hazardous to the aquatic environment	PE, PVC
	Fluorescent brightening agents	–	–	PE, PP, PET, PVC
Fillers	Barium sulphate Calcium carbonate Clay Kaolin Metal powder Mica Talc Zinc oxide	–	–	For all plastic products
Reinforcements	Carbon fibres Glass fibres	–	–	For all plastic products

Different from the distribution pattern of MPs in the water column, the spatial distribution of sedimental MPs varies greatly in different aquatic environments worldwide. For lake sediments, the highest abundance of MPs was reported in Lake Ontario, Canada, reaching 27,830 particles kg^{-1} (Ballent et al., 2016), followed by 18,000 particles kg^{-1} in Bizerte lagoon, Tunisia (Abidli et al., 2017). While for river sediments, relatively high abundance of MPs was detected in Thames River, Canada, ranging from 6 to 2444 particles kg^{-1} (Corcoran et al., 2020), whereas only 0.46–1.62 particles kg^{-1} MPs in Carpathian basin ponds, Hungary (Bordos et al., 2019), 10–52 particles kg^{-1} in Brisbane River, Australia (He et al., 2020b), and 0–78 particles kg^{-1} in Po River, Northern Italy (Nel et al., 2017), respectively.

Most researchers attribute the heterogeneity of MPs abundance in aquatic systems to catchment characteristics, since terrestrial emissions via stormwater runoff and wastewater discharge are considered as major contributors of MPs to aquatic environments (Blettler et al., 2018; Freeman et al., 2020; He et al., 2020c), followed by aquatic-related activities (such as fisheries and aquaculture) and atmospheric deposition (Waldschlager et al., 2020) (Fig. 3). Moreover, considering the differences in physicochemical properties of MPs, namely, type, size, shape and colour, researchers have attempted to link MPs characteristics to specific sources in order to interpret the source-fate processes of MPs (Fahrenfeld et al., 2019). For example, polyethylene (PE), polypropylene (PP), polyamide (PA), and polyethylene terephthalate (PET) are the most commonly observed MPs in both, the water column and sediments (Blettler et al., 2018; Li et al., 2020a). Commercial activities significantly contribute to the occurrence of MPs such as PE, PP and PET, as they have the largest market share for food packaging and single-use plastic products. Polyamide (PA) fibres primarily originate from synthetic clothing and emissions from domestic wastewater discharge (Cesa et al., 2017; Gago et al., 2018). Rubber particles (commonly considered as MPs), can originate from road wear and vehicle tyre wear and transported via stormwater runoff and discharged into receiving waters (Baensch-Baltruschat et al., 2020; Wagner et al., 2018). However, though the concept of physicochemical-based source tracking has been widely adopted to determine the contribution of various sources on MPs in freshwater environments, such methodology is not always reliable due to the complex behaviour of MPs in aquatic systems as described in Section 2.2.

2.2. *Movement behaviour*

According to available data, an aquatic environment with relatively lower flow velocity would retain more MPs (Collins and Hermes, 2019). This is because, when water flow changes from high to low, MPs would be deposited in sediments rather than being transported downstream (Matsuguma et al., 2017). For MPs under the similar hydrodynamic conditions, the physiochemical characteristics such as shape and density would dominate their movement behaviour. For example, irregularly shaped particles are more likely to be drawn down from the surface into the sediment bed when compared with linear and spherical particles (Ballent et al., 2012), while high-density particles will infiltrate deeper compared to those with relatively lower density (Waldschlger and Schüttrumpf, 2020). Additionally, deposited MPs can be further buried in the core sediments or resuspended in the water column and transported by water flow when hydraulic conditions and particle physicochemical properties change. For example, an increase in density of MPs would facilitate deeper deposition, while a decrease in density would make the particles more prone to resuspension (Kooi et al., 2018) (Fig. 3). The relatively long-life span of MPs increases the frequency of deposition and resuspension processes, as well as the possibility of being transported for a considerably long distance from the emission source. In this context, it is difficult to confirm whether the MPs detected in the water column and sediments are freshly released from offshore sources or have originated from pre-deposited particles. These findings further imply that after being discharged to an aquatic environment, the influence of the combined effects of hydrodynamic conditions (such as water depth, flow velocity, salinity), environmental factors (such as wind and sediment clay) and physicochemical properties (such as density, size and shape) on MPs presence should be emphasized rather than only focusing

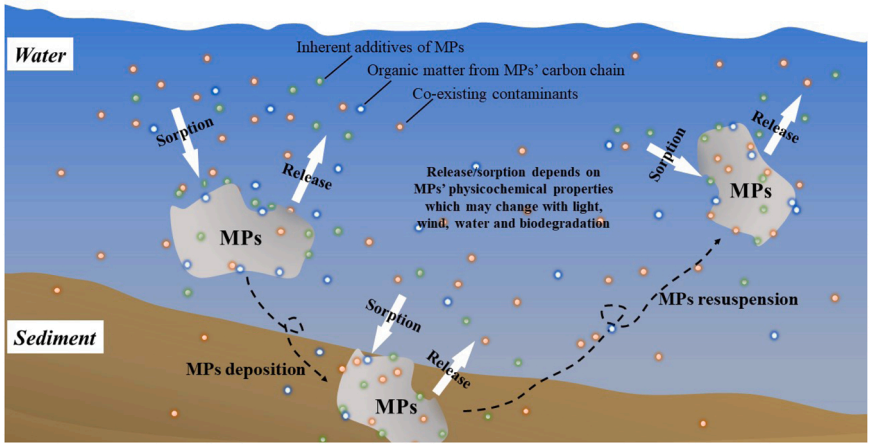

Fig. 4. Conceptual diagram illustrating the release and sorption of MPs.

on anthropogenic activities to further explicate the hydrodynamic behaviour MPs in rivers, lakes, and ponds.

