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Research Paper

Effects of microplastics and food particles on organic pollutants bioaccumulation in equi-fugacity and above-fugacity scenarios

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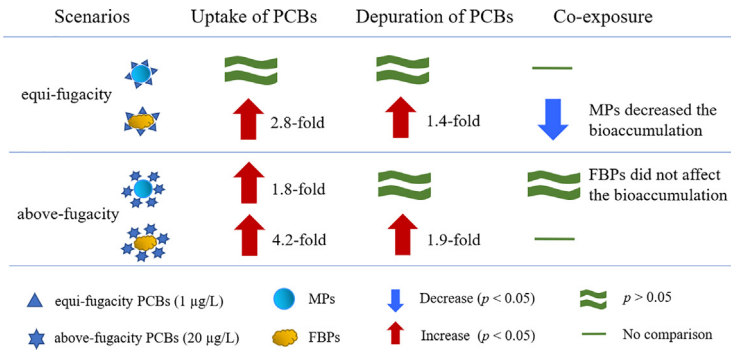
HIGHLIGHTS

- Microplastics had limited effects on PCBs bioaccumulation in equi-fugacity scenario.
- Microplastics enhanced the bioaccumulation of PCBs in above-fugacity scenario.
- Food is easily assimilated by fish, making associated PCBs more bioavailable.
- Food increased the bioaccumulation of PCBs under both scenarios.
- Microplastics facilitated depuration of PCBs accumulated via food.

GRAPHICAL ABSTRACT



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ABSTRACT

Microplastics (MPs), as emerging contaminants, sorb organic pollutants from the environment or leach out additives, thereby altering the fate of co-existing pollutants to organisms. We chose equi-fugacity and above-fugacity concentrations of polychlorinated biphenyls (PCBs) as background contamination and plastic additive concentrations, respectively, to investigate the effects of MPs on PCB bioaccumulation; we compared the effects of MPs with those of food-borne particles (FBPs). Co-exposure to MPs and FBPs at both the equi-fugacity and above-fugacity PCB concentrations had no obvious toxic effects (ROS generation and *cyp1a* expression) on zebrafish. When the zebrafish were exposed to the equi-fugacity PCB concentrations, the PCB concentrations reached 177.7–400.5 ng/g after a 7-d uptake; the presence of MPs did not significantly enhance PCB bioaccumulation. The remaining PCB concentrations in the fish after a 4-d depuration were 58.4–125.1 ng/g; the effects of MPs were the same as those during the uptake period. However, at the above-fugacity PCB concentrations, the MPs markedly increased the PCB bioaccumulation (by 1.8–fold) to 712.9 ng/g. This is because at above-fugacity concentrations, PCBs on MPs migrate to organisms as there were high fugacity gradients. The FBPs enhanced PCB bioaccumulation in zebrafish more effectively than the MPs, even after depuration. In the presence of FBPs, PCB bioaccumulation increased by 2.8- and 4.2- fold after uptake in the equi-fugacity and above-fugacity scenarios, respectively, both of which were significantly higher than that observed for the MPs. This is probably because FBPs are easily assimilated by fish, making the associated PCBs more bioavailable. Finally, during the co-existence of MPs and FBPs, MPs facilitate the depuration of PCBs accumulated via FBP vectors; conversely, FBPs did not affect PCB accumulation via MP vectors. Thus, this study elucidated the effects of MPs and FBPs on the bioaccumulation of pollutants at equi-fugacity or above-fugacity concentrations in aquatic environments.

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1. Introduction

Pollution caused by microplastics (MPs, plastic particles <5 mm in size) has become an emerging issue due to their ubiquitousness in the atmosphere, aquatic, and terrestrial environments (Auta et al., 2017; Chen et al., 2020a; Yu et al., 2020). MPs have the following characteristics: small particle size, floatability, and refractory degradation; thus, they can be randomly ingested by organisms and cause direct adverse effects. These effects include physiological damage (Wright et al., 2013), retardation of growth and development (Mak et al., 2019; Pannetier et al., 2020), or even death (Ziajahromi et al., 2017). Meanwhile, indirect effects that arise via MPs as vectors for organic pollutants are also worthy of our attention (Menéndez-Pedriza et al., 2022). MPs can transport co-existing pollutants and lead to the alteration of their bioaccumulation in organisms (Tanaka et al., 2020; Tourinho et al., 2019), which may consequently change the toxic effects of these pollutants (Browne et al., 2013; Zhu et al., 2018).

Pollutants associated with MPs can originate via sorption from the surrounding environment (Chen et al., 2021; Tang et al., 2021) or additives incorporated during manufacturing (Nurlatifah et al., 2021). Regardless of the source, MPs will release pollutants after entering organisms due to desorption (Browne et al., 2013; Chen et al., 2020b; Coffin et al., 2019). After the exposure of contaminated MPs to clean test organisms, the concentration levels of organic pollutants in these organisms will increase (Xia et al., 2020). For instance, the bioaccumulation of phenanthrene in zebrafish was shown to increase from 0.268 ± 0.054 μg/g to 0.475 ± 0.052 μg/g in the presence of polystyrene MPs (Xu et al., 2021). However, such exposure scenarios may not be representative of those occurring in the natural environment. In environments with organic pollution, the impact of MPs on the bioaccumulation of pollutants can reduce or be negligible compared with that in an environment without organic pollutants (Besseling et al., 2017; Zhu et al., 2018). Koelmans and other researchers have reported that the MP-mediated cleaning or scrubbing effects of hydrophobic organic pollutants (HOCs) in organisms may be more prominent than the vector effects of MPs in an open coastal environment (Besseling et al., 2013; Koelmans et al., 2013). The main reason for this contradiction is the different exposure scenarios that were considered (Ziccardi et al., 2016); such scenarios have not been distinguished well in previous studies. When MPs accumulate HOCs from the surrounding aquatic environment, this process will continue until the fugacity of the MPs and water is equal, at which time the exposure gradient of the MPs towards the fish will not increase. When HOCs, such as phthalates, are added during the manufacturing process of MPs, the HOCs loaded onto the MPs will have an above-fugacity state with regard to the surrounding water/organisms. Fugacity is a thermodynamic quantity related to chemical potential or activity that characterizes the escaping tendency of pollutants from a phase. At equilibrium, the fugacities (depicted via units of pressure; Pa) of the MPs and surrounding water are equal (Mackay and Paterson, 1982).

When MPs and food-borne particles (FBPs) mainly picked up pollutants from the surrounding environment that they were exposed to, their concentrations may reach the same level in exposure scenarios. However, their concentration is not a good indicator to represent their chemical activity. As MPs are indigestible, the migration of the pollutants from MPs to organisms mainly depends on the concentration of the associated pollutants. Because FBPs can be digested and assimilated, the gastrointestinal (GI)-magnification effect can occur in organisms, which may enhance their chemical activity and fugacity over the course of their migration from the digestive tracts to organisms. Thus, fugacity can better illustrate the possible direction of the migration of pollutants than the pollutant concentration (Gobas et al., 2021). Ultimately, the ingested food fugacity in an organism is the combination of its food basket, i.e., the weighted concentration of different food items it eats or the fugacity of the different HOCs within these items weighted by the proportion of each food item in the overall diet of the animal. The MPs in these foods are just a part of the food basket but possess a different digestibility (zero digestibility) compared with that of the other ingested food items.

