

# Interaction and combined toxicity of microplastics and per- and polyfluoroalkyl substances in aquatic environment

Yanhui Dai<sup>1</sup>, Jian Zhao (✉)<sup>1,2</sup>, Chunxiao Sun<sup>1</sup>, Diying Li<sup>1</sup>, Xia Liu<sup>1</sup>, Zhenyu Wang<sup>3</sup>,  
Tongtao Yue<sup>1</sup>, Baoshan Xing (✉)<sup>4</sup>

<sup>1</sup> Institute of Coastal Environmental Pollution Control, Key Laboratory of Marine Environment and Ecology (Ministry of Education),  
Frontiers Science Center for Deep Ocean Multispheres and Earth System, Ocean University of China,  
Qingdao 266100, China

<sup>2</sup> Laboratory for Marine Ecology and Environmental Science, Qingdao National Laboratory for Marine Science and Technology,  
Qingdao 266237, China

<sup>3</sup> Institute of Environmental Processes and Pollution Control, and School of Environmental and Civil Engineering, Jiangnan University,  
Wuxi 214122, China

<sup>4</sup> Stockbridge School of Agriculture, University of Massachusetts, Amherst, MA 01003, USA

## HIGHLIGHTS

- Adsorption of PFASs on MPs and its mechanisms are critically reviewed.
- MPs could alter the transport and transformation of PFASs in aquatic environments.
- Combined toxicity of MPs and PFASs at organismal and molecular levels is discussed.

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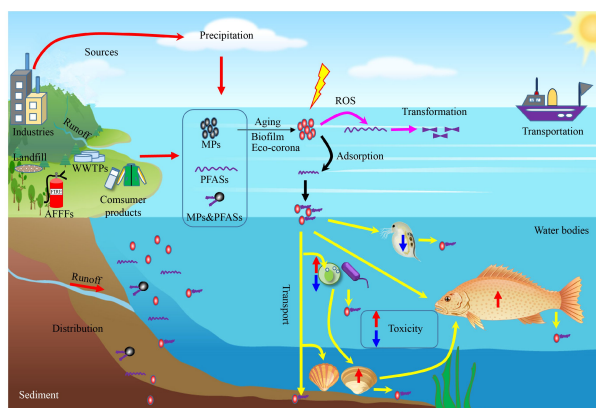
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## GRAPHIC ABSTRACT



## ABSTRACT

Microplastics (MPs) are recognized as vectors for the transport of organic contaminants in aquatic environments in addition to their own adverse effects on aquatic organisms. Per- and polyfluoroalkyl substances (PFASs) are widely present in aquatic environments due to their widespread applications, and thus coexist with MPs. Therefore, we focus on the interaction of MPs and PFASs and related combined toxicity in aquatic environments in this work. The adsorption of PFASs on MPs is critically reviewed, and new mechanisms such as halogen bonding,  $\pi$ - $\pi$  interaction, cation- $\pi$  interactions, and micelle formation are proposed. Moreover, the effect of MPs on the transport and transformation of PFASs in aquatic environments is discussed. Based on four typical aquatic organisms (shellfish, Daphnia, algae, and fish), the toxicity of MPs and/or PFASs at the organismal or molecular levels is also evaluated and summarized. Finally, challenges and research perspectives are proposed, and the roles of the shapes and aging process of MPs on PFAS biogeochemical processes and toxicity, especially on PFAS substitutes, are recommended for further investigation. This review provides a better understanding of the interactions and toxic effects of coexisting MPs and PFASs in aquatic environments.

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✉ Corresponding authors

E-mails: [jzhao@ouc.edu.cn](mailto:jzhao@ouc.edu.cn) (J. Zhao); [bx@umass.edu](mailto:bx@umass.edu) (B. Xing)

## 1 Introduction

Plastic pollution has drawn great attention as an emerging threat to the aquatic ecosystems (Borrelle et al., 2020; MacLeod et al., 2021). Estimates of the global emissions of plastic waste to aquatic ecosystems were 19 to 23 million metric tons in 2016, and the annual emissions will increase up to 53 million metric tons per year by 2030 (Borrelle et al., 2020). Plastics with sizes smaller than 5 mm were considered as microplastics (MPs), composed of different polymer types, such as polypropylene (PP), polyethylene (PE), polystyrene (PS), polycarbonate (PC), rubbers, and polyvinyl chloride (PVC) (Table S1). Generally, there are two different kinds of MPs, primary and secondary MPs. Primary MPs are defined as plastics with a microscopic size that are manufactured for use, such as cosmetic products, while secondary MPs are tiny plastic fragments derived from larger plastic debris during physical, biological and chemical processes in the environment (Cole et al., 2011). Upon release into the environment, MPs have shown deleterious effects on aquatic organisms (MacLeod et al., 2021). For example, MPs, serving as “a widespread poor food”, could be ingested by organisms (e.g., fish) (Li et al., 2021) and increase the starvation risk (Santos et al., 2021). Accordingly, MPs ingestion induced physiological disruption and the impairment of gastrointestinal functions, thus decreasing the health, growth, and reproductivity of aquatic organisms (de Sá et al., 2018; Rebelein et al., 2021). The toxicity of MPs to aquatic organisms has been previously reviewed (de Sá et al., 2018; Rebelein et al., 2021) and will not be specifically discussed in this review.

Poly- and perfluoroalkyl substances (PFASs) have been commonly used in various fields and products, such as waterproof materials, aqueous film-forming foams (AFFFs), and electronics (Xiao, 2017). Perfluoroalkyl acids (PFAAs), the most widely distributed types of PFASs in the environment, include perfluoroalkyl carboxylic acids (PFCAs), perfluoroalkyl sulfonic acids (PFSAs), perfluoroalkane sulfinic acids (PFSIAs), perfluoroalkyl phosphonic acids (PFPAs), and perfluoroalkyl phosphinic acids (PFPIAs) (Buck et al., 2011) (Table S2). Due to their ongoing emissions and persistent properties, PFASs have been widely detected in different surface waters and sediments (Pan et al., 2017; Lee et al., 2020b; Muir and Miaz, 2021). Their accumulation in and adverse effects on aquatic organisms have been investigated at both physiological and molecular levels (Lee et al., 2020c). The aquatic environment is considered as a main sink for emerging pollutants. MPs have migration pathways similar to those of PFASs from different environmental compartments (e.g., soil) to aquatic environments, and their interaction deserves systematic investigation. Recent studies reported that PFASs (e.g., perfluorooctanesulfonate (PFOS)) could be adsorbed on MPs (e.g., PE) (Wang

et al., 2015; Llorca et al., 2018), and hydrophobic interactions dominated the partitioning of organic pollutants into MPs (Atugoda et al., 2021). MPs types and environmental factors highly affect MPs-PFASs interactions. For example, biofilms on the MPs surface could supply sorption sites for PFASs (Bhagwat et al., 2021), but their roles in PFAS adsorption on MPs need systematic evaluation. In addition to adsorption, the presence of MPs may further change the transport and transformation of PFASs, which will be addressed in this review.

In addition to the interaction between MPs and PFASs in aquatic environments, MPs and PFASs could be released simultaneously from a given consumer product, such as textile with durable water repellent (Holmquist et al., 2016; Supreeyasunthorn et al., 2016), car seats (Wu et al., 2021), and carpet (Chen et al., 2020a), during the usage and washing processes. Upon these co-exposure pathways, the combined toxicity of MPs and PFASs has attracted increasing concern. Combined toxicity (e.g., antagonistic toxicity) to aquatic organisms was observed during the co-exposure of MPs and PFASs (Chen et al., 2020b; Yang et al., 2020). However, in-depth explanations for the observed contradictory results are still lacking. This is because our knowledge on the interaction and combined toxicity of MPs and PFASs is still fragmentary. Therefore, this review will focus on the adsorption of PFASs on MPs and related mechanisms, the effects of MPs on the transport and transformation of PFASs, and the individual and combined toxicity of PFASs and/or MPs. This review will fill in the knowledge gaps and advance our understanding of the environmental behavior and potential risk of both MPs and PFASs in aquatic environments.