Currently, though experimental studies and theoretical modelling approaches offer an analytical approach for source tracking and predicting the transport pathways of MPs in aquatic systems (Waldschlaeger and Schüttrumpf, 2019a, 2019b), it should be noted that such methodologies are based on the principles of classical sediment transport. Compared with natural sediment particles, the density, shape and size of MPs can be significantly altered by biofilm colonization (Kaiser et al., 2017), interaction with sediment clays (Li et al., 2019b), and degradation processes (Kowalski et al., 2016). These factors would consequently result in totally distinct movement behaviour of MPs compared to natural sediment particles within fluvial environments. However, very little is known about how and to what extent these factors influence the mobility of MPs in different aquatic environments. Consequently, this gives rise to the important issue of how to infer the current age of particles in an aquatic environment by identifying the accumulated biofilm on their surface, and to determine the effects of accumulated biofilm on their movement behaviour. Another important knowledge gap in relation to MPs movement behaviour is the critical thresholds of hydraulic parameters such as shear stress and flow velocity that enable different MP particles to start to move, deposit, and resuspend. Hence, in order to gain an in-depth understanding of MPs origination and transportation in a fluvial environment, factors including, the duration of presence, biofilm colonization, and degradation trends of different MPs when they are exposed to different aquatic environments should be investigated for accurate source tracking and simulation of transport behaviour.

3. MPs as complex environmental pollutants

MPs are complex environmental pollutants as they contain a wide range of toxic chemicals to meet the functions that are desired for a certain application of the original plastic materials (Lithner et al., 2009; Teuten et al., 2007, 2009). The additives used in plastic materials can be primarily divided into four categories, namely, functional additives, colorants, fillers, and reinforcements (Hahladakis et al., 2018). Table 1 presents a brief description of the substances and applications of the aforementioned additives. These additives are not chemically bound to the plastic materials and can thus be released from the material matrix to ambient substrates (Bridson et al., 2021) (Fig. 4). In addition to toxic leachate, the hazard impacts of MPs also extend to the sorption capacity of co-existing contaminants (Fig. 4). Currently, studies on the release and sorption mechanisms between MPs and toxic contaminants are mainly focused on metals and organic contaminants in aquatic systems (Binda et al., 2021; Sun et al., 2019). Moreover, related studies are basically for determining the contribution of factors in relation to MPs physicochemical characteristics (Bridson et al., 2021). For other factors, such as the properties of water and sediments, few systematic studies have been conducted. In this context, this paper discusses the release and sorption capacity of MPs with regards to metals and organic contaminants mainly in the context of changes to MPs physicochemical properties which occurs over time.

3.1. Release of toxic compounds

3.1.1. Metals

The release of metals typically occurs when the physicochemical properties of MPs change during the degradation process (Fig. 5). Cu, Cr, Pb and Zn are the most frequently reported metals from past bench leaching experiments of plastics (Brennecke et al., 2016; Holmes et al., 2014). For example, Nakashima et al. (2012) reported the leaching mass of Pb with an estimated 23 ± 11 g from PE, 6 ± 4 g from PP, and 284 ± 247 g from PVC, respectively, within 120 h. Boyle et al. (2020) measured approximately 84.3 ± 8.7 μg Pb being released from 500 mg PVC L^{-1} in aqueous medium after a 24 h bench experiment. In an in-situ

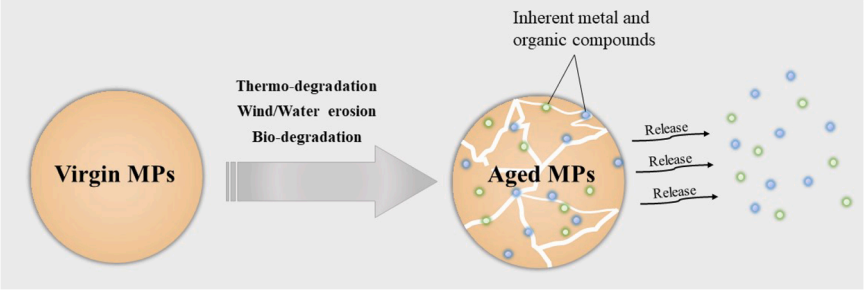

Fig. 5. Schematic illustrating the release of metal and organic compounds from MPs.

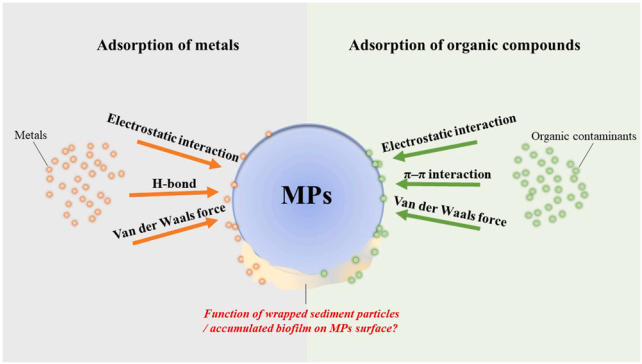

Fig. 6. Schematic illustration of major interaction mechanisms the sorption of metals and organic contaminants by MPs in aquatic environments.

survey by Nakashima et al. (2012) at Ookushi Beach, Japan, considerable leachate of Cr, Cd, Sn, Sb, and Pb were detected from the collected plastic litter.