It has been reported that in living organisms in which FBPs and MPs are jointly present, the abundance of MPs accumulated will decrease. For instance, the presence of abundant FBPs reduced the ingestion of polyethylene MPs in *Tripneustes gratilla* sea urchin larvae by 4–10-fold (Kaposi et al., 2014); this was because organisms can distinguish nutritious food from other suspended particles. However, the extent to which FBPs and other environmental matrices can affect the bioaccumulation of MPs and co-existing pollutants is largely unknown and deserves more research.

Polychlorinated biphenyls (PCBs) are typical classical HOCs; they can easily partition into lipids and plastic particles (Kodavanti and Loganathan, 2014; Velzeboer et al., 2014), and served as flame retardants that were added to plastics (Erickson and Kaley, 2011). Although PCBs are no longer used as additives and generally reach equilibrium in the environment, they can be used as model additives to study the bioaccumulation effects of both traditional and emerging flame retardants on organisms. PCBs are still widely detected in plastic debris despite they have been phased out, most of which had similar fugacities with the surrounding aquatic environment. Furthermore, the scenario of above-fugacity can still be seen in case of some newly disposed plastics or MPs transported from highly polluted wastewater. Where the MPs pick their PCB loads from, i.e., from wastewater vs ambient water, can determine whether or not the MPs achieve an equi-fugacity state with ambient water. The fugacity of MPs in organisms is fixed due to their indigestibility; the differences in their concentrations can be directly translated into differences in their fugacity. However, the fugacity of FBPs can be magnified in the GI tract. To the best of our knowledge, this study is the first to systematically provide an objective theoretical basis for understanding the role played by MPs as carriers in affecting the bioaccumulation of co-existing pollutants in the presence or absence of food particles.

2. Materials and methods

2.1. Materials

Polyethylene MPs (180–430 μm) were supplied by Guanbu Electromechanical Technology Co., Ltd. (Shanghai, China). PCB standards were purchased from Anpel Laboratory Technologies Inc. (Shanghai, China). Other reagents were of analytical purity and purchased from Titan Technology Co., Ltd. (Shanghai, China). We choose six kinds of PCBs and divide them into 3 pairs of PCBs with similar properties (log $K_{ow,}$ log K_{PEw} and molecular weight) (**Table S1**). The two PCBs within a pair were spiked into artificial freshwater (AFW) as an equi-fugacity state (1 μg/L), and an above-fugacity state (20 μg/L), respectively. Food particles were purchased from Yuyue Pet Products Store (Shanghai, China), and then they were ground and sieved into 250–300 μm FBPs, which size was similar to the dominant size range of MPs. The morphology, distribution, and polymer composition characteristics of MPs and FBPs are shown in **Fig. S1**. One hundred particles of MPs and FBPs were taken to measure their size distributions ($n = 6$). The surface of MPs is relatively smooth overall, and the partition capacity of pollutants on MPs depends on the polyethylene (PE)-water equilibrium partitioning constants (Lohmann, 2012; Wang et al., 2021). While the FBPs surface is more porous with greater surface heterogeneity, and the lipid, protein, and moisture contents of FBP are approximately 5%, 41%, and 11%, respectively. According to (Debruyn and Gobas, 2007), the sorptive capacity is not only contributed by lipids, but also non-lipid parts (such as proteins) that can't be ignored. For hydrophobic chemicals, the food particles/water partition coefficient (K_{FW}) should be calculated as $K_{FW} = (Ø_L + 0.05Ø_{NLOM}) K_{OW}$, where $Ø_L$ and $Ø_{NLOM}$ are fractions of lipid and non-lipid organic matter, respectively. These properties made the partition of FBPs relative to octanol was different compared to MPs.

2.2. Preparation of PCBs associated MPs and FBPs

All glassware used for the sorption experiment of PCBs associated MPs and FBPs was soaked with a dilute nitric acid solution for 24 h before use, and rinsed with ultrapure water after washing, and then they were

transferred to a muffle furnace at 450 °C for 4 h burning at least. First, we conducted a preliminary experiment of sorption kinetics to determine the time required for sorption equilibrium. MPs (0.2 g) were added into each 40 mL-brown glass bottle, and stock PCB solutions in n-hexane were spiked into bottles to assure a final concentration of 100 μg/L in AFW and the final n-hexane concentration was below 0.1%. The samples were shaken continuously at 80 rpm and 25 ± 1 °C for 96 h in a thermostatic oscillator (HZ-9612 K, China). The sampling time points were at 8, 12, 24, 48, 72, and 96 h.

The sorption of PCBs on MPs is mainly divided into two processes: surface adsorption and intraparticle diffusion (Tourinho et al., 2019). The first step of surface adsorption is dominated by the specific surface area of MPs, which can reach equilibrium in a short time (Guo et al., 2018). Kinetic experiments suggested that the sorption reached pseudo-equilibrium within 24 h (Xu et al., 2019) which is also verified in the present study (**Fig. S2**). After sorption, PCBs were extracted with the Oasis HLB μ-Elution extraction columns (Waters, Milford, USA). Two hundred microliter methanol and water were eluted through to active the columns successively, and then 750 μL of sampled solutions and 200 μL of Milli-Q water were loaded on columns successively. After loading, PCBs on columns were eluted by adding 75 μL of methanol. The activation, loading, and elution flow rates were set at 1.0 mL/min. Finally, internal standards were spiked into the eluted solutions to reach a concentration of 20 μg/L. The recovery rate of PCB samples was 71.0–89.5%. We measured the concentrations of PCBs remaining in the aqueous solution, which were converted to the sorbed PCB concentrations on MPs and/or FBPs.

For the preparation of associated PCBs on MPs or FBPs, we added 200 mg of MPs or FBPs into each 40 mL-brown glass bottle, and stock PCB solutions were spiked into bottles to achieve a final concentration of 1 μg/L or 20 μg/L in AFW with n-hexane concentration below 0.1%. The PCB concentrations in the solution were extracted by Oasis HLB μ-Elution columns as described above. The MPs and FBPs particles in the aqueous solution are obtained by filtration.

Sorption kinetics results showed that the six PCB congeners required 24 h to reach sorption pseudo-equilibrium. When the concentration of the PCB congeners was 1 μg/L (equi-fugacity scenario), the sorption amount of PCBs on MP was around 0.26 μg/g, and that on FBP was around 0.34 μg/g. When the concentration of PCBs was 20 μg/L (above-fugacity scenario), the sorption amount of PCBs on MP was around 3.9 μg/g, and that on FBP was around 4.6 μg/g (Fig. 1a and b). The fugacity of MP was not equal to the fugacity of FBP in the equilibrated and unequilibrated treatments (**Table S2, S3**), which caused a significant difference ($p < 0.05$) between the sorption amount on MPs and FBPs under both PCB exposure scenarios.

2.3. Zebrafish uptake and depuration experiments

Adult wild-type zebrafish (*Danio rerio*) were purchased from FishBio Co. (Shanghai, China), and the male: female ratio is 1:1. The fish were acclimatized in a circulating water system with a constant temperature of 25 ± 1 °C, 14:10 h of light: dark for one month before exposure.

The PCBs uptake and depuration experiments were divided into five groups: [Control], [PCB], [PCB + MP], [PCB + FBP], and [PCB + MP + FBP] (Fig. 1c). The specific group settings are as follows: [Control]: AFW solution; [PCB]: 1 μg/L PCBs in the solution; [PCB + MP]: both equi-fugacity and above-fugacity PCBs on MPs, and 1 μg/L PCBs in the solution; [PCB + FBP]: both equi-fugacity and above-fugacity PCBs on FBPs, and 1 μg/L PCBs in the solution; [PCB + MP + FBP]: equi-fugacity PCBs on FBPs and above-fugacity PCBs on MPs, and 1 μg/L PCBs in the solution.