## 2 Distribution and sources of MPs and PFASs in aquatic environments

### 2.1 Global distribution and sources of MPs

Currently, MPs are globally detected in different marine and freshwater environments. PP and PE were the most common polymer types in most surface waters, and fibers and fragments were the primary shapes found in the aquatic environment (Tables S3 and S4). The contents of MPs in seawater reached 362000 particles/m<sup>3</sup> (Chabahar Bay, Iran) (Hosseini et al., 2020) or 389 µg/m<sup>3</sup> (Atlantic Ocean) (Pabortsava and Lampitt, 2020). The high concentrations may be due to the improved sampling and analytical methods, which could detect very small particles. Additionally, the abundance of MPs in fresh water is much higher, up to 519000 items/m<sup>3</sup> in the Saigon River (Southern Vietnam) (Lahens et al., 2018), because of the contribution from textile and plastic industries and the untreated wastewater from the dense

urban districts and industrial zones. This suggests that the freshwater environment acts as an important sink for plastics from land to ocean. The highest abundance of MPs in the sediment of freshwater samples was 9597 items/kg (Pearl River, China) (Lin et al., 2018), which is much lower than that in marine sediment (33300 particles/kg) (South Pacific) (Bakir et al., 2020), illustrating that marine sediments might be the final fate of MPs.

The sources of MPs in aquatic environments can be divided into point sources and nonpoint (diffuse) sources. The point sources of MPs mainly include human activities (e.g., littering, synthetic textiles, coastal tourism, recreational and commercial fishing), transportation (e.g., rubber tires), and industry (marine vessels and marine industries, WWTPs, raw sewage discharge) (Cole et al., 2011; Wang et al., 2021a). For the nonpoint source, MPs derived from atmospheric deposition (Allen et al., 2019), runoff (e.g., agricultural and stormwater runoff) (Grbić et al., 2020), and extreme weather (e.g., flooding, typhoon) (Chen et al., 2021; Treilles et al., 2022) are directly or indirectly introduced into the aquatic environment. For example, typhoons significantly increased MPs abundance in surface water (Chen et al., 2021). Atmospheric deposition is reported as the dominant pathway for MPs transport to remote mountainous lake basins (Dong et al., 2021) and in snow samples of Europe and the Arctic (Bergmann et al., 2019).

## 2.2 Distribution and sources of PFASs

PFOS and perfluorooctanoic acid (PFOA) are the dominant PFASs in aquatic environments. In addition, emerging PFASs (e.g., chlorinated polyfluoroalkyl ether sulfonates (F-53B)) were frequently detected in global surface waters and sediments (Pan et al., 2018; Lee et al., 2020a; Li et al., 2020a; Marchiandi et al., 2021). The concentrations of PFASs in freshwater were extremely high, up to 5500 ng/L in water bodies (Melbourne, Australia) (Marchiandi et al., 2021) and 345.7 µg/kg in sediment (Las Vegas Wash, USA) (Bai and Son, 2021), which is much higher than that found in the water bodies (maximum at 684 ng/L) and sediment (maximum at 25 ng/g) of marine environments (Liu et al., 2019). Freshwater samples were collected near fire sites storing PFAS waste products, fluorochemical production plants, and downstream of airports and WWTPs, which should be the reason for the high concentrations of PFASs in freshwater environments.

The point sources of PFASs were generally from landfills, manufacturing plants, the application of PFAS-containing products (e.g., AFFFs) and the transformation of PFAS precursors (Ahrens and Bundschuh, 2014). Industrial complexes and wastewater treatment plants (WWTPs) are considered as the major emission sources (Yao et al., 2016; Seo et al., 2019; Tenorio et al., 2020; Nickerson et al., 2021). Another notable source of PFASs is the plastics/rubber and consumer products (e.g.,

textiles, carpets) (Herzke et al., 2012; Gremmel et al., 2016; Schellenberger et al., 2019; Chen et al., 2020a; Hu et al., 2021), from which PFASs and MPs-PFASs complexes could be released into the environment during the production, usage and washing processes. Hu et al. (2021) found that the sources of plastic/rubber and textile products caused the highest PFAS contribution in New Hampshire (USA) private wells, as predicted by machine learning. Among nonpoint sources, atmospheric deposition and runoff are the main sources of PFASs to the aquatic environment. Twenty PFASs were detected in the Czech Republic air samples, of which the airborne concentrations were controlled by long-range atmospheric transport (Paragot et al., 2020). PFASs in snow can be recognized as an important atmospheric tracer (Wang et al., 2019b), indicating the atmospheric transport of PFASs. Additionally, Chen et al. (2017) demonstrated that riverine flow input was the primary source for PFASs discharged to the Bohai Sea (China).

## 2.3 Occurrence and distribution of MPs and PFASs

As displayed in Tables S3–S6, MPs and PFASs could coexist in the same water bodies and sediment. For example, both MPs and PFASs are widely detected in the Yellow River (Zhao et al., 2016; Duan et al., 2020), Yangtze River (Zhao et al., 2017; Hu et al., 2018), and Pearl River (Pan et al., 2014; Lin et al., 2018) of China; Mälaren Lake in Sweden (Pan et al., 2018; Rotander and Kärman, 2019); the Thames River in UK (Pan et al., 2018; Corcoran et al., 2020); the Rhine River across Germany and the Netherlands (Pan et al., 2018; Mani et al., 2019). The high abundance of MPs and PFASs in freshwater is closely related to the proximity of population density and anthropogenic activity. For example, samples from the Pearl River of China (e.g., MPs at 7924 items/m<sup>3</sup> in surface water and 9597 items/kg in sediment, PFOS at 290 ng/L) were collected in or near very large cities (e.g., Guangzhou), which could be considerably influenced by sewage discharge, WWTPs, roadways, and tributary inputs.

However, for marine environments, the greatest overlap of MPs and PFASs is found in the Atlantic (Zhang et al., 2019; Silvestrova and Stepanova, 2021), the eastern coast of China (e.g., Bohai Sea) (Zhang et al., 2017b; Liu et al., 2019), the Yellow Sea of China (Sun et al., 2018; Wang et al., 2019a; Feng et al., 2020; Zhong et al., 2021), the North Sea (Zhao et al., 2015; Mani and Burkhardt-Holm, 2020), and the Bering Strait (Kakhkashan et al., 2019; Mu et al., 2019). The highest concentration of PFASs in Bohai of China was collected near an oil production plant that had strict fire prevention measures and intensive oil industry activities (Liu et al., 2019), and the inputs of PFASs from the discharge of riverine flow, coastal wastewater and effluents to the Bohai Sea of China were estimated to be 87.3 tons per year (Chen et al., 2017). While the MPs in the Bohai Sea of China and North Sea were mainly from fishing lines and rope material, respectively, indicating the importance of commercial

fishing, marine farming and heavy ship traffic (Dubai and Liebezeit, 2013; Zhang et al., 2017b). This coexistence of MPs and PFASs in aquatic environments inevitably results in their direct interaction (e.g., adsorption), and combined toxicity to aquatic organisms.

