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Review

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Microplastics as carriers of per- and polyfluoroalkyl substances (PFAS) in aquatic environment: interactions and ecotoxicological effects

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Abstract

MPs act as carriers of organic pollutants, including per- and polyfluoroalkyl substances (PFASs) in aquatic environments and exhibit harmful effects on organisms. Although there has been extensive research on the abundance and distribution MPs and PFASs individually, their co-occurrence and combined ecotoxicological effects have been poorly understood. In this context, the present review aims to present an up-to-date understanding of PFAS sorption onto MPs and their bioaccumulation in different organisms. The combined sources and prevalence of MPs and PFASs are summarized, with some daily-use products identified as important sources of both pollutants. MPs occurrence indicates considerable diversity in their levels, with concentrations ranging from 0-7 particles/m³ (air), 1-4712 particles/kg (soil), 1-26 particles/L (aquatic matrices), and 0-199 particles/organism (species) worldwide. Further, the literature survey provides evidence of PFAS concentrations ranging from < 0.0011 to 95.378 ng/m³ in the air, 6.90 to 294000 ng/kg in soil, 4 to 268 ng/L in aquatic environments, and 0.0014 to 796 ng/g in species. Additionally, the review investigates MPs-PFAS interaction in the aquatic system, thereby highlighting the factors that influence PFAS sorption onto MPs. Various mechanisms of PFAS sorption onto MPs have been examined and compared. Moreover, the review covers the ecological consequences of ingesting PFAScontaminated MPs and their synergistic effects on aquatic biota. Conclusively, more research is needed to evaluate the combined exposure to MPs and PFAS, particularly under environmentally relevant concentrations to understand the significance of MPs as a vector for emerging contaminants in the environment.



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Keywords: Microplastics (MPs), per- and polyfluoroalkyl substances (PFASs), co-occurrence, sources, exposure, sorption, ecotoxicological impacts, secondary pollution

INTRODUCTION

Microplastics (MPs) are plastic particles that measure less than 5 mm and are considered as significant pollutants that pose a threat to both human and ecological well-being^[1,2]. Boucher and Friot^[3] reported an annual discharge of approximately 1.5 million tons of MPs into waterways globally. MPs found in waterways have a range of harmful impacts on aquatic organisms, including obstructed digestion, reduced reproductive capacity, and instances of choking and starvation as a result of ingestion^[4]. Additionally, MPs have a large specific surface area and strong hydrophobicity, making them excellent carriers of toxic organic chemicals that gradually seep into waterways, leading to severe environmental and health problems^[5]. Several studies have investigated the adsorption and adherence of specific emerging contaminants to the surface of MPs in various aquatic environments, resulting in secondary pollution sources^[6-10]. For example, per- and polyfluoroalkyl Substances (PFAS) are among such emerging contaminants that can attach to the surface of MPs and cause toxicity in the environment^[7,9]. Similar to other chemicals that are known to adsorb onto MPs, PFAS possesses characteristics that can enhance the ability of MPs to act as carriers for these toxic chemicals^[2,3].

PFAS, are complex and ever-evolving group of synthetic chemicals comprised of alkyl-based materials that contain the C_nF_{2n+1} group, where some or entire hydrogen atoms are substituted by fluorine atoms^[11,12]. PFAS have been extensively utilized in industrial and commercial sectors, including as lubricants, surface coatings, building paints, cosmetics, insecticide formulations, wax, cookware, and aqueous film-forming foams^[9,13,14]. They have gathered considerable scientific attention due to their resistance to natural decomposition and ability to easily enter the aquatic environment, leading to both acute and chronic toxicity in aquatic organisms as well as humans^[15-17]. It has been investigated that MPs could adsorb and concentrate PFAS^[10,18,19] due to the hydrophobicity of plastic waste^[20]. Considering the widespread occurrence of MPs and PFAS in different aqueous systems and their prolonged persistence, these emerging pollutants may work in tandem within food chains to produce harmful impacts on fish, wildlife, and humans^[21].

PFAS possess certain characteristics that make them prone to being carried by MPs and researchers are currently investigating the interaction between the two by utilizing MPs for the adsorption of PFAS. For instance, Wang et al. investigated that polyethylene (PE) exhibited a stronger adsorption capacity for PFAS groups than polyvinyl chloride (PVC) and polystyrene (PS) MPs^[22]. Bakir et al. examined the desorption and adsorption of perfluorooctanoic acid (PFOA) on PE and PVC MPs^[18]. Similarly, Scott *et al.* conducted experiments to examine the interactions between PFAS and MPs in both real-world and controlled settings^[10]. They deployed various types of MPs in a lake for 1 month and 3 months, measuring the abundance of PFAS on these MPs. Comparing the results to laboratory conditions with only plastic and water, they found that PFAS adsorption was much higher in the field due to the presence of inorganic and/ or organic matter associated with the environment. The results suggest that the existence of organic or inorganic debris linked with MPs significantly boosts the sorption of PFAS. Cormier et al. examined the effects of MPs size on the sorption of PFOS for 6 months using PE MPs of four different sizes^[23]. As the particle size of the PE MPs decreased, a linear increase in the concentration of sorbed PFOS onto the PE was observed. Also, the sorption and desorption of such contaminants on MPs can be influenced by several parameters, such as the inherent characteristics of the MPs (e.g., surface charge, size, density, and aggregation), the physicochemical properties of the contaminants, and the environmental conditions (e.g., pH, salinity, weathering, and exposure to sunlight)^[24,25].