Feeding was fixed within two hours from 9:30 to 11:30 in the morning and take place every day during the 7-d uptake period. When group [Control], [PCB], [PCB + MP] were fed clean food, [PCB + FBP], [PCB + MP + FBP] were fed FBP particles loaded with different PCB concentrations. Each group was fed clean food during the 4-d of the depuration period. It was found that zebrafish stopped feeding behavior after two

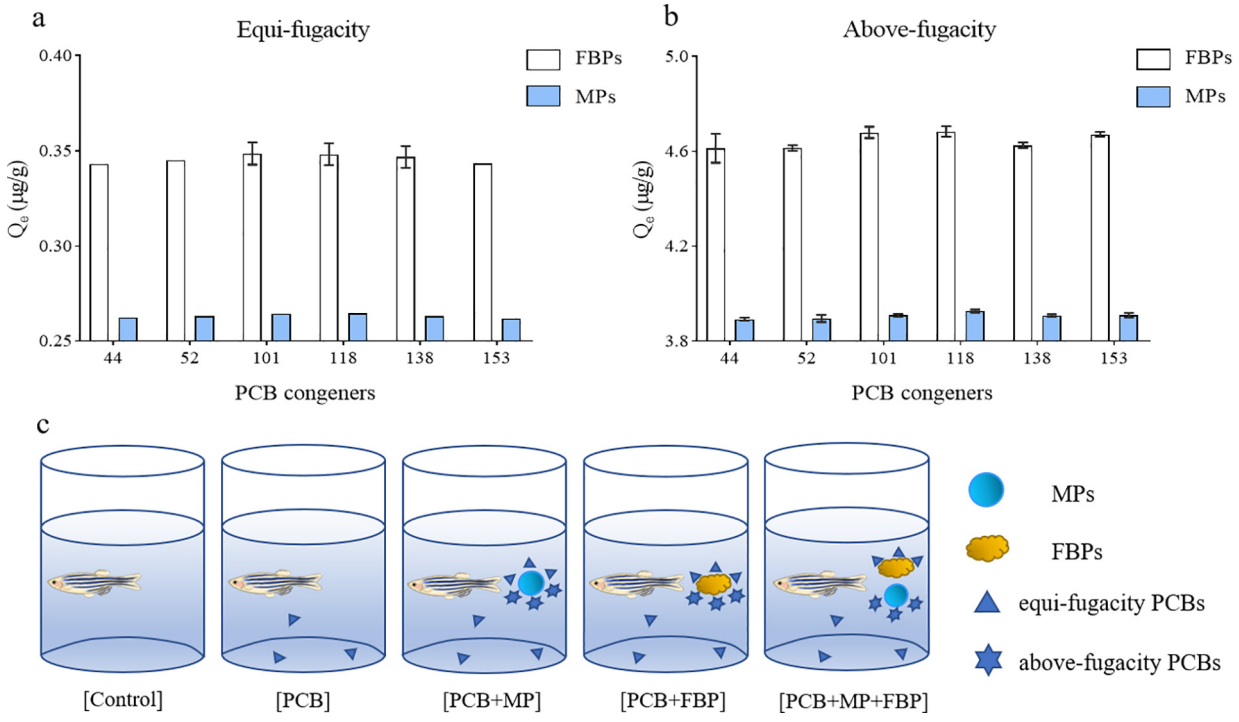

Fig. 1. (a) Sorption amount of PCBs onto polyethylene MPs and FBPs when the aqueous PCB concentrations were in the equi-fugacity scenario, and (b) in the above-fugacity scenario; (c) Set-up of exposure groups for PCBs bioaccumulation analysis in zebrafish with or without the presence of MPs/FBPs. Blue particle: MPs; Brown irregular particle: FBPs; Blue triangle: represents equi-fugacity PCBs (1 μg/L); Blue hexagon: represents above-fugacity PCBs (20 μg/L). Note: [Control]: AFW solution; [PCB]: 1 μg/L PCBs in the solution; [PCB + MP]: both equi-fugacity and above-fugacity PCBs on MPs, and 1 μg/L PCBs in the solution; [PCB + FBP]: both equi-fugacity and above-fugacity PCBs on FBPs, and 1 μg/L PCBs in the solution. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

minutes, which made it negligible for the adsorbed PCBs desorbing into the aqueous solution. Fresh PCB contaminated water refreshed each day of the 7-d uptake period and after every feeding event. For MPs containing groups, the added amount of MPs was 300 particles/L (which is an environmentally relevant concentration (Dubaish and Liebezeit, 2013)). For FBPs containing groups, the amount of FBPs was 5% of the wet weight of zebrafish, the concentration of FBPs is about 360 particles/L. After feeding, aqueous solutions for each group were completely renewed, and water samples were taken before and after feeding to determine the total organic carbon (TOC) by a TOC-V device (Shimadzu, Japan) to calculate the feeding rate of zebrafish. For each sampling, took 500 μL of water samples from the surface, middle, and bottom layers of the aqueous solution and mixed them into one sample. And avoid disturbing zebrafish when sampling, we used a glass rod to slightly agitate the aqueous solution while adding the food to make the particles evenly distributed.

The uptake period lasted for 7 d, followed by a 4-d depuration. Fish samples were taken and dissected on the 1st, 3rd, 5th, and 7th d during the uptake and on the 1st, 3rd, and 4th d during the depuration. At each sampling point, six fish (three males and three females) were taken from each group. Then, the fish were euthanized using benzocaine solution (200 mg/L) and dissected. One male and one female fish were pooled together as one replicate. All samples were fast frozen with liquid nitrogen immediately after dissection, and stored in a refrigerator at −80 °C for further analysis. The conduction of zebrafish experiments was obedient to the requirements of the Animal Experimental Ethics Committee of East China Normal University, Shanghai, China (Ethical review number: Z20180301).

2.4. Quantification of MPs in the fish

The intestines and gills of each fish were collected for MPs quantification, the number of replicates is three to ensure the accuracy of the experimental data. Fish intestines and gills were transferred to 50 mL centrifuge tubes, and 10 mL of 30% hydrogen peroxide were added. Then, the tubes were shaken in an incubator at a constant temperature of 65 °C at 80 rpm for 24 h. The digested solutions were filtered through hydrophilic polycarbonate membranes (5.0 μm pore size, Merck Millipore, Germany), and residues on filter membranes were observed on an Olympus microscope (BX53, Japan) to quantify MPs concentrations numerically in intestines and gills.