### 3 Interactions between MPs and PFASs in aquatic environments

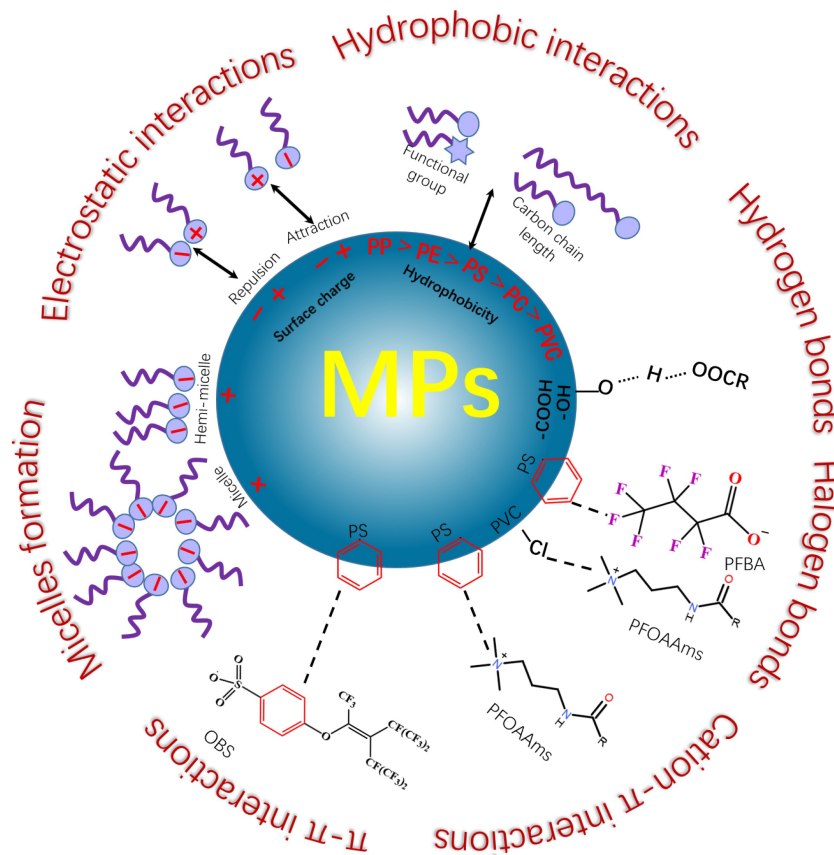
#### 3.1 Adsorption mechanisms of PFASs on MPs

Due to the coexistence of PFASs and MPs in aquatic environments, the adsorption of PFASs occurs on the surface of MPs. Actually, high concentrations of PFASs (up to  $9.07 \times 10^3$  ng/g) on MPs in the drain outlets were detected (Cheng et al., 2021). Adsorption is the most important environmental process between MPs and PFASs, and PFAS adsorption on PE, PS, and PVC MPs were previously studied (Wang et al., 2015; Llorca et al., 2018; Yang et al., 2020). For instance, Wang et al. (2015) reported that the lower adsorption capacity of PFOS on PE MPs mainly resulted from the electrostatic repulsion between anionic PFOS and negatively charged PE MPs in comparison with nonionic perfluorooctanesulfonamide

(FOSA). Llorca et al. (2018) reported that the adsorption of PFASs generally increased with increasing carbon-chain length (C11–C14) due to stronger hydrophobic interaction with MPs. In addition, the polymer type of MPs is important for PFAS adsorption, as the partition/distribution coefficient ( $K_d$ ) values of FOSA were PE > PVC > PS (Wang et al., 2015). Therefore, the adsorption capacity depends on the properties of both MPs and PFASs. Adsorption mechanisms have been reported in previous studies (Wang et al., 2015; Llorca et al., 2018), and electrostatic and hydrophobic interactions are considered as the dominant interactions, which is similar to the interaction of MPs with other organic contaminants (e.g., antibiotics) (Wang et al., 2021b) and PFASs with other particles (e.g., granular activated carbon) (Du et al., 2014). Other possible interactions, such as hydrogen bonding, halogen bonding,  $\pi$ - $\pi$  interaction, and cation- $\pi$  interaction are also proposed in this review (Fig. 1).

##### 3.1.1 Confirmed adsorption mechanisms

Electrostatic interactions would occur between anionic PFASs and the positively charged MPs, or between cationic PFASs and negatively charged MPs. Additionally, the charge of PFASs can be generated from both the



**Fig. 1** Adsorption mechanisms of PFASs on MPs. Electrostatic and hydrophobic interactions are confirmed mechanisms in the literature, while micelles formation,  $\pi$ - $\pi$  interaction, cation- $\pi$  interaction, halogen bonding and hydrogen bonding are proposed mechanisms which need to be verified. "R" group represents C-F chain.



functional groups and their unique molecular structures (Xiao et al., 2011). For example, in PFOS, the positively charged carbon framework was surrounded by negatively charged fluorine, oxygen and sulfur atoms (Xiao et al., 2011). The positively charged functional groups of adsorbents electrostatically adsorb the anionic functional groups of PFASs, while negatively charged adsorbents repel them. Also, with the decrease of pH, the MPs surface is more positively charged or less negatively charged by protonation, therefore altering the electrostatic interactions. Since the  $pK_a$  value of PFOS is lower than 1.0 (Table S1), they exist in the form of anions when  $pH > 1.0$ . Therefore, the adsorption of anionic PFOS on the surface of protonated PE MPs increased with the decrease of pH (3.0–7.0) due to the reduced electrostatic interaction (Wang et al., 2015). In addition,  $Na^+$  and  $Ca^{2+}$  are the common ions in natural waters. The increase of NaCl and  $CaCl_2$  concentrations increased the adsorption amount of PFOS on PE MPs, but did not increase the amount of PFOS on PVC MPs, indicating that electrostatic interaction was not the only force responsible for PFOS adsorption on MPs (Wang et al., 2015).

Previous studies confirmed that hydrophobic interactions played an important role in the adsorption of PFASs on MPs (Wang et al., 2015; Llorca et al., 2018). Both functional groups and carbon chain length can affect the hydrophobicity of PFASs. Generally, PFASs with longer carbon chain lengths are more hydrophobic, and possess stronger adsorption on MPs (Llorca et al., 2018). In addition to the length of the perfluoro-carbon tail, the functionality of the head group also plays important roles. For instance, carboxylic acids make PFASs difficult to be adsorbed onto MPs, compared with sulfonic and sulphonamide groups (Llorca et al., 2018). The sulfonate moiety is slightly larger than the carboxylate moiety, and contributed to higher hydrophobicity (Higgins and Luthy, 2006). In addition, the increase of hydrophobicity in carbon-based materials could improve the adsorption (Deng et al., 2012), as influenced by the molecular composition and structure of polymers (Llorca et al., 2018). The hydrophobicity of the common MPs follows the order: PP > PE > PS > PC > PVC, which has a high correlation with their functionalities of MPs (Min et al., 2020). This could be the reason for the higher adsorption amount of FOSA on PE MPs compared with PVC and PS MPs (Wang et al., 2015). In natural environments, MPs can undergo aging and increase the contents of oxygen-containing groups (e.g., -COOH) on their surface (Wang et al., 2020c). These polar groups could decrease the hydrophobicity of MPs. For example, the adsorption of PFASs on PS-COOH MPs was lower than that on pristine PS MPs (Llorca et al., 2018). However, oleophobicity is another property of C-F chains. Accordingly, the extracellular polymeric substances (EPS) (containing proteins, lipids, and polysaccharides) adsorbed on the MPs surface could improve the hydrophobicity of MPs

(Fadare et al., 2020), which would possibly increase the adsorption between PFASs and MPs. Unfortunately, there is scarce information to address the role of EPS on PFAS adsorption, which needs further investigation.