When PFAS substances are adsorbed or adhered to the surface of MPs, they can create secondary pollution in the environment^[6,18,22]. As MPs and PFAS have long persistence times in the environment, they can accumulate and persist for extended periods, leading to chronic exposure to these pollutants^[26]. Additionally, these PFAS-contaminated MPs can be ingested by aquatic organisms and transferred up the food chain, potentially causing harmful effects on the ecosystem and human health^[27,28]. The leaching of PFAS from contaminated MPs can also contribute to the spread of PFAS in the environment, therefore exacerbating the problem. Previous studies have shown the adsorption of PFAS by MPs that has been primarily focused on single-component analysis without considering additional factors or components that may influence the adsorption process. However, the nature of PFAS interactions with the surface of MPs and their potential effects on the surface chemistry remain largely unknown.

Given the widespread and enduring presence of MPs and PFAS in the environment, it is crucial to comprehend their interactions and potential impacts on both human health and the environment. In this context, this work presents a systematic review of the occurrence, distribution, and sources of PFAS and MPs in diverse aqueous matrices. The work discusses the role of MPs as a carrier of PFAS chemicals, which leads to secondary pollution through combined exposure. Further, the mechanisms of PFAS sorption on MPs and the features that influence their adsorption behavior in the aquatic environment have also been summarized. The synergistic ecotoxicological effects of MPs and PFAS in the aquatic environment have been explored. Additionally, this review also highlights existing research gaps and future research directions that need to be taken to predict the risks associated with the presence and adsorption of PFAS compounds onto the MPs in different environmental matrices.

REVIEW METHODOLOGY

A systematic literature search was performed using various search engines, including Google Scholar, Google, Scopus, ScienceDirect, Web of Science, and Connected Papers. In the first phase of the literature review, different keywords were utilized such as "MPs and PFAS co-occurrence", "MPs and PFAS in the environment", "MPs and PFAS distribution", "MPs and PFAS sources", "MPs and PFAS eco-toxicological effects", "PFAS sorption on MPs", and "MPs and PFAS combined toxicity" to retrieve relevant studies. Our focus was on selecting studies that extensively examined MPs and PFAS co-occurrence, distribution, sources, and combined toxicity while providing promising results. To ensure the reliability of the collected data and information, we applied filters to narrow down the search results to include only peer-reviewed research papers, scientific reports, data reports, and organization documents published until March 2023. This filtering process aimed to maintain the credibility of the sources included in the review. The initial search yielded around 163 research papers from databases such as Scopus, ScienceDirect, Web of Science, and Google Scholar. Furthermore, we retrieved approximately 3 organizational documents/reports and 2 scientific reports from Google. To ensure the dataset remained focused on the topic of the review article, a manual screening process was implemented. This process involved evaluating the summary for scientific and organizational reports, as well as the abstracts for journal papers. Articles that did not primarily address the relevant review topic were excluded from consideration. As a result of this screening process, a total of 130 journal papers were deemed relevant and included in the review.

MPS AND PFAS IN THE AQUATIC ENVIRONMENT: SOURCES, OCCURRENCE, AND

DISTRIBUTION

Potential sources of MPs and PFAS

There are several sources of MPs and PFAS co-occurrence in aquatic systems, which are closely linked to human activities. The sources of MPs and PFAS in the aqueous matrices are diverse and include both point and non-point sources. Mainly, MPs can come from the fragmentation of larger plastic items, plastic pellet

release during manufacturing and transportation, and disposal of plastic waste^[5]. Similarly, PFAS sources include industrial activities, non-stick coatings, firefighting foam, water repellents, and consumer products like food packaging and textiles. Both MPs and PFASs are commonly found in urban and industrial areas, particularly in areas with high population density and anthropogenic activity, such as coastal waters, estuaries, reservoirs, lakes, and rivers^[28]. The discharge of treated and untreated wastewater, as well as runoff from urban areas and agricultural land, can transport these pollutants into water bodies^[29,30]. In addition, the use of fertilizers and pesticides in agriculture can also contribute to the release of PFAS and MPs in aquatic systems^[31]. The main point sources of MPs and PFASs that may be released into the environment are illustrated in Figure 1, depicting various ways that could be responsible for their release.

Studies found that polyamide (PA) textiles treated with water-repellent which is commonly used in products like rainwear, carpets, and plastic fabrics, can emit both microfibers and residual PFAS^[7,32]. Certain consumer products (textiles, carpets, plastics, and rubber) are recognized sources of PFAS and MPs through the production, utilization, and cleaning processes of these products resulting in MPs-PFAS complexes in the environment^[32-34]. For example, machine learning predictions identified textile and plastic stuff as the primary sources of PFAS in private wells in New Hampshire, USA^[35]. Recent research highlighted that plastic food containers are often coated with PFAS to provide characteristic properties but potentially contaminate food^[7,36]. For example, Teflon-based cookware, which is composed of polymers and PFAS complexes while cooking^[37].

Among non-point sources, atmospheric deposition and runoff, unconventional waste disposal, and the discharge of wastewater treatment plants (WWTPs) are other major co-sources of PFAS and MPs to the aqueous matrices. Long-range atmospheric transport was found to control the airborne concentrations of around 20 PFAS substances identified in the Czech Republic which can also be recognized as important atmospheric tracers in the snow^[38]. Similarly, the prime source for PFAS released into the Bohai Sea in China was demonstrated to be riverine flow input^[39,40]. Recent studies have reported high levels of MPs in landfill soil and leachate, ranging from 570 to 14,200 items/kg, as well as high levels of PFAAs, including dominant compounds like PFOA and perfluorobutanesulfonic acid (PFBS), in landfill leachate in China^[41,42]. This showed that with time, the breakdown of products containing MPs and PFAS can result in their release into the environment. Simultaneously, Ziajahromi et al. reported that despite WWTP treatments, pollutants can still be discharged with effluent into the aquatic environment^[43]. Houtz et al. detected perfluorohexanoic acid (PFHxA), perfluorobutanoic acid (PFBA), and PFOA in effluent samples from 6 WWTPs at median concentrations of 24 ng/L, 19 ng/L, and 23 ng/L, respectively. Overall, MPs and PFAS have diverse sources in aquatic systems which are linked to human activities and pollution^[44]. Despite this, their coexistence and direct interaction are inevitable, thereby posing serious combined toxicity in the aquatic environment.