2.5. Zebrafish mRNA expression analysis

The livers of every two zebrafish (1 female and 1 male) were pooled together and used to measure the mRNA expression of the *cyp1a* gene at the end of uptake and depuration periods according to Zhao et al. (Zhao et al., 2017), we performed 3 replicates at each point to test in the toxicity test. Frozen zebrafish livers were homogenized in Trizol Reagent (Invitrogen, USA) for RNA extraction and subsequently centrifuged at room temperature for 10 min at 12 000 g. Total RNA concentrations were determined by the 260/280 nm ratio using a NanoDrop 2000 Spectrophotometer (Thermo Fisher, USA). cDNA was obtained via reverse transcript from RNA samples using cDNA reverse transcription kits (Takara, Japan). Quantitative reverse transcriptase-polymerase chain reaction (qRT-PCR) assay was performed using SYBR Green Master Mix (Takara, Japan) and the fluorescence quantitative PCR instrument (Bio-Rad CFX Connect System, USA). Primer sequences for *cyp1a* and reference gene *gapdh* were F: AAAGACACC TGCGTGTTTGTAA and R: GAGGGATCCTTCCACAGTTCT, and F: TTCCAG TACGACTCCACCCA and R: TGACTCTCTTTGCACCACCC. The *cyp1a* gene's expression was normalized to the expression of reference gene *gapdh*.

2.6. Reactive oxidative species measurement

To evaluate the oxidative stress, the reactive oxidative species (ROS) content was measured with zebrafish liver samples collected on the 1st, 3rd, and 5th d of uptake and 1st and 2nd d of depuration with a ROS commercial kit (Nanjing Jiancheng Bioengineering Institute, Nanjing, China) by using the dichlorofluorescein diacetate (DCFH-DA) method (excitation and emission wavelengths were of 502 nm and 530 nm, respectively) and a bicinchoninic acid (BCA) protein quantification kit according to manufacturer's instructions.

2.7. Sample preparation and PCBs quantification

The zebrafish muscle samples without gills, liver, and intestines were extracted to measure PCB concentrations. First, zebrafish muscle samples were freeze-dried on a lyophilizer (Trx-FD-1A-50, Shanghai, China) after 4 h of pre-frozen at −80 °C. Then, two zebrafish muscle samples were ground into powders with mortars and pestles, which were mixed with diatomaceous earth and later extracted in an accelerated solvent extraction equipment (ASE, Dionex 350, USA) with 30 mL dichloromethane/n-hexane (1:1, *v*/v) at 1500 psi and 100 °C. The extraction procedure for ASE was set as follows: static extraction time: 10 min; solvent flushing time: 180 s; the number of cycles: two. After extraction, samples were further concentrated on a rotary evaporator (Senco R206D, Shanghai, China) at 45 °C and 100 rpm to around 1 mL. Then, samples were purified with gel chromatography columns which had been packed with 5.0 g of 100–140 mesh silica gel (burned at 180 °C for 8 h), 5.0 g neutral alumina (burned at 270 °C for 8 h), and 0.5 g anhydrous sodium sulfate (burned at 450 °C for 8 h) from bottom to top with a volume ratio of 12:6:1, and pre-activated with 10 mL of n-hexane. Later, 10 mL of hexane/acetone (1:1, *v*/v) were added to wash gel chromatography columns, and the eluted extracts were collected and rotated again to leave about 1 mL, which were blown to dryness by a nitrogen blowing concentrator (MD 200, China) and used 60 μL of n-hexane and 20 μL of 100 μg/L of PCB 31 and PCB 180 each (internal standards) for solvent replacement, to achieve a final solution volume of 100 μL.

PCB concentrations in samples were quantified by a gas chromatography-mass spectrometer (GC–MS) (7010B Agilent, USA), with a DB-5MS capillary column in the single ion monitoring (SIM) mode. Helium was used as carrier gas at a flow rate of 1.0 mL/min. The injection port and detector temperatures were set at 250 and 310 °C, respectively. The temperature elevation program was set as follows: 60 °C held for 1 min, then ramped to 170 °C at 40 °C/min and no hold time, and finally rose to 280 °C at a rate of 7 °C/min and kept for 8 min. Samples were injected with 1 μL in the splitless mode.

The LOD and LOQ for PCBs were 2.0–4.0 ng/g and 1.2–6.0 ng/g, respectively. PCBs standard curves were run intermittently between samples to monitor changes in the measurement sensitivity. The internal standard method was used for calibration, using PCB 31 (for PCB 44, 52, and 101) and PCB 180 (for PCB 118, 138, and 153) at 20 μg/L as internal standards. PCB 44, 52, 101, 118, 138, and 153 were quantified by comparing with the peak area of the internal standards.

2.8. Statistical analysis

The obtained results were statistically analyzed using SPSS (IBM Statistics, ver 23, USA). The mean ± standard error (SEM) was determined based on three replicates for each experimental treatment. Statistical differences among means were assessed by the one-way analysis of variance (ANOVA) method followed by Turkey's posthoc test. The statistical difference was set at $p < 0.05$.

3. Results and discussion

3.1. Toxicity assessment

3.1.1. cyp1a mRNA expression

PCB exposure may induce aromatic hydrocarbon receptor (AhR)-dependent endpoints changes in aquatic organisms at high concentrations (Zhou et al., 2010). However, in this study, it was found that there were no obvious toxic effects after exposure to environmentally relevant concentrations of MPs. CYP is an important detoxification enzyme in organisms that

can detoxify exogenous substrates; its gene expression is regulated by AhRs (Navas and Segner, 2000; Wu et al., 2001). When the presence of aromatic compounds induces *cyp1a* expression, high mRNA levels and activity of CYP can be detected. In this study, we investigated the expression of *cyp1a* gene to determine the toxic effects of PCB exposure on the detoxification metabolism system in zebrafish. Our results showed that the exposure to MPs alone or the co-exposure to MPs and PCBs did not result in marked changes in the *cyp1a* mRNA expression levels in zebrafish, comparison with the case for the control during both the uptake and depuration periods; however, some fluctuations in the *cyp1a* mRNA expression levels were observed (**Fig. S3a and b**).

3.1.2. ROS content

Moreover, no significant differences of ROS content in the exposure groups were found relative to the controls, except for the [PCB + FBP] group on day 1 (**Fig. S3c**). However, with the extension of the uptake time, the level of oxidative damage in the zebrafish decreased, and no significant differences were found between the [PCB + FBP] and control groups after day 3 (**Fig. S3c and d**). As the increase of ROS content reflects cellular oxidative damage in fish (Jin et al., 2020), our results imply that the exposure to PCBs and MPs did not cause notable oxidative damage to the zebrafish throughout the experiment; thus, PCB bioaccumulation will not be negatively impacted. This also indicates that our experimental setting is reasonable, and is thus, favorable for observing the differences in PCB bioaccumulation in the presence of equi-fugacity and above-fugacity PCB concentrations.

3.2. Interaction of MPs and FBPs during the exposure

3.2.1. MPs accumulation in zebrafish

The concentrations of MPs were the highest in both the intestines and gills of the zebrafish after 1 d of exposure: 17.7 and 2.3 MPs items/tissue in the [PCB + MP] group. With the extension of the experimental period, zebrafish can recognize MPs as inedible materials (Kim et al., 2019) or avoid ingesting them (Ryan et al., 2019), thereby causing a reduction in the amount of MPs ingested. Thus, the MPs concentrations gradually decreased (Fig. 2). At the end of the uptake period, the concentrations of MPs were significantly lower than that at day 1 ($p < 0.05$). The number of MPs reduced by 43% to 10.0 items/tissue in case of the intestines and by 71% to 0.7 items/tissue in case of the gills. The depuration period further decreased the MPs concentrations by 35% and 100% in the intestines and gills, respectively, but these decreases were not statistically significant ($p > 0.05$).