### 3.1.2 Unconfirmed adsorption mechanisms

Although electrostatic and hydrophobic interactions were found to be the dominant adsorption mechanisms, other possible interactions could also significantly affect PFAS adsorption on MPs, such as hydrogen bonding, halogen bonding,  $\pi$ - $\pi$  interaction, cation- $\pi$  interactions, and micelle formation. Due to the hydrophobicity of PFASs, it is hard for C-F chains of PFASs to form hydrogen bonds with MPs, while oxygen atoms in the functional groups of PFASs make it feasible. It is showed that hydrogen bonds were able to form between the carboxyl group in the solute and the carboxylate groups on the adsorbent surface (Li et al., 2013). PFCAs (e.g., PFOA) contain carboxylate groups, and aged MPs are enriched in oxygen-containing groups (Wang et al., 2020c). Therefore, in contrast to pristine MPs, the aged MPs possibly improve the adsorption of PFCAs by forming hydrogen bonds. In addition, Bhagwat et al. (2021) reported that N-H bonds on nylon (PA) MPs enhanced PFOS partitioning in comparison with other MPs (e.g., PP, PE), and speculated that hydrogen bonding played an important role in PFOS adsorption although direct evidence was not provided. As for  $\pi$ - $\pi$  interactions, it mainly exists in aromatic compounds. Benzene ring-containing MPs (e.g., PS MPs) and PFASs (e.g., sodium *p*-perfluorononenoxybenzene sulfonate, OBS) may form  $\pi$ - $\pi$  bonding interaction through  $\pi$ - $\pi$  electron coupling (Fig. 1), which deserves in-depth investigations. In the case of cation- $\pi$  interactions, they mainly exist between cationic and aromatic compounds. Benzene ring-containing MPs (e.g., PS MPs) and cationic/zwitterionic PFASs (e.g., perfluorooctaneamido ammonium salt (PFOAAmS)) may form cation- $\pi$  bonds (Fig. 1), which merits further investigations. Furthermore, Wu et al. (2019b) reported that halogen bonds formed between chlorides on PVC MPs and electron donors (e.g., hydroxyl groups, benzene rings) on bisphenol A, thus promoting its adsorption. For PVC MPs, it is speculated that halogen bonds could also be formed during the adsorption of amidogen-containing PFASs (e.g., perfluorooctaneamido ammonium salt (PFOAAmS)). In addition, fluorine atoms (electron acceptors) on PFASs may form halogen bonds with hydroxyl groups and benzene rings on MPs (e.g., weathered PS), which deserves further investigation.

Interestingly, PFASs with long chains (e.g., perfluorooctadecanoic (PFODA)) can form hemi-micelles or micelles by the hydrophobic aggregation of C-F chains (Chen et al., 2011; Llorca et al., 2018). It has been reported that PFASs can form hemi-micelles and then increase adsorption onto carbon-based adsorbents (e.g., maize

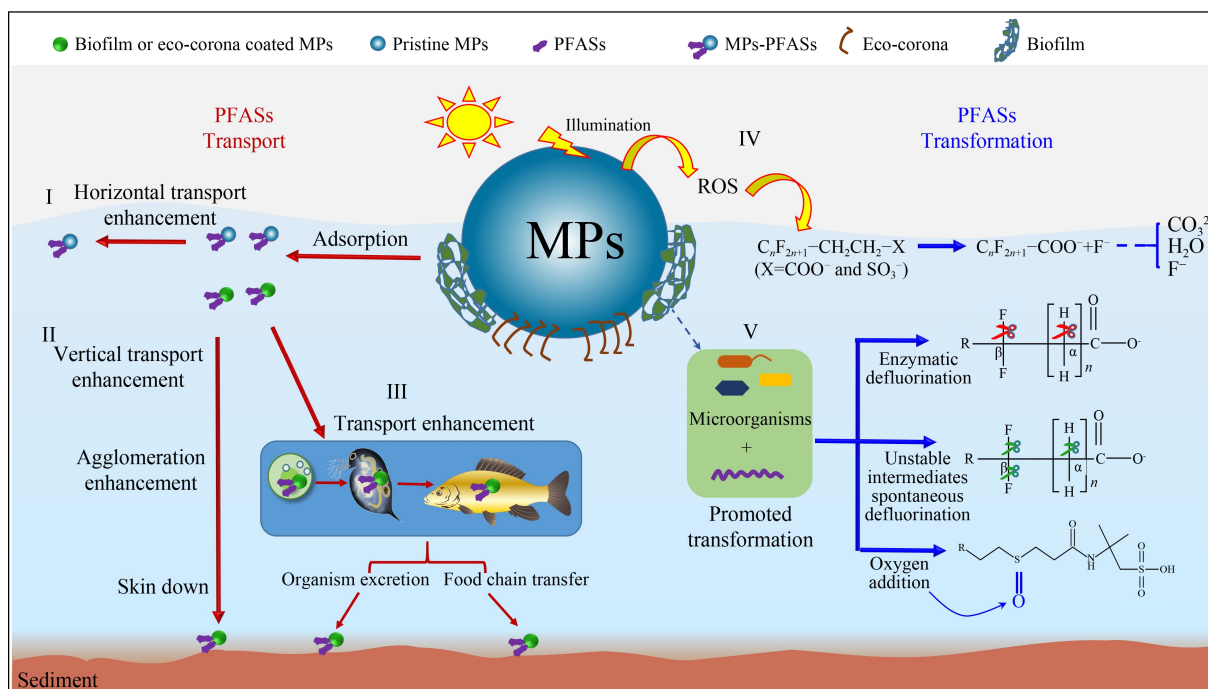
straw-origin ash) (Chen et al., 2011). Also, PFASs in the form of micelles were supposed to be adsorbed onto MPs (e.g., protonated MPs), but solid evidence is still lacking. If PFASs could be adsorbed onto MPs in the form of micelles, the processes are explained as follows: First, PFASs gather around the surface of positively charged adsorbents (e.g., protonated MPs), and the hydrophobic tails tend to aggregate together to form hemi-micelles. Second, the PFASs adsorbed onto adsorbent surface are covered by hemi-micelles. Finally, the whole adsorbent surface can be coated with micelles of PFASs (Fig. 1), thus enhancing the adsorption capacity toward PFASs when their solubilities are higher than the critical micelle concentrations (e.g., perfluorodecanoic (PFDA)) (Llorca et al., 2018). Although adsorption mechanism has not been reported during PFASs-MPs interactions, it can be speculated that the hemi-micelles/micelles of PFASs on the surface of protonated MPs or MPs with positively charged functional groups such as  $-NH_2$  could promote this adsorption process.

### 3.2 Effects of MPs on the transport and transformation of PFASs

Upon adsorption, MPs may serve as a vector for PFASs, thus altering the transport of PFASs (Fig. 2). On the one hand, pristine MPs, as carriers (e.g., over 16000 ng/mg FOSA adsorbed onto the PS surface (Llorca et al., 2018)), could increase the horizontal transport of PFASs (Process

I, Fig. 2). On the other hand, the aggregation and biofouling process induced the sedimentation of MPs (Process II, Fig. 2), which are related to the particle physiochemical properties (e.g., shape) (Van Melkebeke et al., 2020). In natural aquatic environments, MPs could form “eco-corona” and/or biofilm on the surface during aging, and then be attached with natural minerals, thus leading to a decrease in particle buoyancy. Meanwhile, EPS in the “eco-corona” or biofilm makes MPs much stickier and promotes heteroaggregation between MPs and organisms (e.g., algae) (Long et al., 2015; Rummel et al., 2017). This process could increase the sinking and vertical transport of MPs and thus the MPs-PFASs (Process II, Fig. 2). Furthermore, PFASs could be adsorbed on MPs and increase their uptake along with MPs ingestion by aquatic organisms, which further changes the transport of PFASs in aquatic environments (Process III, Fig. 2).