The release mass for a given metal element from different MPs vary with their proportions for a particular plastic species. However, it should be noted that the release characteristics such as release rate for a certain metal element over a specific time duration can significantly change over time with changing physicochemical properties such as size and the degree of degradation of the plastic particles. According to Bandow et al. (2017), exposure to 1000 h of photooxidation resulted in proportionately much lower Cu release from a certain MP type compared to after 2000 h. Similarly, Luo et al. (2020) confirmed that a longer photooxidation time promoted a higher rate of Cr and Pb leaching from MPs. This is because of aging-induced fracture of chemical bonds which result in the deterioration of the mechanical stability of MPs materials. This causes polymer chain interruption and reduces the polymer molecular weight (Binda et al., 2021)which enhances the interchange of metal ions and allows the ions situated in the deep portions to become more accessible for extraction and leaching (Bandow et al., 2017). In addition to the degree of degradation, MPs particle size also influences the rate and mass of metal ions released. For smaller-sized MPs, higher surface area to volume ratio can accelerate metal ions diffusion within the polymer matrix and thus, enhance the leaching of metals (Bridson et al., 2021). In this context, MPs act as an additional emission source of metals in aquatic environments. As different plastics contain different metal species and amounts, quantifying the release capacity such as the critical parameters when metal release commences, especially those identified as toxic metal species for different sizes and aging levels would greatly benefit risk assessment in relation to both, MPs and MPs-related metals. However, current studies on source evaluation of metals in aquatic environments primarily focus on the correlation with sediment physicochemical properties and anthropogenic activities (Miranda et al., 2021; Wijesiri et al., 2019). Knowledge on the impact of different MPs on the magnitude and types of toxic metals in aquatic environments is still scarce. This knowledge gap constrains the accurate assessment of the source of origin and the hazards posed by specific toxic metals, based on MPs occurrence. Therefore, further studies for the investigation of indicator metal species for different MPs types are recommended to enable the development of a conceptual model in order to interpret and predict the MPs-metals interaction from a risk assessment perspective.

3.1.2. *Organic compounds*

In addition to toxic metals, significant past efforts have focused on detecting leachate of various organic compounds that are used as additives in different plastics which are of major toxicological concern (Cheng et al., 2020a). Similar to metals, the leaching behaviour of organic compounds from MPs also depends on the physicochemical properties, namely, type, size and degradation level of the plastic particles (Ricardo et al., 2021) (Fig. 6). Glass transition temperature of plastics, which indicates the temperature at which plastics change from being rubbery to glassy, is the paramount factor that influences the desorption capacity of organic compounds from MPs (Ateia et al., 2022; Cheng et al., 2020a). The diffusion model derived by Sun et al. (2019) characterised the releasing behaviour of flame retardants from millimetre-sized waste plastic granules through molecular diffusion. The research outcomes from the study confirmed that the diffusion coefficients of organic additives originating from plastics with relatively lower glass transition temperature can be much higher than those with higher glass temperature. This is because, plastics with low glass transition temperature have higher molecular mobility, which allows additives to move easily from inside the plastic particle to the surface (Cheng et al., 2020a). Further, Chen et al. (2019) found that small sized MPs yielded 1093.0 ± 1607.5 μg kg^{-1} organic endocrine disrupters, which was significantly higher than those in the medium sized (250.0 ± 445.3 μg kg^{-1}) and large-sized (270.6 ± 557.6 μg kg^{-1}) plastic

Table 2
The adsorption of heavy metals by MPs (Note: "√" refers tested MPs type).

Heavy metals	MPs type	Physicochemical properties of MPs				Adsorption capacity (ug g^{-1})	Reference
		Shape	Size (mm)	Virgin	Aged		
Ag	PE	–	0.18 0.30	√	√	3510 (For virgin MPs) 2440 (For aged MPs)	(Kalčíková et al., 2020)
As(III)	PTFE	Pellets	–	√		≤ 1050	(Dong et al., 2019)
Cu(II)	PS and PET	Pellets	0.025 0.18		√	67–102 mg g^{-1}	(Wang et al., 2022)
	PET	Piece	1.0	√	√	40.3–55.1 (For virgin MPs) 140.3–182.6 (For aged MPs)	(Wang et al., 2020a)
	PS	–	0.0001 0.02	√		20080 (MPs with 0.0001 mm diameter) 16430 (MPs with 0.02 mm diameter)	(Qiao et al., 2019)
Cr(IV)	PE	Spheres	0.15	√		3.82	(Liao and Yang, 2020)
	PP	Spheres	0.15	√		3.81	
	PVC	Spheres	0.15	√		3.50	
	PS	Spheres	0.15	√		5.07	
	PLA	Spheres	0.15	√		2.88	
Pb(II)	PA	Rope	2.0		√	1030	(Tang et al., 2020)
	PE	Pellets	2.0–4.0		√	~ 0.05	(Turner et al., 2020)
Cd(II)	PE	–	–	√		113.5	(Guo et al., 2020)
	PP	–	–	√		123.6	
	PS	–	–	√		134.1	
	PVC	–	–	√		151.4	
Zn	PET	Piece	1.0	√	√	24.7–44.5(For virgin MPs) 54.7–86.8 (For aged MPs)	(Wang et al., 2020a)