Co-occurrence and distribution of MPs and PFAS

Despite the increasing evidence indicating the existence of PFAS and MPs in various environmental compartments like the atmosphere, rivers, lakes, ocean, and soil/sediments [Table 1], there has been relatively little investigation into their co-occurrence and distribution in the ecosystem. Figure 2 depicts the concentration range of MPs (particles/L or particles/m³) and PFAS (pico/nano/micro gram/L) detected in various environmental compartments such as air, living species, and marine and freshwater systems around the world.

Sample studied	Study type	Contaminants detected	Source of contamination	References
Air				
Outdoor air	Field-scale	MPs (0-4.18 particles/m ³)	Textile	[45]
		PFAS (0.0013-0.923 ng/m ³)	Chemical industrial park	[46]
Indoor air	Lab-scale	MPs (1414 \pm 1022 particles/m ^{2/} day)	Fabrics	[47]
		PFAS (< 0.0011-3.100 ng/m ³)	NA	[48]
Outdoor and	Field and	MPs (0.45-6.64 particles/m ³)	Synthetic fibers, home furniture, and plastic fibers	[49]
indoor	lab-scale	PFAS (3.531-95.378 ng/m ³)	Industrial activities	[50]
Soil				
Shallow soils	Field-scale	MPs (78 \pm 13 particles/kg)	NA	[51]
		PFAS (640-294000 ng/kg)	Industrial areas, airports, landfills, fire stations, and agricultural areas	[52]
		MPs (1.3-4712 particles/kg)	Human activities on the coastal beaches	[53]
		PFAS (6.90 ng/kg)	Fluorochemical plants and large manufacturing industry	[19]
Aquatic matric	es			
Taihu lake	Field-scale	MPs (3-26 particles/L)	Fibers, WWTPs	[54]
		PFAS (5-78 ng/L)	NA	[55]
Pearl river		MPs (0.57 \pm 0.7 particles/L)	Fragmentation of discarded plastic wastes	[56]
		PFAS (6-14 ng/L)	Fluorochemical industry, WWTPs	[57]
Bohai sea		MPs (0.33 \pm 0.34 particles/10 ³ L)	NA	[58]
		PFAS (4-118 ng/L)	Fluorotelomer industry	[59]
Yangtze River		MPs (2 × 10^{5} -9 × 10^{5} items/km ²)	NA	[60]
		PFAS (39-220 ng/L)	Fluororesin coatings/metal plating, surface runoff/textile, effluent discharge/food packaging, and leather/fabrics	[61]
Atlantic Ocean		MPs $(2.46 \pm 2.43 \text{ particles}/10^3 \text{ L})$	NA	[62]
		PFAS (0.011-0.069 pg/L)	Atmospheric deposition and continental sources	[63]
Great lakes		MPs (1.26 × 10 particles/km ²)	NA	[64]
		PFAS (11-121 ng/L)	NA	[65]
Rhine River		MPs $(2.7 \pm 0.4 - 6.3 \pm 2 \text{ particles}/10^3 \text{ L})$	NA	[66]
		PFAS (119-268 ng/L)	WWTPs	[67]
Living organis	ns			
Zooplankton	Field-scale	MPs (2 ± 0.2 particles/organism)	NA	[68]
		MPs (0.07-1.2 particles/organism)	Chemical industries like papermaking, coatings, inks, textile printing	[69]
		MPs (0.058 \pm 0.01 particles/organism)	NA	[70]
		PFAS (0.1-12.98 ng/g)	WWTPs	[71]
		PFAS (4.18 ng/g)	Discharges from municipal wastewater	[72]
		PFAS ($17 \pm 13 \text{ ng/g}$)	NA	[73]
Shellfish	Field-scale	MPs (198.82 particles/organism)	NA	[74]
		MPs (3.35 ± 1.85 particles/g)	Storm drains	[75]
		PFAS (0.0156-64.0 ng/g)	NA	[76]
		PFAS (0.0054-7.543 ng/g)	NA	[77]
Fish	Field-scale	MPs (0.65-3.82 particles/g)	NA	[78]
		MPs (2.88 \pm 3.12 particles/organism)	NA	[79]
		MPs (2.15 \pm 1.55 particles/organism)	NA	[79]
		PFAS (180-145000 ng/L)	WWTPs	[71]
		PFAS (64200 ng/L)	WWTPs	[72]
		PFAS (0.0014-1.627 ng/g)	NA	[77]

Table 1. Studies reporting MPs and PFAS occurrence and distribution in the environment worldwide

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Seal	Field-scale	MPs (0-16 particles/10 organism)	NA	[80]
		MPs (0-43 particles/organism)	NA	[81]
		PFAS (12.6-796 ng/g)	Wastewater effluent and sediment	[82]
		PFAS (111 \pm 5 ng/g)	NA	[83]

MPs: Microplastics; NA: not available; PFAS: per- and polyfluoroalkyl substances; WWTPs: wastewater treatment plants.