In the presence of FBPs, fewer MPs accumulated in zebrafish intestines (Fig. 2a). At the end of the uptake period, there was an average of 3.5 ± 1.4 items/tissue in case of intestines of the fish from the [PCB + MP + FBP] group; the MPs concentration was only 35% of that in the fish from the [PCB + MP] group. At the end of the depuration, an average of 2.3 ± 1.4 items/tissue were found in case of the intestines of the fish from the [PCB + MP + FBP] group, which was only 47% of that in the intestines of the fish from the [PCB + MP] group (Fig. 2b). These findings suggest that FBPs can reduce the accumulation of MPs in the zebrafish intestine.

The main difference in the feeding rates among the fish from the different groups probably lies in the states in which the FBPs existed during feeding. Compared with the fish in the [PCB + MP] group, which were fed with clean, unsoaked FBPs, the fish in the [PCB + MP + FBP] group were fed with contaminated soaked FBPs (which were softer and more digestible). As shown in Fig. 2g, the fish in the [PCB + MP + FBP] group ingested more food than those in the [PCB + MP] group. It has been found that the availability of food negatively affected the ingestion of MPs by organisms (Kaposi et al., 2014; Wang et al., 2019). In a nutrition-rich habitat with adequate food, zebrafish can selectively ingest available food, resulting in a reduction in the ingestion of MPs (Kim et al., 2019; Wang et al., 2019). In 2017, Grigorakis et al. found that MPs were excreted from the fish viscera at the same rate as food/feces (Grigorakis et al., 2017). Thus, since the fish in the [PCB + MP + FBP] group ingested a lesser amount of MPs than those in the [PCB + MP] group, the concentrations of remaining MPs in the zebrafish intestine were also lesser after elimination.

Much fewer MPs were accumulated in the gills of zebrafish than in the intestines (Fig. 2c and d), which was probably due to the lack of the interception functions of the gills (Batel et al., 2018; Koongolla et al., 2020); furthermore, no significant differences between the accumulated MPs concentrations in the two groups were found during the whole exposure experiment.

In general, we found that the amount of MPs in intestine in the fish from the [PCB + MP] group during the uptake was 12.8 items/tissue; the concentration of MPs remaining even after depuration was 6.2 items/tissue (Fig. 2e). The presence of FBPs significantly reduced the abundance of MPs by 39.8% and 69.4% ($p < 0.05$) after uptake and depuration, respectively. However, there were no significant differences between the abundance of MPs in the gills from zebrafish in the two MPs-exposure groups (Fig. 2f).

3.2.2. Food feeding rate during the exposure

During the uptake period, the feeding rates of the fish from the groups subjected to treatment with FBPs, namely, the [PCB + FBP] and [PCB + MP + FBP] groups (0.038 ± 0.002 mg food/mg fish/d and 0.028 ± 0.001 mg food/mg fish/d, respectively), were always higher than those of the fish from the other groups. This may be because the PCBs associated FBPs were softer than the fresh FBPs, as the former had previously been soaked in exposure solutions for PCBs sorption, which made their uptake and digestion by the fish easier. On the contrary, the fish subjected to MPs exposure had relatively lower feeding rates than those from their corresponding groups. For instance, the feeding rates of the fish in the [PCB + MP] and [PCB] groups were 0.018 ± 0.001 mg food/mg fish/d and 0.019 ± 0.001 mg food/mg fish/d, respectively, and those of the fish the [PCB + MP + FBP] and [PCB + FBP] groups were 0.028 ± 0.001 mg food/mg fish/d and 0.038 ± 0.002 mg food/mg fish/d, respectively. This may be because the MPs were excreted out more easily after the addition of the food (Khosrovyan et al., 2020). Another explanation for this is that the spitting behavior, which had been observed in zebrafish after MPs ingestion, may have led to a lower feeding rate (Kim et al., 2019). During the depuration stage, the zebrafish in all five groups were fed with clean FBPs; no differences of feeding rates (0.021–0.023 mg food/mg fish/d) were found among the groups (Fig. 2g and h).

3.3. Equi-fugacity scenario: MPs did not affect, but FBPs enhanced PCBs bioaccumulation

When PCBs were present in the aquatic environment at the equi-fugacity concentration, the total PCB concentrations were 544.0–1153.5 ng/g in the fish from the [PCB + FBP] group, which was significantly higher than that in the fish from the [PCB] (177.7–400.5 ng/g) and [PCB + MP] groups (159.4–378.2 ng/g) during the entire uptake period ($p < 0.05$) (Fig. 3a–d); the differences in the PCB concentrations in the latter two groups were not significant.

During the depuration period, significant differences among the three groups gradually disappeared (Fig. 3e–g). However, the PCB concentrations in the fish from the [PCB + FBP] was still 1.44–fold higher than that in the fish from the [PCB] group at the end of depuration ($p > 0.05$) (Fig. 3g). The accumulated PCB concentrations in the zebrafish from the [PCB + MP] group were comparable to those in the zebrafish from the [PCB] group during the entire depuration period.

Next, the PCBs fugacities (f; Pa) of the MPs, FBPs and zebrafish muscle were calculated; the f values were determined according to previous studies (Debruyn and Gobas, 2007; Golding et al., 2007; Golding et al., 2008). When the PCBs loaded on MPs and FBPs at the equi-fugacity concentration, the f values ranged from 0.16–6.36E-06 Pa for MPs, 0.59–11.5E-05 Pa for FBPs, and 0.22–9.17E-05 Pa for zebrafish muscle (**Table S2**). A $f_{matrix1}/f_{matrix2}$ value of >1 indicates that the direction of the migration of organic pollutants was from matrix 1 to matrix 2. The average value of f_{FBP}/f_{muscle} was >1, suggesting that the transfer of PCBs from the FBPs to fish muscle