It was reported that PFASs could undergo transformation in aquatic environments (Zhang et al., 2021). The transformation mechanisms are related to the cleavage of C-F bonds, and the conversion of functional groups by oxidation, dealkylation, and defluorination (Zhang et al., 2021). For example,  $\cdot OH$  is able to trigger defluorination process and thus shorten the carbon chain length of PFASs. Liu et al. (2021) found that the  $\cdot OH$  oxidation cleaved 35%–95% of C-F bonds, converting fluorotelomer carboxylates (FTCAs,  $C_nF_{2n+1}-CH_2CH_2-COO^-$ ) and sulfonates (FTSAs,  $C_nF_{2n+1}-CH_2CH_2-SO_3^-$ ) to PFCAs



**Fig. 2** Effects of MPs on the transport and transformation of PFASs. Process I and II: MPs increase the horizontal and vertical transport of PFASs in water, respectively. Process III: MPs enhance the PFAS dispersion by organism excretion and food chain transfer. Process IV and V: MPs promote the transformation of PFASs through photodegradation and microbial decomposition, respectively.

( $C_nF_{2n+1}COO^-$ ). Importantly, Zhu et al. (2020) demonstrated that the photo-transformation of PS MPs could stimulate the generation of reactive oxygen species (ROS), including  $O_2^{\cdot-}$ ,  $^1O_2$ ,  $H_2O_2$  and  $\cdot OH$ . Therefore, PFASs may be transformed/degraded during the interaction with photo-transformed MPs under light irradiation (Process IV, Fig. 2); however, to the best of our knowledge, there is no report to support this hypothesis. In addition, it has been reported that PFASs could be transformed/degraded by aerobic microbes (Harding-Marjanovic et al., 2015; Che et al., 2021). Activated sludge communities contributed to the defluorination of short-chain fluorinated carboxylic acids (FCAs) (Che et al., 2021). The cleavage of C-F or C-C in the biotransformation process is possibly due to: 1) the enzymatic defluorination of functional groups such as C-F bonds at the  $\beta$ -position, 2) the spontaneous defluorination of the unstable intermediates, and 3) the cleavage of C-C bonds by oxygen addition (Harding-Marjanovic et al., 2015; Che et al., 2021). MPs surfaces are usually colonized by microorganisms and form biofilms in freshwater, estuarine and marine environments (Rummel et al., 2017; Johansen et al., 2019). Therefore, although there is currently no report, we speculate that the enriched microorganisms on the MPs surface could contribute to PFAS transformation and increase the transformation rate (Process V, Fig. 2).

#### 4 Accumulation and individual toxicity of MPs or PFASs to aquatic organisms

Previous studies have reported the toxicity of MPs or PFASs to aquatic organisms at the organismal and/or molecular levels. In the present review, we summarized the responses of the four most studied organisms, including shellfish, *Daphnia*, algae, and fish, to MPs or PFASs. For shellfish, the studies of “distribution and accumulation” accounted for the greatest number, 55% and 57% for MPs and PFASs studies, respectively. “Growth and inhibition” have been widely addressed for both *Daphnia* and algae in response to MPs or PFASs. For fish, “growth and inhibition” (57%) and “molecular level” (47%) were more prevalent in MPs and PFAS toxicity studies, respectively (Fig. 3).

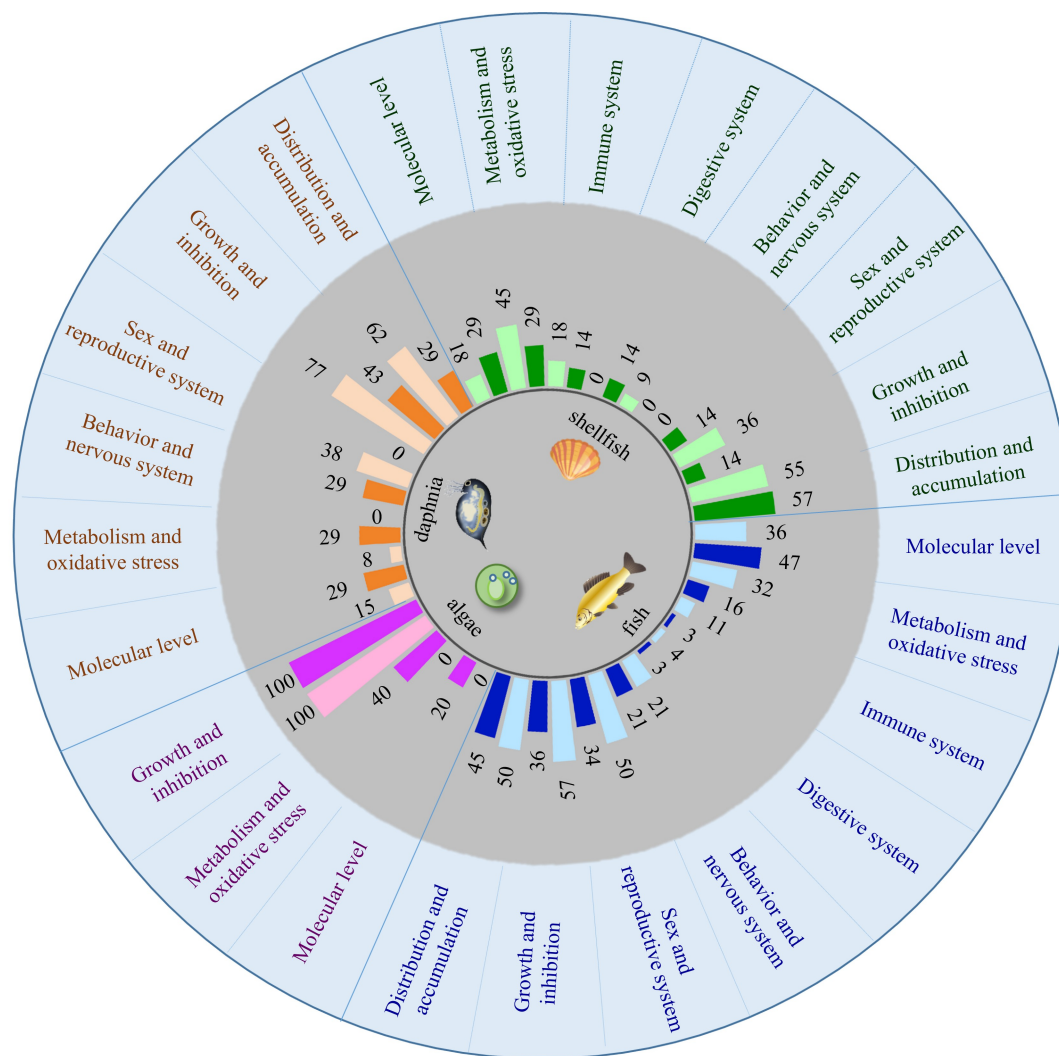
##### 4.1 Accumulation and toxicity of MPs to aquatic organisms