Figure 1. Sources of MPs and PFAS co-occurrence in the environment. MPs: Microplastics; PFAS: per- and polyfluoroalkyl substances.

Understanding the interactions between the two types of pollution requires a focus on their coexistence in the same location^[28]. While variations in sampling sites across studies may limit direct comparison, available data suggests the presence of both pollutants in common locations. Previous studies have indicated that closed or inland waters have greater concentrations of PFAS and MPs compared to open ocean waters due to factors such as the restricted ability of self-cleaning, retention processes, and land-based sources that impact ocean current transmission. The co-occurrence of PFAS and MPs in the marine ecosystem is particularly prevalent in certain regions, including the Atlantic, eastern coast of China (e.g. the Bohai Sea), Yellow Sea of China, North Sea, and Bering Strait, as evidenced by several studies^[84-89]. For example, water samples collected from coastal areas of Japan contained PFOA levels that were 1000 times greater than those



Figure 2. Occurrence and distribution of MPs and PFAS in the environment worldwide. MPs: Microplastics; PFAS: per- and polyfluoroalkyl substances.

found in the central Pacific Ocean^[s9]. According to Dai *et al.*, the quantity of MPs in the Bohai Sea is as high as 7667 pieces/m³, which is more than what has been observed in the open ocean, such as the northeast Pacific Ocean where it was 2080 pieces/m^{3[90]}. The elevated concentrations of the two contaminants were typically detected in locations where the associated products were produced, utilized, and discarded, such as industrial wastewater discharge points and waste disposal sites. For example, the PFAS concentration in Bohai of China was highest in proximity to an oil plant with tight fire protection actions and severe activities of the oil industry, and the estimated inputs of PFAS to the Bohai Sea from coastal wastewater, riverine flow, and effluents were substantial^[39,40,91]. In contrast, fishing lines were identified as the main source of MPs in the Bohai Sea in China, while rope material was found to be the primary source in the North Sea. This emphasizes the significance of commercial fishing, marine farming, and the high volume of ship traffic in these areas^[92,58]. The presence of MPs and PFAS can be influenced by their own physical and chemical characteristics as well as the environmental conditions, which play a significant role in determining their distribution and presence in the ocean.

Likewise, the persistence of PFAS and MPs in freshwater is strongly linked to human activity and population. The concentration of MPs in Taihu Lake, China, was found to be significantly higher than in Qinghai Lake, in Northwest China, where levels were comparatively low. Likewise, PFOA and PFOS were detected at higher levels in Huangpu River as opposed to remote surface water areas in China. For instance, both PFAS and MPs have been extensively detected in several freshwater systems in China including the Yellow River^[93,94], Yangtze River^[58,95], and Pearl River^[59,60], as well as Mälaren Lake in Sweden^[61], the Thames River in the UK^[96,97], and the Rhine River in Germany and the Netherlands^[55,98]. The concentration of MPs in Taihu Lake, China, was found to be significantly higher (ranging from 3400 to 25800 p/m³) than in Qinghai Lake, in Northwest China, where levels were comparatively low^[64,99]. Also, the presence of PFOA and PFOS was detected at higher levels in Huangpu River (139.6 ng/L and 46.5 ng/L, respectively) as opposed to remote aquatic zones in China (0.4 ng/L and 2.4 ng/L, respectively)^[57,100]. In addition, samples collected from the Pearl River in China revealed high levels of MPs at 7924 items/m³ in surface water and 9597 items/kg in sediment, as well as PFOS at 290 ng/L. These samples were taken in or near densely populated urban areas,

such as Guangzhou^[55]. These sites are likely to be significantly impacted by sewage discharge, tributary inputs, WWTPs, and roadways. Overall, the co-existence and distribution of PFAS and MPs in various environmental compartments, particularly in areas of high population density and anthropogenic activity, highlights the importance of studying their interactions and potential impact on aquatic organisms and human health.

MPS AS CARRIERS OF PFAS IN THE AQUATIC ENVIRONMENT

Type, properties, and concentration of PFAS adsorbed on MPs

PFAS (logKow: 1.35-9.95, logKoc: 0.93-6.20 L/kg) are emerging contaminants that are persistent, ubiquitous, toxic and bioaccumulated in organisms in aquatic medium as shown in Figure 3. They share properties with other chemicals that adhere to MPs, potentially enhancing the ability of MPs to carry them^[101]. Studies have shown that MPs can act as vectors for PFAS, meaning that they can adsorb and transport these substances in the environment. The type of PFAS that adsorbs to the surface of MPs can vary depending on the properties of the PFAS, including their molecular weight and chemical structure^[31,85,97]. Long-chain PFAS, such as PFOA and PFOS, have been shown to adsorb to the surface of MPs more readily than short-chain PFAS. For example, Wang *et al.* found that perfluorooctanesulfonamide (PFOSA), despite having similar hydrophobic free energy to PFOS, is more easily adsorbed by PE MPs^[22]. This is because anionic PFAS molecules electrostatically repulse from the PE surface.

Notably, PFASs, including perfluoroalkyl carboxylic acids (PFCA), PFOA, and PFOS, persist in the environment because of their stable C-F backbone with a high bond energy of 485-585 kJ/mol due to Fatoms' electronegativity^[38,55,102]. They can cause harmful developmental effects, infertility, carcinogenicity, and cell cycle alteration, even at concentrations as low as 70 ng/L^[28,32]. In addition, the concentration of PFAS adsorbed on MPs can vary widely, depending on the concentration of PFAS in the surrounding environment and the amount of exposure time. Some studies have suggested that the concentration of PFAS adsorbed on MPs can be several orders of magnitude greater than the concentration of PFAS in the surrounding water^[55,101]. For instance, high concentrations of PFAS (up to 9.07 × 10³ ng/g) on MPs in the drain outlets were detected^[6]. The concentration of PFAS adsorbed on MPs can increase over time as the MPs are exposed to PFAS-contaminated environments. For example, a significant increase (24-259 times) in PFAS adsorption on MPs reported was compared to laboratory experiments, thereby highlighting the role of inorganic and/or organic matter associated with MPs in enhancing PFAS adsorption^[10].