particles. This can be attributed to the fact that MPs with high surface area to volume ratio would accelerate the release of organic compounds (Sun et al., 2016). Furthermore, basic water quality parameters such as pH, temperature and salinity can also influence the release of organic compounds from MPs. For example, the amount of BPA (bisphenol A) released from PE and PC has been observed to increase with increasing pH and temperature due to enhanced ionization of BPA molecules in alkaline conditions (Liu et al., 2019). This is also in agreement with the findings by Biedermann-Brem et al. (2008) who detected that the extracted BPA mass increased as the pH and temperature increased in the aquatic environment. Conversely, an increase in salinity would be expected to inhibit BPA release from MPs due to the salting-out effect (Liu et al., 2019).

Currently, though the release of organic contaminants from MP particles in aquatic environments is a particular concern, past leaching experiments have primarily focused on a limited number of compounds, such as brominated flame retardants (Cheng et al., 2020b; Sun et al., 2019) and endocrine disrupting chemicals (Chen et al., 2019; Shabbir et al., 2022; Trujillo-Rodríguez et al., 2021). Moreover, reported data are typically obtained in laboratory-scale experiments by selecting representative parameters for a specific environment. However, in the real-world aquatic environment, environmentally relevant factors such as temperature, pH, salinity and flow velocity would have significant impacts on the leaching of organic compounds, since these factors can alter the physiochemical properties of MPs, as well as the functional groups of organic compounds. However, very limited knowledge exists in relation to the combined effect of environmentally relevant factors on the rate and extent of organic compounds released from MPs. Hence, comprehensive evaluation of the diffusion coefficients and leaching rate of organic contaminants from MPs by taking into consideration the complexity of the natural environment factors including pH, salinity, and media properties merits further research.

3.2. Sorption of existing contaminants in the surrounding environment

3.2.1. Metals

The sorption capacity of MPs is available for a variety of metals as shown in Table 2. Adsorbed metal content on MPs surface varies with the metal species and MPs type. For example, the sorption capacity of Pb was found to be 1030 ug g^{-1} for PA MPs, whereas it is only about 0.05 ug g^{-1} for PE MPs. However, for MPs such as PE, 3510 ug g^{-1} of Ag, 40.3–55.1 ug g^{-1} of Cu (II), and 3.82 ug g^{-1} of Cr (IV) have been found to be adsorbed (Table 2). This can be explained by the different interaction mechanisms between different MPs and metals. The sorption mechanism for metals on MPs is primarily electrostatic interactions (Ding et al., 2022) (Fig. 6). Since most plastics are generally negatively charged (such as PVC, PS, PP and PE) (Ren et al., 2021), the change of MPs oxidation status caused by the degradation process would enhance the electrostatic attraction between charged sites on surfaces and metal ions (Cao et al., 2021). For example, by examining the sorption of metals (Cd, Co, Cr, Cu, Ni, Pb) to both, virgin and aged MP pellets in river water and sea water, Holmes et al. (2014) demonstrated that aged plastic pellets have considerably greater adsorption capacity for all tested metals compared to virgin pellets. A laboratory-scale experiment conducted by Turner and Holmes (2015) also confirmed that the adsorption of trace metals (Ag, Cd, Co, Cr, Cu, Hg, Ni, Pb, Zn) to aged plastic pellets was greater than to virgin pellets. This can be explained by the fact that the deterioration of the plastic matrix due to weathering can increase the surface area of micro-sized fractures. This in turn results in a larger area for interaction and higher adsorption capacity for metals (Bhagat et al., 2021a; Kalčíková et al., 2020; Singh et al., 2021).

Environmentally relevant parameters such as pH, temperature and salinity can also influence the interaction between metals and MPs by impacting the surface properties (Binda et al., 2021; Rozman and Kalcikova, 2022). Increased pH enhances the sorption capacity of MPs for metals as the low concentration of hydrogen ions would not compete with cationic metals for the adsorption sites (Binda et al., 2021). Compared with pH, salinity shows a negative influence on metal-MPs adsorption behaviour. This is because, the increased salinity will on the one hand lead to high competition between Na^+ and metal ions adsorption on MPs surfaces, and Cl^- on the other hand can enhance complexes with dissolved metal ions. This will result in lower activity and accessibility of free metal ions to MPs binding sites (Fu et al., 2021; Huang et al., 2021). Additionally, MPs are recognised as a suitable substrate for biofilm growth and accumulation (Baptista Neto et al., 2019). The biological film adhering to MPs enhances surface wettability and thus, modulates the plastic-metal interaction (Binda et al., 2021; Johansen et al., 2019; Wang et al., 2020b). According to published data, MPs with relatively low toxic additives such as PE and PP would encourage greater biological growth than those with more toxic compounds such as PVC (He et al., 2020a). As such, the more biofilm is accumulated, the higher the degradation rate of MPs, which can then lead to greater release of toxic chemical compounds, namely, metals and organic compounds. On the other hand, greater biofilm accumulation on MPs surface would facilitate the sorption of co-existing contaminants from the surrounding environment as more functional groups are established. It is therefore reasonable to hypothesise that within a certain degradation time period, MPs with low toxic additives could pose a relatively high risk to the surrounding environment due to the combined effects of accumulated biofilm. Unfortunately, the role of biofilm on the release and adsorption of co-existing contaminants in relation to MPs remains unclear. This knowledge gap constrains not only the robust evaluation of the outcomes of studies on source tracking and distribution patterns of metals through MPs movement, but also the risk assessment from the synergistic effects of MPs bonded contaminants. Consequently, this is an important area that merits further research.