It was reported that both primary and secondary MPs posed a threat to aquatic organisms, including algae, shellfish, *Daphnia*, and fish (Table S7). Based on laboratory results, the most studied polymer type of MPs is PS, followed by PE and PP. In the case of algae, previous studies have mainly focused on growth inhibition and mortality (Fig. 3). In contrast to PFASs, the

size of MPs is an important factor for MPs toxicity. Generally, no toxic effect was observed when the cell size/MPs size ratio ranged from  $-1.85$  to  $1.33$  (log scale) (Chae et al., 2019). In contrast to large-sized MPs, small-sized MPs were mainly adsorbed on the cell surface, damaged cell walls (Liu et al., 2020a), increased membrane permeability (Wang et al., 2020a) and reduced photosynthetic efficiency (e.g., microalgae) (Zhang et al., 2017a). In the case of filter feeders (e.g., *Daphnia* and shellfish), filter feeding is the primary way for MPs uptake and accumulation in the digestive system, such as PE MPs in the *Daphnia* gut (Kokalj et al., 2018) and PS MPs in the mantle, visceral masses and gills of *C. fluminea* (Li et al., 2020b). Additionally, small particles ( $\leq 87.6$  nm) could accumulate in the liver through endocytosis (Li et al., 2020b). Polymer type is also an influence factor of toxicity. For example, the  $EC_{50}$  values of PVC, polylactic acid (PLA) and polyurethane (PUR) to *Daphnia magna* are 45.5, 122, and 236 mg/L, respectively (Zimmermann et al., 2020). MPs exhibited toxicity to filter feeder by 1) impairing the filtration capacity and digestive system (Colomer et al., 2019; Pedersen et al., 2020; Wang et al., 2020b) and 2) causing oxidative stress by inhibiting the activity of antioxidant enzymes (e.g., catalase) (Paul-Pont et al., 2016) and triggering related genes (e.g., *CYP4C34*) (Wu et al., 2019a) (Fig. 3).

For fishes, they could ingest MPs that have sizes similar to those of plankton and other food particles. MPs initially accumulated in the digestive tract, and gill and digestive organs such as the intestine are the main accumulation sites for MPs (Fig. 4A). Importantly, the accumulation showed size preference (Abarghouei et al., 2021). For instance, prey-size plastics possibly increased the encounter and ingestion rate by larval fish (Gove et al., 2019). MPs of smaller size ( $5\ \mu m$ ) tended to accumulate in fish gill, liver, and gut, while those of larger size ( $20\ \mu m$ ) were mainly in the gill and gut (Lu et al., 2016). Besides, it has been reported that nanoscale plastics ( $39.4$  nm) were able to penetrate the blood-brain barrier and eventually reach the brain (e.g., medaka) (Kashiwada, 2006). In addition to size, other properties of MPs (e.g., shape and color) could influence ingestion, and fibers were the more common shape of MPs that were found in the fish gut by field investigation (Su et al., 2019). In addition to classical endpoints, such as growth inhibition and mortality, current investigations on MPs toxicity also involve behavioral and physiological changes in aquatic organisms (Fig. 4B). After MPs ingestion by fish (e.g., yellow croaker, zebrafish), the consequent gut blockage and the inhibited activity of digestive enzymes may reduce energy availability (Gu et al., 2020). The change in enzymes mediated by the neuroendocrine system further explained the abnormal behavior (Umamaheswari et al., 2021). Additionally, MPs enhanced the intracellular ROS levels, decreased the activity of antioxidant enzymes (e.g., CAT), and





**Fig. 3** Ecotoxicological effects of MPs and PFASs on the four selected aquatic organisms. The bars with dark and light colors represent the toxicity of PFASs and MPs, respectively. Numbers indicate the frequency percentage (%) of this subject in the total selected papers. The total papers of MPs in shellfish, Daphnia, algae and fish are 11, 13, 5, and 28, respectively, while those of PFASs are 7, 7, 5 and 58, respectively.

downregulated the expression of oxidative stress and antioxidant defense related genes such as *cat*, *sod1*, and *gpx1a* (Umamaheswari et al., 2021).

More importantly, secondary MPs (e.g., photodegraded MPs) were reported to show higher toxicity due to the formation of functional groups (e.g., -COOH) on the surface and smaller size (Wang et al., 2020c). Similarly, the leachates (e.g., plasticizer, Zn, and polycyclic aromatic hydrocarbons (PAHs)) released from MPs were also toxic to aquatic organisms, which can not be ignored (de Silva et al., 2016; Schrank et al., 2019; Kolomijec et al., 2020). Although the leachate types were not identified, leachates from beached pellets did result in higher toxicity than virgin pellets (de Silva et al., 2016). PVC MPs without plasticizer diisononylphthalate (DiNP) did not result in obvious toxicity, while those with DiNP

significantly increased the mortality of *Daphnia magna* (Schrank et al., 2019). Further research is necessary to thoroughly reveal the toxicity mechanisms induced by secondary MPs (or aged MPs).

#### 4.2 Accumulation and toxicity of PFASs to aquatic organisms

The accumulation and toxicity of PFASs were mainly associated with their carbon-chain length and functional groups. It was reported that the long-chain PFASs showed higher accumulation in organisms (e.g., carp), because of stronger partitioning to phospholipids and higher binding energy with specific protein or fatty acid (e.g., L-FABPs) (Shi et al., 2018b; Shi et al., 2020). Moreover, the functionality of PFASs is a better predictor for distribution than the carbon chain length (Shi et al.,



2018b). In the case of PFCAs, bioconcentration factors (BCFs) of perfluoropentanoic acid (PFPeA), PFHxA, perfluorohexanoic acid (PFHpA), PFOA, perfluorononanoic acid (PFNA) in zebrafish were 0.9, 3.0, 18, 100 and 610, respectively (Menger et al., 2020). While for PFASs, the BCFs for perfluorobutanesulfonate (PFBS), perfluorohexanesulfonate (PFHxS), and PFOS were 18, 100, and 2700, respectively (Menger et al., 2020). In addition, in the presence of long-chain PFASs, the uptake, elimination rates in tissues and BCFs of short-chain PFASs in zebrafish were decreased, mainly due to the competition of transporters and binding sites of proteins between these two types of PFASs (Wen et al., 2017). PFAS accumulation was tissue-specific and mainly accumulated in protein-rich tissues, such as blood, liver, muscle and gonads (Fig. 4(A)). Sex-dependence was also observed. For example, F-53B in male zebrafish blood was much higher than that in female blood (Shi et al., 2018a).