Water chemistry affects PFAS adsorption through salting out, anions competing with ionized PFAS, and cation bridging^[70]. Short-chain PFAS have stronger electrostatic interactions, while longer-chain PFAS have more hydrophobic interactions per net charge^[103]. Likewise, Higgins and Luthy, suggest hydrophobic/ electrostatic interactions are the main sorption processes of PFASs onto organic material. However, the differences in their sorption efficiency on MPs may be due to functional group differences^[104]. For example, the hydrophobic chain of PFASs is surrounded by an electronegative C-F bond, which generates a negative coating around it and makes the interactions more complex^[105]. Additionally, PFOS has a sulfonate group that is more polar than its carboxylic group, and the smaller carboxylic head reportedly results in greater sorption on the MPs^[18,106]. Oliver *et al.* found that hydrophobic interactions were the main factor in the successful sorption of PFOS onto PE MPs^[106], therefore, resulting in substance-specific sorption behavior. Nevertheless, limited studies are available to date that investigate PFAS sorption behavior on MPs, and it can further vary based on MPs properties in the aquatic environment. Therefore, more thorough research is necessary to comprehend the mechanisms of PFAS adsorption on MPs and its potential environmental and human health impacts.



Figure 3. MPs as a carrier of pollutants in the environment: (A) sorption of PFAS on MPs; (B) desorption and release of PFAS from MPs. MPs: Microplastics; PFAS: per- and polyfluoroalkyl substances.

Type and properties of MPs affecting PFAS sorption

MPs can act as carriers of contaminants in water, as they accumulate these substances at higher concentrations than the surrounding water due to their anthropogenic surfaces. Therefore, understanding the sorption behaviors of microplastics and contaminants is crucial to reduce the environmental harm caused by the increased MPs pollution^[107,108]. The sorption behaviors of MPs for contaminants like PFAS^[109] are shown in Figure 4.

The shape of MPs can affect their ability to adsorb PFAS and release contaminants into the environment. The shape of MPs, such as those with rough or porous surfaces, can also increase their ability to sorb PFAS^[34,93,97,103]. MPs in the environment exist in various shapes, such as fibers, fragments, beads, films, and foam, which impact their surface area and subsequent adsorption behavior^[1,2]. The shape of MPs can influence the formation of pores and cracks on their surface due to weathering or biodegradation, which can increase the surface area and promote the accumulation of PFAS^[107,108]. Similarly, MPs' sorption behavior can also be affected by their size, which impacts their adsorption capacity, adsorption/desorption rate, and equilibrium rate^[110,111]. While smaller MPs tend to have higher sorption capacities due to increased surface area, particle sizes < 235 nm can experience decreased sorption rates due to self-aggregation^[5]. For example, Cormier *et al.* observed that the sorption of PFOS on different sizes of MPs was more efficient for smaller particles than larger particles in all tested concentrations^[23]. The chemical composition of MPs, such as the type of plastic and the presence of additives, have been reported to affect the adsorption and



Figure 4. Factors affecting the sorption of PFAS on MPs in the aquatic environment. MPs: Microplastics; PFAS: per- and polyfluoroalkyl substances.

degradation of PFAS. Some studies have shown that MPs made from certain types of plastic, such as PE, can adsorb more PFAS than MPs made from other types of plastic. According to Pascall *et al.*^[112] and Karapanagioti and Klontza^[113], PE particles exhibit significant segmental mobility and free volume, making them capable of adsorbing chemicals. Similarly, when studying MPs as a potential carrier of emerging contaminants in the aquatic system, it is crucial to evaluate the polarity of the polymer. Non-polar polymers are known for their chemical resistance and structural strength, which arise from the presence of C-C and C-H covalent bonds. However, the introduction of various functional groups to the C, H, or sidechains of a polymer can lead to polarity^[91].

In addition, the surfaces of MP particles can provide a substrate for the growth of biofilms, which can result in the sorption of other pollutants, including hydrophobic PFAS^[10,113]. Hydrophobic MPs can provide a favorable environment for the adsorption of PFAS due to their nonpolar surfaces. In contrast, hydrophilic MPs tend to repel PFAS and are less likely to sorb these compounds. The hydrophobicity of PFAS can also influence their interaction with MPs. PFAS with higher hydrophobicity tends to adsorb more strongly onto MPs due to stronger intermolecular interactions between the hydrophobic surfaces. Increasing the hydrophobicity of MPs can enhance their sorption capabilities. For instance, PE MPs were found to exhibit greater sorption of FOSA compared to PVC and PS MPs. It is noteworthy that the hydrophobicity of common types of MPs generally follows the sequence: PP > PE > PS > PC > PVC^[15]. Nevertheless, the research on the influence of MPs properties on PFAS sorption is an ongoing and evolving field. While some studies have explored the impact of specific physical and chemical properties of MPs on PFAS sorption as mentioned above, there is still much to learn about the extent to which these properties affect PFAS sorption in aquatic environments.