Furthermore, current studies in relation to the release or sorption capacity of MPs has been analysed separately. However, such processes can occur simultaneously in the natural environment. Hence, determining the dominant process of MPs-induced occurrence of metals in an aquatic environment would greatly benefit in identifying sources, understanding pollutant mitigation and improving water and sediment quality from the perspective of ecological risk assessment. Additionally, since MPs can be easily ingested by aquatic fauna, the co-existence of MPs and MPs mediated metals would affect the bioavailability and toxicity to aquatic organisms. Hence, accurately determining the contributions of various MPs to the abundance of toxic metals in the natural aquatic environment is vital for biotoxicity assessment.

3.2.2. Organic compounds

Organic compounds have relatively higher sorption capacities for MPs compared to metals owing to their diverse functional groups (Ricardo et al., 2021). Electrostatic forces, hydrophobic interactions, and non-covalent bonds are the three main sorption mechanisms in the adsorption of organic compounds by MPs as illustrated in Fig. 6 (Ding et al., 2022). Similar to the interaction between metals and MPs, aged MPs would have a greater sorption capacity for organic contaminants as the increase in the surface area during degradation would result in increased number of oxygen-containing surface functional groups (Rai et al., 2022). For example, adsorption of tetracycline increased from 0.90 to 1.97 mg g^{-1} for polylactic acid (PLA), and 0.75–1.36 mg g^{-1} for PVC with ageing due to UV radiation (Fan et al., 2021). Further, results derived by Li et al. (2019a) and Ma et al. (2019) confirmed that the sorption capacity of triclosan (TCS) for both, polystyrene (PS) and PVC increase with decreasing particle size due to the relatively larger surface area in the case of smaller particles.

Current studies on factors that influence the interaction between MPs and toxic organic pollutants primarily focus on the degree of MPs degradation, species and background concentrations of the adsorbates (Ricardo et al., 2021). Based on available knowledge, for non-planar structure MPs such as PE, mechanism in the adsorption of organic compounds is mainly electrostatic interaction, while for planar MPs such

as PS, their sorption capacity for organic chemical components is attributed to π–π interactions (Bakir et al., 2012; Lee et al., 2014; Wang et al., 2018). It is thus reasonable to hypothesise that different functional groups in plastics would result in totally distinct interaction mechanisms between MPs and adsorbed organic chemical contaminants. As such, a clear understanding of such heterogeneity among different types of MPs can contribute to predicting the species content of typical toxic organic contaminants based on the presence of different MPs. However, there is paucity of systematic research data available to qualitatively determine the sorption capacity of organic compounds among different MP species, therefore meriting further research. Additionally, in a real-world aquatic environment where numerous substances such as sediment clays and plant litter co-exist, the adsorption capacity of organic contaminants on MPs can be changed by reactive media. For example, for sediment-wrapped MPs, the molecular interactions between MPs and adsorbates are inhibited by reduced accessible surface area regardless of their physicochemical status. However, there is limited knowledge in relation to the magnitude and type of such adsorption processes between MPs and adsorbates, taking into consideration the complexity of the conditions in natural aquatic environments. Therefore, further research is recommended to enable the reliable assessment of aquatic toxicity posed by the co-existence of MPs and organic contaminants.

4. Aquatic toxicity associated with MPs occurrence and mobility

Plastics have been manufactured to be durable, which allows them to remain and pose long-term hazards to aquatic environments and organism for many years. A high number of aquatic organisms are known to be subjected to irreversible toxicological effects including nutrition loss, physical damage or even death (Nava and Leoni, 2021). However, due to the complexity of MPs presence and settling mechanism in freshwater systems, it is difficult to relate the specific contaminants to their sources or pathways. To improve knowledge on the hazards of MP-associated impacts to water and sediment quality and fauna in freshwater ecosystems, it is important to build a framework which considers the migration pathways and potential risks as a whole.

4.1. Environmental risks

MPs present an environmental risk to aquatic environments through two ways, namely, MPs alone and with associated contaminants. Regardless of land-based MPs discharge, the continuous breakdown and degradation of existing larger plastic debris into smaller size plastics is regarded as the main source of MPs pollution in aquatic systems (Hidalgo-Ruz et al., 2012; Reisser et al., 2013). The fragmentation of plastic debris leads to the wide occurrence of MPs in an aquatic environment. Specially, for floating MPs, which are easier to be transported by water flow, it is important to consider the environmental risk posed by their longitudinal dispersion, as the aggregation of MPs is highly affected by chemical conditions such as pH and salinity of water. For example, when floating MPs are transported from inland freshwater to estuaries, the increased pH and salinity would reduce the electrostatic repulsion and prompt outer-sphere surface complexation between MP particles, consequently increasing MP aggregation (Ateia et al., 2022). This further induces MPs-related radiative effects by absorbing and scattering radiation (sunlight rays can change direction when coming into contact with objects such as MPs) (Revell et al., 2021), which can further influence water temperature, nutrient concentration and fauna structure in aquatic systems. However, to-date, very little is known about MPs-radiation interactions in aquatic systems. This lack of knowledge constrains the accurate evaluation of the environmental consequences of floating MPs on the aquatic environment. In this context, studies on the radiative effects of floating MPs based on particle size, shape, colour and surface conditions in various aquatic environments are recommended.