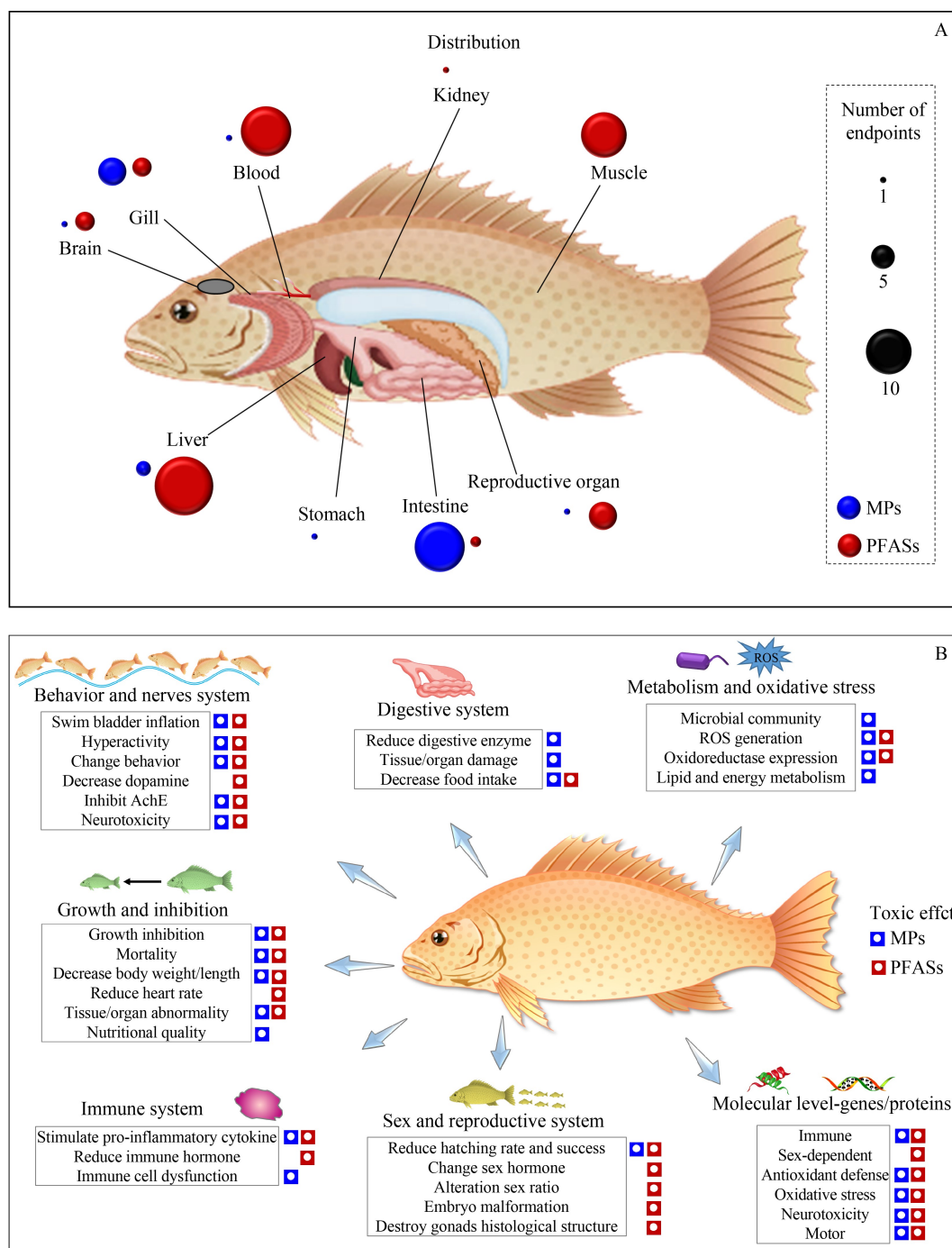
Both legacy and emerging PFASs could cause adverse effects on organisms (Fig. 4(B), Table S8). The  $EC_{50}$  values of PFOA were 0.594, 78.2, 207.46, and 350 mg/L for green mussel (*Perna viridis*) (Liu et al., 2014), *Daphnia carinata* (Logeshwaran et al., 2021), green algae (*Chlorella pyrenoidosa*) (Xu et al., 2013), and zebrafish (Ulhaq et al., 2013), respectively. Obviously, green mussels were much more sensitive to PFOA exposure, possibly due to the strong genotoxicity of PFOA to mussels. PFASs with longer carbon chain lengths as well as with an active group (e.g., sulfonate) displayed higher toxicity to aquatic organisms (e.g., zebrafish) (Hagenaars et al., 2011; Ding et al., 2012; Menger et al., 2020). In addition to growth inhibition, PFASs altered the sex and reproductive systems of aquatic organisms, induced the atrophy of oocytes in gonads (Liu et al., 2020b), reduced the yolk sac area (Tu et al., 2019), delayed the hatching rate (Shi et al., 2017b), altered the sex ratio of F1 (Lee et al., 2017), destroyed erythroid differentiation during embryonic development (Shi et al., 2017a), and even decreased the first brood quantity (Liang et al., 2017). Differentially expressed genes of vitellogenin (*vitel*), and choriogenin (*chgh*, *chghm*, *chgl*) indicated the damage of PFASs to the reproductive system at the molecular level (Kang et al., 2019).

In addition, PFASs could change the behaviors of fishes, including failed swim bladder, abnormal ventroflexion of the tails, hyperactivity (Gaballah et al., 2020), reduced total traveled distance and time of immobility, and increased thigmotaxis behavior (Jantzen et al., 2016). These abnormal behaviors induced by PFASs were attributed to the disruption of the nervous system. Neuroactive compounds such as neurotransmitters (e.g., acetylcholine (ACh)) and dopamine interacting with the host immune system are important for the regulation of the nervous system (Morais et al., 2021). After exposure of zebrafish embryos to PFASs, the ACh content was reduced, and dopamine was upregulated

(Guo et al., 2018). These results are consistent with the genes related to hormones that are important for neurodevelopment, such as the upregulation of *tgfa* (locomotive effects), *bdnf* (brain derived neurotrophic factor) (Jantzen et al., 2016), and *manf* (mesencephalic astrocyte-derived neurotrophic factor) (Guo et al., 2018) genes, and the downregulation of genes involved in hypothalamus-pituitary-somatotropic (HPS) such as *gh* and *igf2* (Tu et al., 2019). Oxidative damage is another mechanism of PFAS toxicity. PFASs induced excessive generation of ROS and changed the antioxidant related indicator contents such as malondialdehyde (MDA), CAT and glutathione (GSH) in algal cells (Xu et al., 2013), clam (*Corbicula fluminea*) (Liu et al., 2020b), *Daphnia carinata* (Liang et al., 2017) and zebrafish (Wu et al., 2019c). The changes in the expression of specific genes (*sod*, *se-gpx*, *gr*, *hsp22*, *hsp40*, *hsp60*, *cyp30*) or proteins (Nrf2, Akt/p-Akt) related to oxidative stress also indicated oxidative damage (Wu et al., 2019c; Liu et al., 2020b).

## 5 Combined toxicity of MPs and PFASs to aquatic organisms

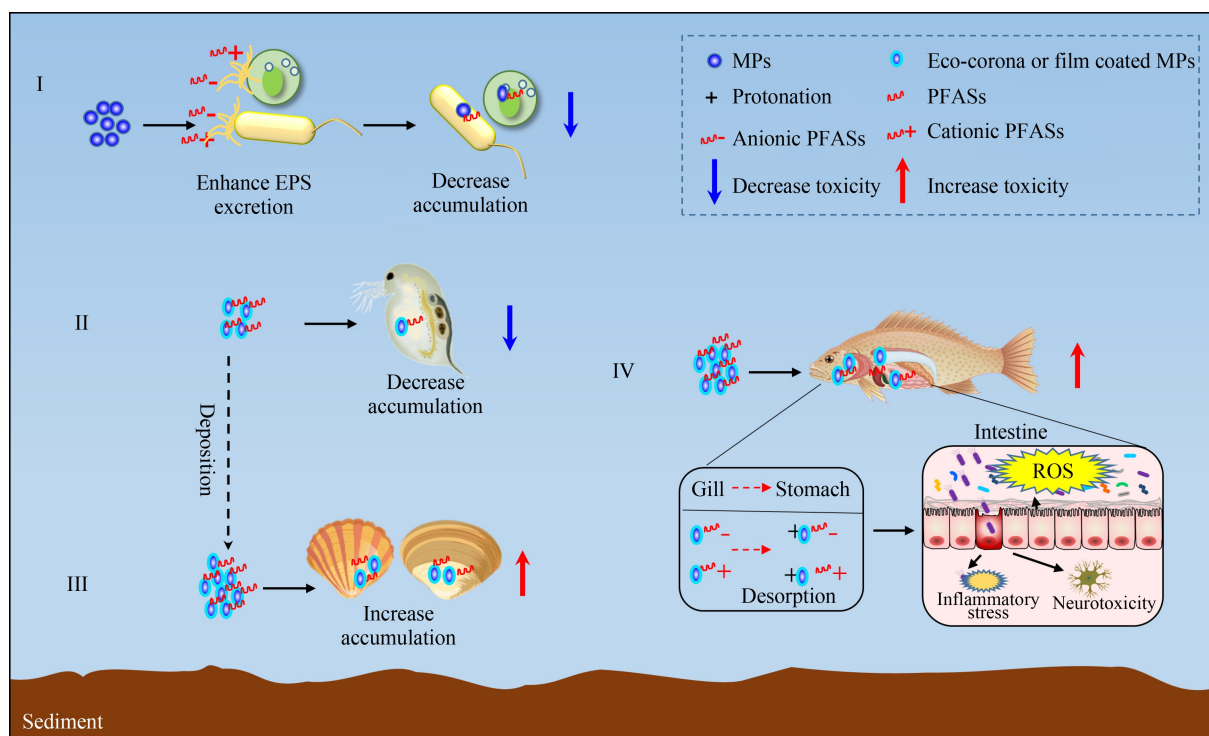
As mentioned above, owing to their small size and large surface area, MPs act as carriers of coexisting PFASs and thus alter their bioavailability. For example, it is clear that the distributions and locations of MPs or PFASs in fishes are largely different (Fig. 4(A)). MPs mainly accumulated in the gill and intestine of fishes, while PFASs were dominantly distributed in the blood, muscle, liver, and reproductive organs of fishes. For their toxicity, both MPs and PFASs could result in adverse effects on “growth and inhibition”, “behavior and nervous system”, and “molecular toxicity”. MPs-induced “digestive system” and “intestinal microbial community” toxicities are remarkably different from PFASs, whereas toxicity to the “sex and reproductive system”, especially to the sex ratio, is mainly induced by PFAS exposure (Fig. 4(B)). Therefore, it is speculated that the co-exposure of MPs and PFASs may lead to different toxic effects, in contrast to PFASs alone. Figure 5 shows the potential effects of MPs on PFAS toxicity to aquatic organisms. To the best of our knowledge, only four studies reported the combined toxicity of MPs and PFASs. With respect to microorganisms, Chen et al. (2020b) demonstrated that co-exposure of PS MPs and PFOS reduced the ROS production and cell permeability of bacteria in contrast to MPs exposure alone. This is because PFOS could promote EPS production, consequently reducing the entry of MPs into bacteria to cause toxicity (Chen et al., 2020b) (Process I, Fig. 5). However, the combined toxicity of PFASs and MPs to other microorganisms, especially algae, is currently unknown and needs further research. As discussed above, aged MPs could adsorb PFASs and deposit into sediment,