Environmental factors influencing PFAS and MPs interaction

Environmental factors such as temperature, pH, salinity, dissolved organic matter (DOM), temperature, and the composition of the water phase can also influence PFAS sorption onto MPs, and this aspect requires

more investigation^[28,31,90]. Changes in pH can affect the ionization state of both the MPs and the PFAS, altering their surface charge and, therefore, their affinity for each other^[102]. For instance, at low pH values, PFAS can become protonated and positively charged, while MPs can become deprotonated and negatively charged^[57,99,105]. This results in a strong electrostatic attraction between the two, leading to enhanced PFAS sorption onto the MP surface. At high pH values, the opposite effect can occur, with PFAS becoming negatively charged and MPs becoming positively charged, resulting in reduced sorption^[98,99,100,114]. Mejías *et al.* found that a decrease in pH from 7 to 4 increased the sorption of PFAS on PA MPs^[102]. At pH greater than the point of zero charges (pHPZC), repulsions between the negatively charged PA surface and PFAS occurred, resulting in lower adsorption^[103]. Other chemical species in the water, such as ions and organic matter, can also compete with PFAS for sorption sites on MPs and are affected by the pH of the aquatic matrices. Therefore, solution pH can have a significant impact on PFAS on MPs^[10,15,99].

The impact of salinity on PFAS adsorption on MPs is an important consideration in the assessment of the fate and transport of these contaminants in aquatic environments. Reportedly, high salinity levels can lead to decreased sorption of PFAS on MPs due to increased competition from other ions present in the water. Similarly, Mejías *et al.* found a significant decrease in the adsorption of PFOS onto PA in the presence of salts^[102], while no significant difference was observed for the other compounds by NaCl content. In addition, high dissolved organic matter content reduces PFAS adsorption onto MPs due to competition with pollutants for sorption sites and complexation with hydrophobic acids^[39,40,115]. The effect is more significant for long-chain PFAS (PFOS and PFOA) and less significant for short-chain PFAS such as perfluorobutanoic acid (PFBuA) and perfluoropentanoic acid (PFPeA). Generally, high content of DOM decreases sorption onto MPs through its competition with pollutants to bind to sorption sites and through complexation with the hydrophobic parts of humic and fulvic acids, which modifies the partitioning between the solid surface and water^[115]. Increasing the concentration of humic acid (25 mg/L) reduces PFAS adsorption on PA, with PFOS and PFOA showing a significant decrease (from 92% to 26% and from 20% to 3.2%, respectively), indicating a negative impact. However, the effect of DOM was less significant for short-chain PFAS (PFBuA and PFPeA).

The adsorption affinity of PFAS onto MPs can be affected by various factors in aquatic matrices, such as intermolecular interactions, electrostatic attraction/repulsion, hydrophobic interactions, salting-out effect, cation bridging, and water cluster formation^[116,117]. Previous studies have revealed two significant impacts of aquatic chemistry on PFAS adsorption onto MPs. Firstly, ionized compounds adsorb more strongly onto PS in seawater than in freshwater due to the salting-out effect and cation bridging between PFAS and the PS surface^[103]. However, the neutral PFAS, FOSA, adsorbs more in freshwater. Secondly, seawater has an overwhelming impact on hydrophobic interactions, resulting in no increasing trend in adsorption coefficients with PFAS chain length in seawater as opposed to freshwater^[9,11,14]. Notably, the scope of research on the influence of different environmental factors on PFAS sorption by MPs is broad, and future studies will likely continue to explore these properties to better understand the behavior of MPs and PFAS in the environment.

PFAS and MPs interaction mechanisms

MPs and PFAS compounds exhibit complex interaction mechanisms in the environment. Figure 5 provides a summary of previous research on the adsorption of PFAS compounds by various MPs and the factors that can impact the intricate interaction between PFAS and MPs in aqueous environments. Gagliano *et al.* found that the sorption processes of PFAS and MPs are influenced by their physicochemical characteristics and solution chemistry^[118], with effective dominant sorption mechanisms being partitioning, electrostatic and hydrophobic interactions^[116].



Figure 5. Different interaction mechanisms of PFAS sorption on MPs: (A) pi-interaction, and cation interaction; (B) electrostatic interactions; (C) hydrogen and halogen bonding; and (D) hydrophobic interactions. MPs: Microplastics; PFAS: per- and polyfluoroalkyl substances.

Partitioning has been suggested as a mechanism for PFAS retention on MPs by several studies^[9,22,23,31,119]. This process involves the distribution of PFAS and MPs between different phases (solid, liquid, and gaseous) and can influence their transportation, fate, bioavailability, and toxicity. For instance, Wang *et al.* attributed sorption differences of PFOS and FOSA by PE, PS, and PVC to MPs' polarity and characteristics^[22]. The study revealed that PFOS, being more polar, partitioned more to PVC, while non-ionic FOSA showed the highest partitioning to PE and the least to PS MPs.

Hydrophobic interactions have been identified as a significant mechanism in sorption behaviors, with longchain PFAS, being more hydrophobic, demonstrating a preference for adsorption compared to short-chain PFAS compounds^[9]. Likewise, Dai *et al.* reported that the sulfonate moiety, being slightly larger than the carboxylate moiety, resulted in an enhanced hydrophobicity during PFAS sorption on MPs surfaces^[90]. In general, greater sorption of FOSA on PE MPs compared to PVC and PS MPs, as shown by Wang *et al.*, can be attributed to the higher hydrophobicity of PE MPs^[22]. The hydrophobicity of typical MPs follows the order: of PP > PE > PS > PC > PVC, thereby confirming that an increase in hydrophobicity can enhance the sorption of PFAS compounds^[28,116]. The sorption of PFASs with the same carbon chain but different functional groups onto MPs increased in the following order: carboxylic groups < sulphonamide groups < sulphonates groups^[9,22].