Flow velocity and particle density are known as the major factors that influence the downward movement of floating MPs (Enders et al., 2015; Kukulka et al., 2012; Reisser et al., 2013). As in ponds and lakes which have low water flow, MPs would be prone to be retained for relatively long periods as they are not likely to be transported downstream by water flow. In this context, most plastic particles would settle down and deposit in benthic sediments over time. These settled plastic particles can later disintegrate into smaller sizes with leaching and adsorption of toxic compounds as discussed in Chapter 3. This in turn could influence the population size of benthic organisms such as macroinvertebrates and algae (Li et al., 2022; Silva et al., 2022), alter the biological community structure in benthic habitats, and consequently influence biogeochemical cycles and nutrient concentrations in aquatic environments (Eerkes-Medrano et al., 2015). As such, it is logical to hypothesise that MPs in aquatic systems with low water velocity would exert more adverse influence on environmental deterioration compared to riverine and marine systems. Specially, plastic particles such as PVC, PET and PUR with high density and associated hazardous compounds resulting in toxic leachate could lead to relatively more severe environmental consequences when compared to PE and PP particles. However, there is limited data available to assess such ecological impacts of MPs among different aquatic systems. This knowledge gap is a significant constraint to risk assessment of MPs occurrence in different aquatic systems and therefore merits further investigation.

Longitudinal transport via water and sediment that are driven by wave, wind, and tide, results in the ubiquitousness of MPs in different aquatic environments worldwide (Mai et al., 2020; Yuan et al., 2022). Rivers are commonly regarded as the main pathway for the transport of MPs from the inland to the marine environment. Therefore, the river-sea transport route is a topic of importance (Besseling et al., 2017; Collins and Hermes, 2019; He et al., 2021). Generally, river catchments with high population density will receive relatively more plastic contaminants from various inland emission sources and introduce greater MPs loads to other receiving water environments including the marine environment based on hydrodynamic forces. However, considering the diverse transport distances among different riverine systems, the extent to which MPs distribution and transportation processes are influenced by hydrodynamic parameters remains largely unknown. Additionally, modelling studies on marine MPs behaviour revealed vertical movement under the influence of tidal forces (Ballent et al., 2012; Collins and Hermes, 2019; Genc et al., 2020). Accordingly, floating MPs in the marine environment would be prone to be transported to inland aquatic systems under particular conditions of forcing parameters such as wind, flow velocity, turbulence, and pressure. However, the migration mechanism such as how much time and how quickly a particular MP particle can be transported in relation to the re-entering process of marine-MPs to inland aquatic environments remains a pending research task. These knowledge gaps can confound the evaluation of the environmental risk posed by MPs from different origins and pathways, and warrants further research.

4.2. Population-level risks to aquatic organisms

MPs can act as ecological habitats for aquatic organisms (Derraik, 2002; Lambert et al., 2014; Zettler et al., 2013). MPs can provide hard-substrates for laying eggs and rafting microbial communities (Goldstein et al., 2012; Gregory, 2009; Zettler et al., 2013). This can consequently influence the population-level of organisms in an aquatic system. For example, a study of the North Pacific Subtropical Gyre has shown that increased MPs pollution has resulted in an overall increase in *Halobates serious* egg densities (Goldstein et al., 2012). As the influence continues, the structure of pelagic and/or bacterial communities were altered accordingly (Anderson et al., 2016). Compared to most natural substrates, MPs provide more opportunities for microbial community colonization and biofilm formation due to their longer life and hydrophobic surfaces (Harrison et al., 2018), and thus act as facilitators for the spread of pathogens and other exotic species (Wagner et al., 2014;

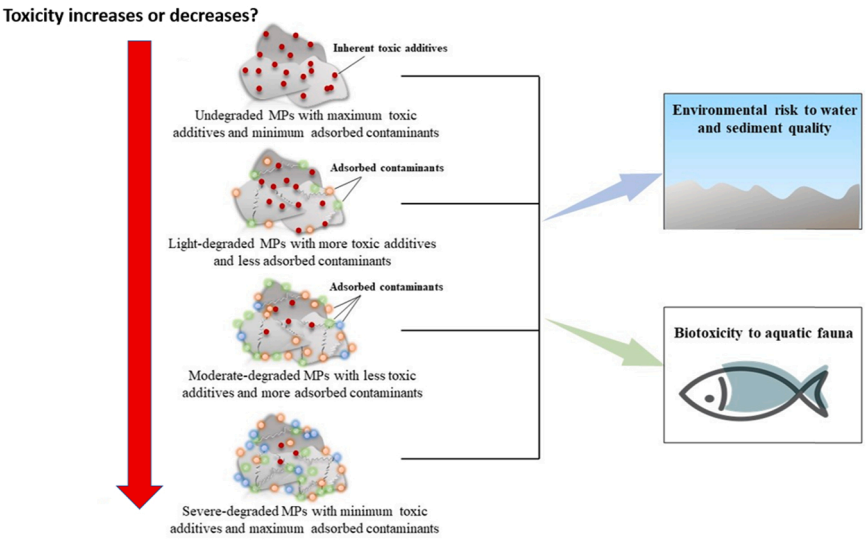

Fig. 7. Schematic illustrating the combined toxicity risk of contaminants associated MPs in aquatic environments.