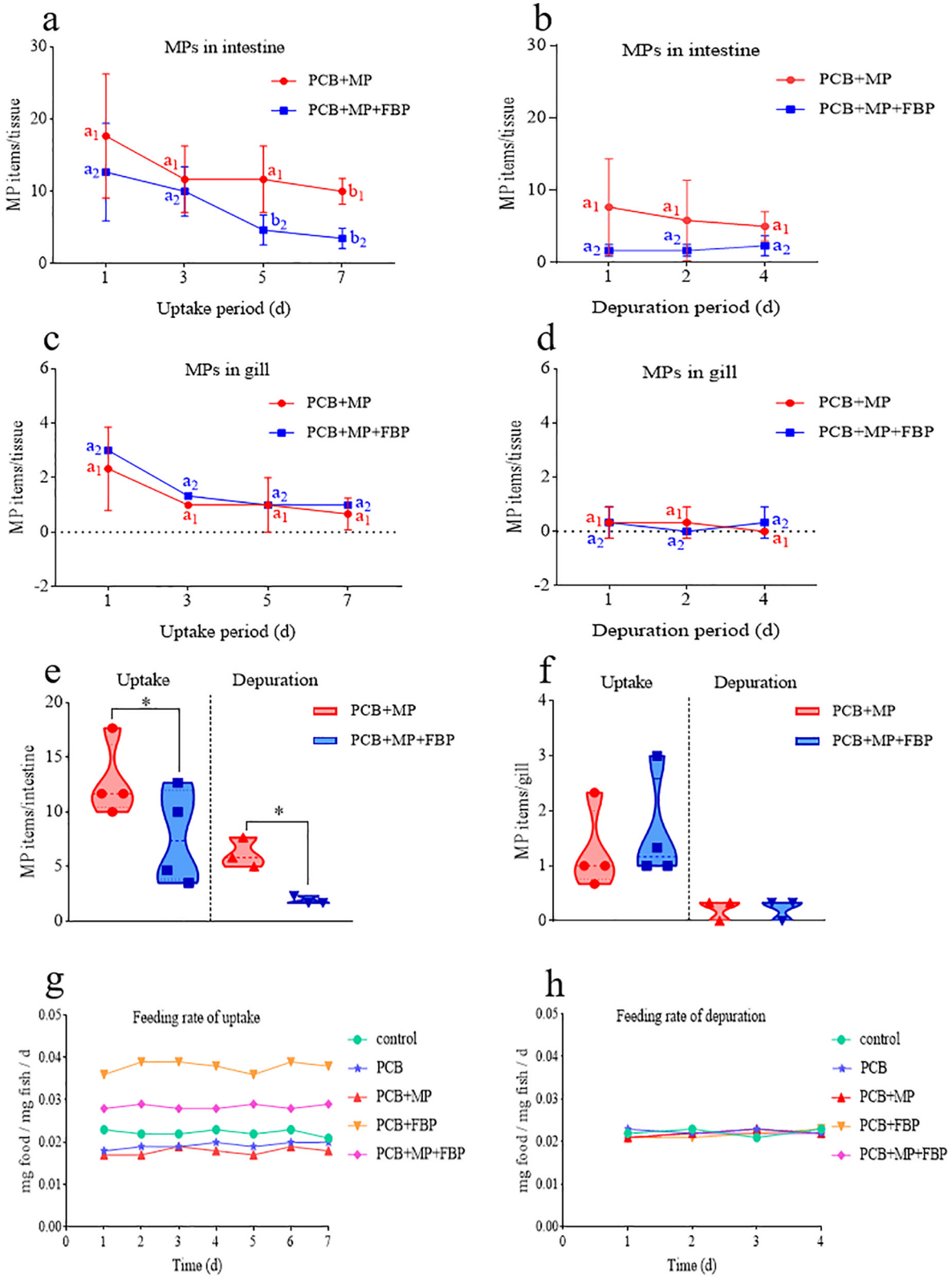

Fig. 2. Concentrations of MPs in the intestines and gills of zebrafish in [PCB + MP] and [PCB + MP + FBP] groups. (a) MPs concentrations in intestine during uptake, and (b) during depuration; (c) MPs concentrations in gill during uptake, and (d) during depuration. a_1/b_1: Represents the significant difference between the [PCB + MP] group at different sampling time. a_2/b_2: Represents the significant difference between the [PCB + MP + FBP] group at different sampling time ($p < 0.05$). Differences in the content of MPs (e) in intestines and (f) gills of [PCB + MP] and [PCB + MP + FBP] groups during uptake and depuration periods. *: Represents significant differences between the [PCB + MP] and the [PCB + MP + FBP] groups at the same sampling time ($p < 0.05$); (g) Food feeding rate of zebrafish during uptake period, and (h) depuration period. Note: [Control]: AFW solution; [PCB]: 1 μg/L PCBs in the solution; [PCB + MP]: both equi-fugacity and above-fugacity PCBs on MPs, and 1 μg/L PCBs in the solution; [PCB + FBP]: both equi-fugacity and above-fugacity PCBs on FBPs, and 1 μg/L PCBs in the solution; [PCB + MP + FBP]: equi-fugacity PCBs on FBPs and above-fugacity PCBs on MPs, and 1 μg/L PCBs in the solution.

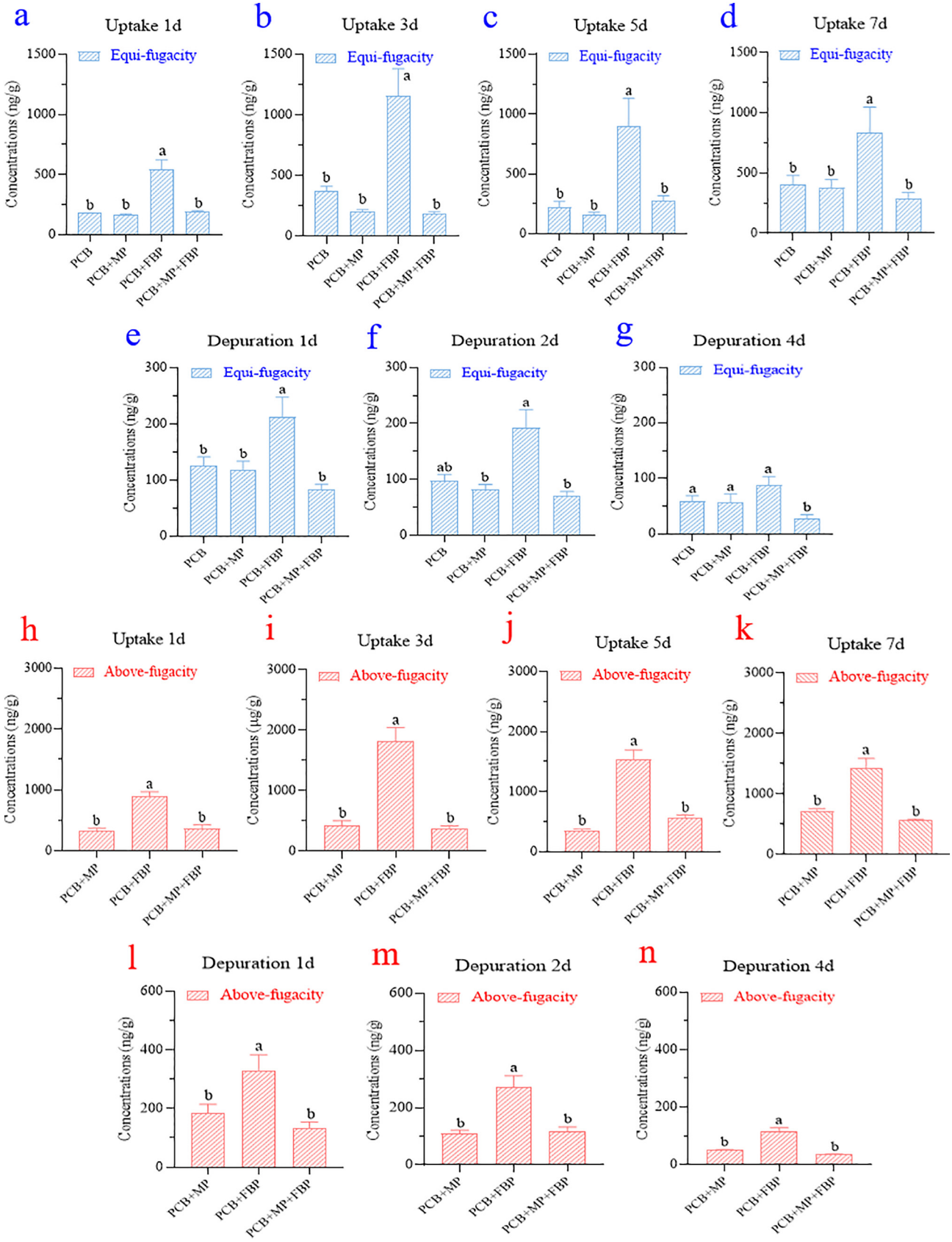

Fig. 3. Total accumulated PCBs in zebrafish under both the equi-fugacity and above-fugacity scenarios during the uptake and depuration periods. (a-g) PCB concentrations in each group under the equi-fugacity scenario; (i-n) PCB concentrations in each group under the above-fugacity scenario. Different letters in each figure mean that there were significant differences among groups ($p < 0.05$). [PCB]: 1 μg/L PCBs in the solution; [PCB + MP]: both equi-fugacity and above-fugacity PCBs on MPs, and 1 μg/L PCBs in the solution; [PCB + FBP]: both equi-fugacity and above-fugacity PCBs on FBPs, and 1 μg/L PCBs in the solution; [PCB + MP + FBP]: equi-fugacity PCBs on FBPs and above-fugacity PCBs on MPs, and 1 μg/L PCBs in the solution.

would occur (Golding et al., 2008). The above results suggest that the presence of MPs did not significantly alter the PCBs accumulation in the presence of the equi-fugacity PCB concentrations, while the FBPs enhanced the PCBs bioaccumulation.