PFASs are different from other hydrophobic contaminants due to the presence of hydrophilic (COOH) and polar groups (SO₃H) in the head of their molecules^[75-110]. This suggests that factors beyond hydrophobic interactions, such as electrostatic or hydrogen bond interactions, may also play a significant role in PFAS adsorption on MPs^[109]. Electrostatic interaction between MPs and PFAS depends on their respective charges, which can be influenced by factors such as functional groups, chemical composition, and

environmental conditions. Opposite charges typically result in electrostatic adsorption, while the same charge can lead to repulsion. For example, PS MPs showed higher sorption capacity for FOSA and PFOA than polyethylene terephthalate (PTFE), PE, and PVC MPs under pH-controlled conditions^[54]. The positive zeta potential of PS facilitated strong electrostatic attraction to the negatively charged PFASs, unlike the other plastics^[104,105]. MPs sorbed with PFOA exhibited a negative surface charge, causing the repulsion of additional PFOA adsorption. Therefore, FOSA (positively-charged amide group), which is readily present in water was found to sorb more quickly on negatively-charged MPs than PFOA. Nonetheless, changes in pH solution and ionic strength can easily affect the efficiency of electrostatic attraction. For instance, the sorption levels of PFOS on PE and PS MPs can be improved by increasing the concentration of common salts in natural water systems, highlighting the role of electrostatic interactions^[38,44,100]. This review suggests additional types of interactions mechanism between MPs and PFAS (hydrogen bonding, halogen bonding, pi-interaction, and cation interaction), as illustrated in Figure 5. Conclusively, there is a need for extensive research to enhance our understanding of the mechanisms behind PFAS sorption onto MPs. This is crucial for predicting PFAS's environmental fate, developing effective mitigation strategies, and combating their toxicity in the environment.

EFFECT OF MPS AND PFAS COMBINED EXPOSURE IN THE AQUATIC ENVIRONMENT

MPs act as a transporter and carrier of PFAS, influencing the availability of PFAS present in the aquatic environment. In this context, alterations in the sorption capacity of MPs can lead to changes in the bioavailability of uncoupled and coupled PFAS with MPs. Figure 6 depicts the occurrence of MPs and PFAS in different aquatic organisms. Studies have indicated that the binding of PFAS to MPs can make them more resistant to degradation in the environment and potentially increase their absorption by different aquatic organisms. The simultaneous exposure of these two pollutants can result in diverse toxic impacts on the organisms present in the aquatic environment^[22]. For instance, studies have investigated that the toxic impact on fish can intensify when they are exposed to MPs and PFAS in conjunction. This may be because MPs and PFAS together can cause higher exposure levels and more disruptions in the regular operations of fish organs and the immune system^[10]. The accumulation of MPs in various fish species was mainly observed in their gill and intestine, whereas PFAS were primarily found in their digestive and reproductive body parts. Also, the accumulation of F-53B in zebrafish larvae was found to be decreased because of the increased sorption rate of F-53B on PS MPs^[120]. PFAS can enter the phospholipid layer and makes it easier for MPs to penetrate the epithelium of the small intestine and enter the bloodstream^[121]. As a result, the number of MPs in various organs and tissues such as the liver and brain may increase, which could potentially lead to an increase in both hepatotoxicity and neurotoxicity.

Both MPs and PFAS can cause harmful effects on the growth, behavior, and molecular toxicity of aquatic organisms. However, the toxic effects of MPs on the microbe's digestive system are distinct from the effects of PFAS. PFAS primarily induces toxicity in the sex and reproductive systems, with a significant impact on the sex ratio^[22]. It is believed that concurrent exposure to both MPs and PFAS may result in diverse toxic effects on aquatic species, which may differ from the effects of MPs or PFAS individual exposure. The potential consequences of combined exposure to MPs and PFAS on aquatic life are illustrated in Figure 7. To date, there are very few studies that have explored the hazards caused by the co-existence of MPs and PFAS in the environment.

Combined exposure to PS MPs and PFAS was found to decrease the production of reactive oxygen species and the cell permeability of bacteria, which was different from the effect observed with exposure to MPs alone^[9,122,123]. This may be due to the fact that PFAS compounds stimulate the production of extracellular polymeric substances. The entering of MPs and PFAS complex into the aquatic organisms' stomach at a low



Figure 6. The occurrence of (A) MPs and (B) PFAS in different aquatic organisms. MPs: Microplastics; PFAS: per- and polyfluoroalkyl substances.

pH value results in the protonation of MPs. Positively charged MPs can attract anionic PFAS and repel cationic PFAS, which could result in an increase in the bioavailability of cationic PFAS and a decrease in the bioavailability of anionic PFAS^[22]. In such a scenario, the uncoupled anionic PFAS bioavailability decreases as they are adsorbed onto MPs resulting in the MPs-PFAS complex in the aqueous phase. Contrarily, the bioavailability of the uncoupled cationic PFAS increases as they ripple to the positively charged MPs. The synergistic effects of MPs and PFAS co-existence in several species are not well understood and future research is warranted. In general, the potential harm caused by the presence of MPs and PFAS as a complex in aquatic environments is a matter of increasing concern. To decrease the harmful impacts of MPs and PFAS can potentially engage together with other micropollutants present in the aquatic environment. This can lead to



Figure 7. Distribution and toxicological effects of MPs and PFAS co-existence in aquatic matrices^[90,120,121]. MPs: Microplastics; PFAS: perand polyfluoroalkyl substances.

alterations in their chemical properties, resulting in an increased toxicological impact on aquatic species which needs to be addressed.