Zettler et al., 2013). According to the research undertaken by Eckert et al. (2018), a significant relationship exists between the increase in MP concentrations and the relative survival rate and richness of microbial community in both, wastewater and natural lake water. In addition to aquatic fauna, MPs could adversely affect the physiological function of aquatic flora (Ge et al., 2021). For instance, floating MPs would be trapped by phytoplankton, inhibiting photosynthetic activity and thus reducing their root growth (Long et al., 2017; Shiu et al., 2020). Additionally, studies have demonstrated that the energy transfer and substances exchange between cells of MPs-contaminated microalgae can be hindered and blocked, causing physical damage to plant structure and negatively affecting the growth of microalgae (Kalčíková et al., 2017; Yu et al., 2020).

The adverse impacts of MPs on aquatic organisms are mainly considered as physical and chemical effects. Since MPs would be retained for the entire life span of organisms after being ingested, they can block the cells, digestive tract, reduce photosynthetic activity, food intake and vigour as well as hinder the growth, mobility and reproduction and even cause death to both, aquatic plants and animals (Bhagat et al., 2021b). Moreover, the toxic chemicals and pathogens carried by MP particles would facilitate the detrimental impacts by translocating across cell membranes and entering into tissues (Bouwmeester et al., 2015; Wright and Kelly, 2017). However, to-date, most biotoxicity studies attribute the adverse effects of MPs on the health of aquatic organisms such as endocrine disorders, mobility retardation and reduced reproduction rate (Sharma and Chatterjee, 2017; Teuten et al., 2009) to MPs itself. It is rare for past studies to have focused on the combined effects of MPs and associated toxic compounds at the molecular level. For identifying the pathogenetic factor that is either posed by MPs themselves or contaminants associated with MPs, response dose, lethal dose, potency of concentrations, and dose levels of MPs and MPs-associated contaminants on different organisms such as whether the additives act independently or in combination requires in-depth investigation. Such knowledge is important since different MPs will affect organisms in different ways due to distinct chemical composition. However, this currently constitutes a knowledge gap. Hence, it is recommended that further research is undertaken to bridge this significant knowledge gap in relation to bioindicator identification of MPs for biotoxicity risk assessment.

5. Limitations of current risk assessment approaches

The risk assessment of MPs in aquatic systems can be analysed quantitatively and qualitatively. A comprehensive understanding of the combined risks posed by contaminants associated with MPs would facilitate the formulation of robust mitigation and management strategies for plastic contaminants. However, to the best of knowledge, the risk assessment approach in relation to MPs is restricted to numerical modelling that is based on hazard scores and concentrations of MPs (He et al., 2020a; Liu et al., 2022; Wang et al., 2021a; Xu et al., 2018). The specific procedure applied for risk assessment of MPs is outlined as (He et al., 2020a):

$$H = \sum P_n \times S_n$$

where H is the calculated hazard score of all detected MPs; P_n is the percent of a given MP type at each selected sampling site; S_n is the corresponding hazard score as determined by Lithner et al. (2011). However, the risk assessments for MPs derived from this approach should be used and interpreted with caution for a number of reasons.

Firstly, the risk assessment results from different studies may not be comparable due to the use of different units. Current studies quantify MPs abundance using one of two typical units, namely, number-based quantification (such as items, particles, number), and mass-based quantification (such as mg, g, μg). When the hazard score for a given MP type is determined, the use of different units used would result in different MPs concentrations. This in turn would result in inaccurate evaluation of MPs abundance. Consequently, this makes it difficult to achieve a realistic determination of the potential risks posed by the detected MPs. For example, though only a limited number of polyvinylidene fluoride (PVDF) and polyacrylonitrile (PAN) particles were detected in the Brisbane River sediments, Australia, the highest hazard grade was for the sampling sites where these two MPs species were detected (He et al., 2020a). This was because the study quantified MPs concentration as particle weight. However, if the units of PVDF and PAN were changed to numbers, the proportion of these two MPs species present would decrease to an extremely low fraction. Accordingly, the hazard scores would decrease. In other words, due to the fragile nature of field collected plastic particles, the hazardous risk of MPs with high numbers, but small particle weight can be inaccurate when MPs concentration is expressed as a unit of item.

Secondly, concentrations of polymer ingredients and changes in MPs physicochemical properties can be underestimated. The hazard scores of diverse polymers that were evaluated by Lithner et al. (2011) are one of the most important reference parameters for MPs risk assessment modelling. However, it should be noted that the hazard scores were calculated solely for hazardous monomers while the most hazardous

Table 3
Current research trends and identified research gaps of MPs in freshwater environments.