3.4. Above-fugacity scenario: MPs did not affect, but FBPs enhanced PCBs bioaccumulation

After seven days of uptake, there were no significant differences between the PCB concentrations in the zebrafish from the [PCB], [PCB + MP], and [PCB + MP + FBP] groups, but significant differences were found between the PCB concentrations in the zebrafish from the [PCB + FBP] group and those in the fish from the three aforementioned groups (Fig. 3). When the MPs or FBPs were added in the presence of the above-fugacity concentration of PCB, no significant difference was found between the PCB concentrations in the zebrafish from the [PCB + MP] and [PCB + MP + FBP] groups, but significant differences were found between the PCB concentrations in the zebrafish from the [PCB + FBP] group and those in these two groups.

PCBs loaded onto different environmental matrices (i.e., MPs and FBPs) showed different levels of bioaccumulation in zebrafish. It is noteworthy that significantly higher PCB concentrations were detected in fish from the [PCB + FBP] group than in those from the [PCB + MP] group ($p < 0.01$), both at most of the scenarios involving the equi-fugacity or above-fugacity PCB concentrations (Fig. 4). This is because zebrafish cannot digest MPs, while food particles can be assimilated easily. Thus, the PCBs carried by FBPs would be more bioavailable to the zebrafish during food digestion (Fig. 4h). Therefore, the PCBs associated with MPs had a much lower bioavailability than those associated with FBPs. When the PCBs were loaded onto the MPs and FBPs at the above-fugacity PCB concentration, the f values ranged from 0.39–5.96E-05 Pa for MPs, 0.10–1.23E-03 Pa for FBPs, and 0.30–16.3E-05 Pa for zebrafish muscle (**Table S3**). Although the f_{PE}/f_{muscle} values of CB-118 and CB-153 were always greater than 1, except after uptake 5 d in the [PCB + MP + FBP] group and after uptake for 7 d. The average f_{PE}/f_{muscle} value was <1, indicating that the MPs loaded with PCBs at the above-fugacity concentration contributed to PCBs bioaccumulation in zebrafish to a lesser degree. On the other hand, the average f_{FBP}/f_{muscle} value was >1, suggesting that at the equi-fugacity PCB concentration, the FBPs were more likely to enhance PCBs bioaccumulation in the zebrafish than the MPs.

3.5. Equi-fugacity scenario: MPs suppressed the effects of FBPs

At the equi-fugacity PCB concentration, the highest PCBs accumulation was observed in the zebrafish from the [PCB + FBP] group (1153.5 ± 225.8 ng/g) on the 3rd day of uptake; the presence of MP significantly decreased the PCBs accumulation by 6.25-fold in the [PCB + MP + FBP] group (184.6 ± 17.2 ng/g; Fig. 3b). A similar phenomenon was observed throughout the uptake period; the concentration of PCBs in the [PCB + MP + FBP] group was significantly lower, i.e., 2.86–4.03-fold lower, than that in the [PCB + FBP] group (Fig. 3a–d). This phenomenon probably occurs because the ingested MPs cannot be assimilated by zebrafish and will finally be excreted out (Ory et al., 2018; Xu et al., 2021). During the excretion, MPs could eliminate a part of the PCBs