Synergistic ecotoxicological effects of MPs and PFAS

In recent years, there has been growing concern about the persistence of MPs and PFAS in various parts of the ecosystem which have harmful impacts on living organisms. Studies have demonstrated that the coexistence of MPs and PFAS can intensify each other's toxicity, resulting in more pronounced adverse impacts on the health and survival of living organisms^[28]. The reason behind the heightened synergistic ecotoxicological effects of MPs and PFAS may be the increased uptake and bioaccumulation of PFAS in living organisms that are exposed to MPs. MPs can serve as carriers for PFAS, thereby increasing the bioavailability of PFAS coupled with MPs and facilitating their transfer across biological membranes. This can result in the accumulation of higher concentrations of PFAS in the tissues of organisms, which can lead to more pronounced toxic effects^[119].

Research on the synergistic ecotoxicological effects of MPs and PFAS is currently limited. Rainieri *et al.* conducted a study on zebrafish, investigating the effects of different feeding regimes involving MPs, individual PFAS compounds, and MPs-containing PFAS^[124]. Results indicated that the combined exposure of zebrafish to MPs and PFAS sorbed to MPs disrupted the homeostasis of the fish's organs, particularly the liver, to a significantly greater extent than exposure to either contaminant alone. Reportedly, earthworms exposed to soil contaminated with MPs showed an increase of approximately 2.5 times in the bioaccumulation factor for PFOA and PFOS^[27]. Notably, mussels that were contaminated with MPs showed higher bioaccumulation rates indicating that they served as a medium for the accumulation of PFAS^[125]. Studies have demonstrated that the presence of MPs can affect the levels of PFAS in an organism's body. For instance, Islam *et al.* conducted an experiment where they exposed the clam *Scrobicularia plana* to different size fractions of MPs either with or without PFOS sorption to assess the environmental threats posed by these emerging pollutants^[126]. Results highlighted that the size of MPs appeared to have a more significant impact on the ingestion of MPs and the accumulation of PFOS in clams than the presence of PFOS itself.

Chen *et al.* investigated the combined toxicity of MPs and ammonium perfluorooctanoate (APFO) on *Daphnia magna* and observed that the effects varied depending on the concentration ratios of the co-exposure^[127]. The study identified three modes of interaction: synergism, antagonism, and partial addition. The findings demonstrated that different ratios of toxic units resulted in differing levels of gut fullness in *Daphnia magna*. The combination of MPs and PFAS in the ecosystem can have both negative and positive impacts. While co-exposure may have detrimental effects, it can also potentially reduce the harmful impact of certain types of pollution.

Several studies suggest that MPs ingestion can be advantageous in species' digestive systems by reducing the bioaccumulation of certain pollutants through sorption behaviors, which is commonly referred to as the "clean effect"^[128]. For instance, the collective toxicity of MPs and PFOS in human intestinal cells revealed that when MP particles were detected in low PFOS concentrations, the negative impacts of PFOS were decreased due to the reduction in the bioavailability of PFOS which are coupled with MPs^[129]. It is worth noting that the adsorption of pollutants by MPs is a reversible process thereby allowing the desorption of the same. The level of sorption and desorption of MPs in an organism can affect the bioaccumulation, bioavailability, and toxicity of sorbed pollutants, as well as their transfer to tissues and organs^[130]. The synergistic ecotoxicological effects of MPs and PFAS emphasize the need to consider the complex interactions among different contaminants in the ecosystem and their potential impacts on living organisms. It also highlights the necessity for research to enhance the understanding of the mechanisms and magnitude of these interactions, as well as to establish effective strategies for mitigating their negative impacts.

CONCLUSIONS AND FUTURE PERSPECTIVES

The co-existence of MPs and PFAS in the environment poses a serious threat to aquatic systems, with MPs acting as a carrier of PFAS and leading to the source of secondary pollution. The review summarizes their co-occurrence, distribution, sources, influencing factors, interaction mechanisms, and toxic effects caused by their combined exposure. Food containers and textiles are major sources common for MPs and PFAS, however, their combined release from these sources has not received much attention. In different environmental compartments, the MPs concentrations range from 0 to 6.64 particles/m³ in the air, 1.3 to 4712 particles/kg in soil, 0.57 to 26 particles/L in aquatic matrices, and 0 to 198.82 particles/organism in living organisms. Moreover, the analysis of existing literature reveals a wide range of PFAS concentrations, with values ranging from < 0.0011 to 95.378 ng/m³ in the air, 6.90 to 294000 ng/kg in soil, 4 to 268 ng/L in aquatic environments, and 0.0014 to 796 ng/g in living organisms. The interaction between the two pollutants is reportedly is affected due to various environmental factors and MPs-PFAS properties. In addition, ingestion of PFAS-sorbed MPs by organisms can result in more severe combined toxic effects compared to exposure to each pollutant alone. The scarcity of studies on the interaction between MPs and PFAS in real environmental settings limits their potential toxicity, highlighting the need for greater emphasis on this topic. Overall, it is vital to determine the relative importance of MPs as a carrier of emerging contaminants in the environment, in comparison to other routes of exposure. Therefore, a comprehensive risk assessment is necessary to evaluate the combined exposure and coexistence of MPs and PFAS in the aquatic environment, as well as their potential as a source of secondary pollution.

DECLARATIONS

Authors' contributions

Investigation, resources, formal analysis, visualization, writing-original draft: Parashar N Writing-original draft, editing, validation, and visualization: Mahanty B Conceptualization, methodology, supervision, writing-review & editing: Hait S

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