Research topic	Current research trends	Research gaps
Source tracking and transport of MPs	• Linking MPs physicochemical properties to catchment characteristics. • Modelling approach for MPs transport conducted based on classical sediment transport theory. • Parameters (e.g., density) for MPs transport modelling are primarily for virgin plastics rather than aged particles.	• How to identify whether the detected MPs are freshly released (e.g., terrestrial, atmospheric, aquatic-related sources) or originating from pre-deposited MPs in up-stream sections? • What are the critical thresholds for hydraulic parameters (e.g., shear stress and flow velocity) that enable different MP particles to start to move, deposit, and resuspend in different freshwater environments? • How to determine the age of detected MPs in surveyed aquatic environment based on their degree of degradation and biofilm accumulation? • What is the explicit impact of biofilm accumulation/sediment wrapping on different MPs density changes?
Release and sorption capacity of typical contaminants	• Influence of MPs physicochemical properties on the release and sorption capacity for metals and organic contaminants.	• What is the dominant process (release or sorption) of MPs with different characteristics (e.g., degree of degradation)? • What is the indicator metal/s and organic compound/s for different MPs presence? • How to differentiate whether the MPs bonded metal or organic compounds result from release or sorption process? • What is the role of biofilm/sediments attached to MPs on MPs and contaminants interaction?
Risk assessment approach for MPs	• Limited risk assessment approaches are available. • Current risk assessment of MPs is solely based on their type and abundance. • Current risk assessment of MPs only considers hazardous monomers while the more hazardous substances such as solvents, catalysts and raw materials have not been investigated. • Current risk assessment does not consider synergistic effects of MPs and other contaminants	• How to develop a risk assessment framework for contaminants associated MPs? • How to quantitatively determine the mixture toxicity (environmental risk and toxicology) of MPs by considering associated contaminants and MPs with different degradation status?

substances such as solvents, catalysts and raw materials were not investigated. Therefore, the calculated toxicity may not exhibit a linear relationship with the concentration of a particular MP type. Additionally, the hazard grade for each polymer type was only validated for substances that are used in virgin plastic products. For MP particles, specially, the secondary MPs, the fragmentation and degradation processes over time can lead to the loss of toxic compounds as discussed in Section 2.1. Hence, this can result in inaccurate assessment of risk posed by MPs. However, there is limited knowledge as to what extent the degree of degradation can influence the changes in the type and concentration of degraded plastic debris. Consequently, it is recommended that further studies should be undertaken to quantify the influence exerted by the degree of degradation on changes to hazardous components in various plastic types in order to provide essential data for robust risk assessment of different MPs under different physicochemical conditions.

Thirdly, the synergistic effects of MPs and other contaminants are commonly neglected. According to past studies on risk assessment of commonly available toxic contaminants in aquatic environments, the contaminants themselves, such as heavy metals (Duodu et al., 2017a; Jayarathne et al., 2020; Ma et al., 2020) and PAHs (Duodu et al., 2017b), would pose adverse impacts on aquatic environments and organisms. Interaction of MPs with various contaminants as described in Section 2.2 would result in mixture toxicity. The combined effects of two or more toxic materials (such as metal accumulated MPs particles) could be much greater than the effect of an individual toxic contaminant (Fig. 7). As discussed in Chapter 3, MPs with different degrees of degradation have different release and sorption abilities for toxic additives and co-existing contaminants, which can thus result in different risks to the aquatic environment and organisms. However, there is paucity of knowledge on the synergistic toxicity effects of MPs and MPs-borne contaminants. This is another important knowledge gap that merits deeper investigations in the future. Further, it is also not clear which conditions are more toxic: less-degraded MPs with more additives, but low adsorbed contaminants or more-degraded MPs with less additives but more adsorbed contaminants (Fig. 7)? This highlights another knowledge gap which merits further investigations.

6. Current research trends and knowledge gaps

Current research studies on aquatic MPs have established a meaningful theoretical basis for the investigation of MPs and biotoxicity assessment based on the available concentrations and types. Nevertheless, available knowledge for source tracking, transport and the toxicity of MPs is still far from comprehensive. Reasons for the differences in the interaction between MPs and contaminants are not clear. This constrains the in-depth understanding of the impacts of MPs and associated contaminants on the aquatic environment in entirety, as well as methodology development for assessing MPs. Accordingly, current knowledge gaps on MPs in freshwater based on existing research trends and deficiencies are summarised in Table 3. These findings are expected to not only contribute to creating a comprehensive knowledge base on current research into MPs, but also create a solid foundation for further research directions.

7. Conclusions

Urban aquatic systems are subjected to considerable MPs contamination. Land-based discharge, atmospheric deposition and aquatic activities are regarded as major contributors to MPs presence in water environments. The distinct physiochemical properties of different MPs types are commonly used for source tracking and interpreting transport patterns of MPs in aquatic environments. However, the combined effects of hydrodynamic parameters, and the physicochemical changes to MPs that occur either due to degradation or interaction with other substances such as sediment particles and microorganisms can alter the movement behaviour and fate of MPs. MPs are associated with numerous toxic compounds that either originate from their manufacturing process or from the sorption of existing contaminants in the surrounding environment. Therefore, it is challenging to associate the specific environmental consequences by linking the synergistic effect on the presence and

migration of MPs and other contaminants. This review systematically discusses the origin, abundance, movement behaviours, and interactions between MPs and typical contaminants, namely, metals and organic compounds from a risk assessment perspective, as well as identifies the current knowledge gaps and provides recommendations for further research. This review is expected to provide insights on the risk assessment approaches and mitigation strategies for MPs in freshwater environments inform researchers and decision makers.

CRediT authorship contribution statement

The presence of microplastics (MPs) are recognized as a significant environmental threat because they are complex environmental contaminants as they bind to a wide range of other contaminants. So far, only limited research has been undertaken to link the environmental risk with MPs occurrence and movement behaviour in aquatic systems. This review discusses the occurrence, sources and movement behaviours of MPs in urban waterways. Then, implications of MPs as a complex contaminant and current risk assessment approaches adopted were discussed. Finally, current knowledge gaps are identified and recommendations are provided for future research from a risk assessment perspective.

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.

Data availability

Data will be made available on request.

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