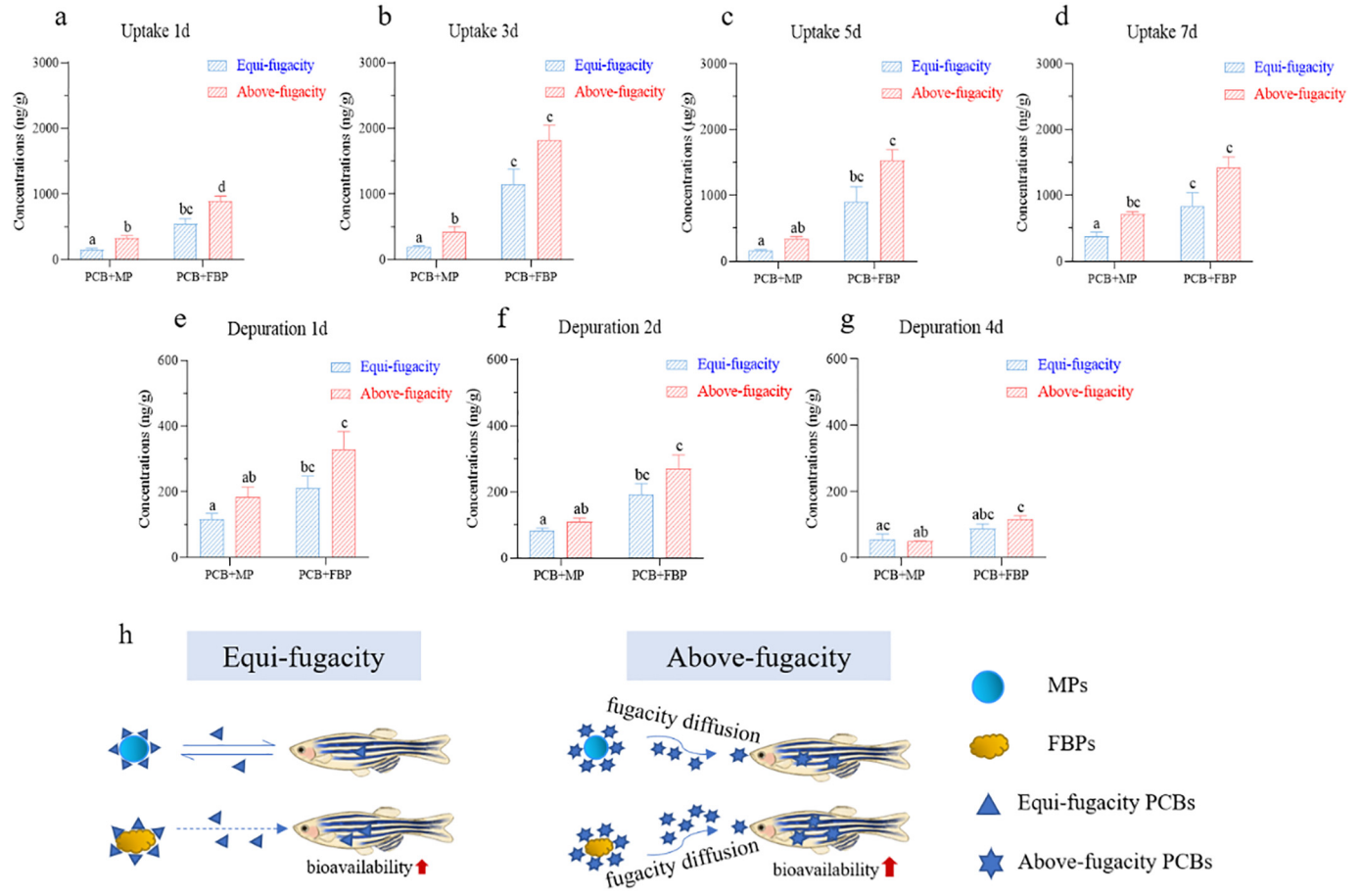

Fig. 4. Comparison of microplastics and food-borne particles on the PCBs bioaccumulation in zebrafish under both equi-fugacity and above-fugacity scenarios. (a-d) PCB concentrations in zebrafish during the uptake period; (e-g) PCB concentrations in zebrafish during the depuration period. Different letters in each figure mean that there were significant differences among groups ($p < 0.05$). (h) Mechanism interpretation sketch for the effects of MPs and FBPs on PCBs bioaccumulation in both equi-fugacity and above-fugacity scenarios. [PCB + MP]: both equi-fugacity and above-fugacity PCBs on MPs, and 1 μg/L PCBs in the solution; [PCB + FBP]: both equi-fugacity and above-fugacity PCBs on FBPs, and 1 μg/L PCBs in the solution.

associated with FBPs in the meantime, and thus, the PCB levels in the bodies of the zebrafish may have reduced. Previous studies have also verified that the presence of MPs can decrease the bioaccumulation of co-existing PCBs on food in goldfish (*Carassius auratus*) (Grigorakis and Drouillard, 2018) and co-existing PCBs as well as brominated flame retardants on food in European seabass (*Dicentrarchus labrax*) (Granby et al., 2018).

3.6. Above-fugacity scenario: FBPs did not influence the effects of MPs

During the entire experimental period, sufficient food (5% of wet weight) was provided to the zebrafish to explore the bioaccumulation of PCBs in simulated ideal environmental conditions (Chae and An, 2020; Korez et al., 2019). When the associated PCBs on MPs were at the above-fugacity states, the presence of FBPs (equi-fugacity) did not alter the PCBs bioaccumulation notably, as no obvious differences were observed between the [PCB + MP] and the [PCB + MP + FBP] groups (Fig. 3h–n). For instance, the highest amount of PCBs accumulated in the zebrafish was detected to be 712.9 ± 41.9 ng/g in case of the [PCB + MP] group on the 7th day of uptake, which was similar to that in the [PCB + MP + FBP] group (567.5 ± 15.2 ng/g). The presence or absence of FBPs (equi-fugacity) had no notable effects on the bioaccumulation of PCBs in zebrafish. Though the competitive sorption of PCBs by FBPs (equi-fugacity) from MPs (above-fugacity) may exist, the presence of FBPs in the aqueous exposure solution only lasted for 2 h per day in this study; this, therefore, had a limited influence on the effects of MPs on PCBs accumulation in zebrafish (Razanajatovo et al., 2018).

3.7. Analysis of the contents of PCB congeners

We focused on the differences in PCB concentrations between the different treatments after 7 days of uptake and 4 days of depuration. The uptake of individual congeners was expected to be proportional to the PCB K_{ow} during the uptake period, but inversely proportional to the PCB K_{ow} during the depuration period (Sun et al., 2016); similar results were found in our study. During the uptake period, only the concentration of CB-138 in each group was significantly higher than that of CB-44 and 101; in the above-fugacity scenario, although the CB-153 level was higher than the CB-52 and -118 levels, there was no significant difference (**Fig. S5**). During depuration, a significantly higher concentration of the remaining CB-138 was again found under the equi-fugacity scenario, except in case of the [PCB] group; similarly, the concentrations of the remaining CB-153 were significantly higher than those of the remaining CB-52 and -118 in the zebrafish under the above-fugacity scenario, except in case of the [PCB + FBP] group.

3.8. Environmental significance

Several studies have detected the presence of organic contaminants accumulated in organisms through their contact with MPs (Fu et al., 2021; Hal et al., 2020; Rainieri et al., 2018), whereas others show no uptake or accumulation in biological tissues (Costanza et al., 2018; Magara et al., 2018) and that MPs may even reduce the concentration of organic pollutants (Kleinteich et al., 2018; Rehse et al., 2018). These different conclusions are mainly due to varying exposure scenarios with different assumptions (Koelmans et al., 2016). In this study, we distinguished, in detail, the scenarios involving the equi-fugacity and above-fugacity concentrations of PCBs to illustrate the migration of the organic pollutants in the organisms under different conditions. Our study also provides a theoretical basis for the bioaccumulation of flame retardants with physical and chemical properties similar to those of PCBs.

When MPs are exposed to an aqueous solution where the distribution of pollutants has reached an equilibrium, their carrier function does not emerge. Moreover, MPs can even reduce the accumulation of organic pollutants loaded onto FBPs in aquatic organisms, which is beneficial for the environment. Nonetheless, the spread of MPs cannot be ignored since the volumes of these plastic particles are growing and ecotoxicological effects may become more prominent with the increase in their concentration (Jiang et al., 2020; Xu et al., 2020).

However, PCBs associated with food matrices are subject to biomagnification, in addition to equilibrium partitioning between the organisms and water. On the other hand, PCBs associated with MPs are subject to equilibrium partitioning in the GI-tract because MPs cannot be digested. When pollutants associated with FBPs are present in an equi-fugacity scenario, the bioavailability of co-existing pollutants would still be altered. Therefore, the combined effects of organic pollutants and FBPs should be considered during the risk assessment of aquatic environmental pollutants.

This study has several limitations. First, the above-fugacity scenario was mainly studied with regard to the primary MPs carrying additives discharged into the environment (Liu et al., 2020). However, the additives on the secondary MPs are very likely to have reached equilibrium with the surrounding environment after long-term degradation and fragmentation. Second, as the actual sorption equilibrium takes months to years to occur (Cormier et al., 2021; Godoy et al., 2020), we used pseudo-equilibrium to mimic the approximate sorption amounts of PCBs on the MPs and/or FBPs, which may under estimate the amounts of PCBs associated with the MPs and FBPs. Further, the uptake and depuration periods were not long enough to estimate kinetics-based bioaccumulation factors (Sun et al., 2016). Third, when the concentration of PCBs loaded onto MPs is lower than the equi-fugacity PCB concentration, the MPs will play a cleaning role for organisms (Devriese et al., 2017; Diepens and Koelmans, 2018); this was not taken into consideration in the present study. Finally, in certain cases, MPs can even alter the metabolism of pollutants, i.e., polyaromatic hydrocarbons, in organisms (Diepens and Koelmans, 2018; Sheng et al., 2021); this was also not considered in the experimental design.

4. Conclusions

Our results showed that MPs can only enhance the PCBs bioaccumulation in fish in the presence of above-fugacity PCB concentrations, but FBPs can enhance PCBs bioaccumulation in the presence of both equi- and above-fugacity PCB concentrations. Moreover, the MPs were conducive to the elimination of PCBs during depuration but the FBPs were not. Finally, during the co-exposure to MPs and FBPs, the MPs accelerate the depuration of PCBs loaded onto the FBPs, while the FBPs do not affect the bioaccumulation of PCBs loaded onto the MPs. Thus, the present study provides new insights into the effects of MPs and FBPs on the bioaccumulation of co-existing pollutants present at equi-fugacity or above-fugacity concentrations in aquatic environments.

Declaration of Competing Interest

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

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2021.152548.

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