**Fig. 4** Distribution (A) and toxicity (B) of MPs and PFASs to fish. The endpoint number indicates the frequency in the papers of fish. The total papers of MPs and PFASs in fish are 28 and 58, respectively.

which will decrease the concentration of PFASs in the upper layer and increase PFAS concentration in the sediments. Therefore, this process may decrease the accumulation of PFASs in organisms in the upper layer (e.g., *Daphnia*) (Process II, Fig. 5). In turn, the accumulation of PFASs could be increased in benthic organisms such as clams (Process III, Fig. 5), in which solid evidence is still needed in future studies.

For aquatic organisms at high trophic levels such as fish, MPs coated with pollutants would mainly remain in the digestive system (e.g., gut). Yang et al. (2020) reported that the bioaccumulation of F-53B in zebrafish larvae was reduced due to the enhanced adsorption of F-53B on PS MPs. PFASs may penetrate into phospholipid bilayer membrane and enhance the permeability (Fitzgerald et al., 2018a; Fitzgerald et al., 2018b). This could facilitate MPs penetration into the



**Fig. 5** Effects of MPs on PFAS toxicity to aquatic organisms. Processes I: MPs and PFASs result in antagonistic effect on microorganisms. Process II: MPs decrease the accumulation and toxicity of PFASs to organisms in the upper water layer. Process III: MPs increase the accumulation and toxicity of PFASs to benthic organisms. Process IV: MPs increase the toxicity of PFASs to fish.

small intestine epithelium and entry into the circulatory system. Accordingly, the proportion of MPs in different organs/tissues (e.g., livers, brain) will be enhanced, which could correspondingly increase the hepatotoxicity and neurotoxicity (Process IV, Fig. 5). After adsorption, the MPs-PFASs complexes could be ingested by aquatic organisms such as fish, and enter the stomach with low pH values. MPs can be protonated at low pH values (Wang et al., 2015), and the positive surface could repel cationic molecules and attract anionic molecules. Hence, cationic PFASs might be released from MPs-PFASs complexes in the stomach. Given that only free PFASs are bioavailable, low pH in the stomach may lead to enhanced bioavailability of cationic PFASs and reduced bioavailability of anionic PFASs, but this hypothesis needs to be further verified.

## 6 Future perspectives and challenges

Currently, MPs and PFASs are widely detected in aquatic environments. This review discussed the interaction between MPs and PFASs and related mechanisms, which further govern their combined toxicity. Unlike other persistent organic compounds such as PAHs, PFASs are hydrophobic and oleophobic, leading to different adsorption behaviors and mechanisms on MPs. The adsorption behavior is controlled by the properties of both MPs and PFASs, and solution chemistry of aquatic

environments. Until now, there are still many gaps related to the adsorption and desorption of PFASs by MPs. It is noted that MPs could affect the transport of PFASs as a vector, and may enhance the transformation of PFASs through oxidation, dealkylation and defluorination during the photo-transformation of MPs. In addition, combined toxicity of MPs and PFASs has been summarized, which is regulated by the adsorption behavior and organism species. However, research on the MPs-PFASs interaction in actual natural environments is limited. This limits the understanding of toxicity, and greater emphasis should be placed on this topic. Further challenges and perspectives are put forward as follows:

1) MPs inevitably undergo photo-aging, thus decreasing their particle size and increasing the content of oxygen-containing functional groups. This will enhance the hydrophilicity of MPs. In addition, “eco-corona” or biofilm on the surface of MPs make them much more electronegative. All these changes in physiochemical properties of MPs could alter PFAS adsorption and interaction patterns, and deserve further efforts. Emerging PFASs (e.g., F-53B) are increasingly released owing to their usage for legacy PFAS substitutes. The carbon chain length and functional groups of these alternatives are different from those of legacy ones. Moreover, PFASs of zwitterions, cations and neutrals account for 54% of total number of emerging PFASs that detected between 2009



and 2017 (Xiao, 2017). Unfortunately, the current literature has focused on the adsorption of legacy and anionic PFAS (e.g., PFOS) on MPs. Adsorption of cationic PFASs in natural soils was reversible, while zwitterionic PFASs showed concentration-dependent hysteresis (Xiao et al., 2019). However, the reversibility of PFAS adsorption on MPs is unknown. Therefore, a thorough understanding of different types of PFASs is urgently needed.

2) Fibers and films are the most common shapes as detected in realistic environments, but spherical particles are currently the most studied shape. From a toxicity point of view, the size and shape are vital properties of MPs that control the interaction pattern of MPs with organisms, and the pathways of MPs passing through biological barriers (e.g., blood–brain barrier, placental barrier). Nanoplastics (< 1000 nm) may possess distinct properties compared with MPs due to their much smaller size and larger surface area, and studies related to their interaction with PFASs and combined toxicity are warranted. In addition, both MPs and PFASs at high doses were used during toxicological investigations, which is meaningful in some cases. However, environmentally relevant doses should be considered to better understand their adverse effects on aquatic organisms and ecosystems.

3) Finally, both MPs and PFASs were found to be transferred along the trophic food chains in aquatic environments. However, their co-transfer along food chains and biomagnification effect are largely unknown. In addition, MPs-PFASs complexes could be ingested by different aquatic organisms (e.g., zooplankton, bivalves, fish), but their bioavailability was unknown. Therefore, the fate (e.g., adsorption-desorption) of MPs-PFASs complexes under different physiological conditions (e.g., stomach, lysosome) should be better understood. Information on the combined effects of MPs-PFASs on the key enzymes and cells of aquatic organisms is also lacking.

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