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Estimating microplastic-bound intake of hydrophobic organic chemicals by fish using measured desorption rates to artificial gut fluid

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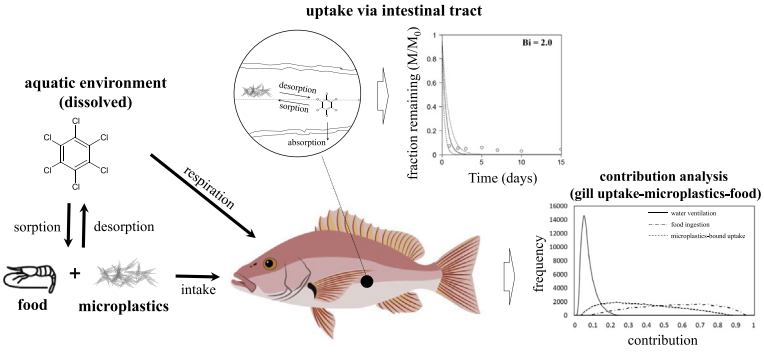
HIGHLIGHTS

- The contribution from ingested MPs to overall uptake of HOCs by fish was evaluated.
- $K_{MP/SIF}$ and $K_{om/SIF}$ were determined for HOCs using a tree-phase partitioning method.
- HOCs leaching from MPs in artificial gut fluid was compared with model predictions.
- The contribution of plastic-bound intake was assessed using a fish bioaccumulation model.
- The results suggest that the role of ingested MPs may play as "dilution" for HOCs.

GRAPHICAL ABSTRACT



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ABSTRACT

One of the most important concerns about marine microplastics is their role in delivery of chemical contaminants to biota. The contribution of microplastic ingestion to the overall uptake of five hydrophobic organic chemicals (HOCs) [α-, β-, and γ-hexachlorocyclohexanes (HCHs), pentachlorobenzene (PeCB), and hexachlorobenzene (HeCB)] by fish is evaluated in this study. Partition coefficients of all five HOCs between surfactant micelles and simulated intestinal fluid (SIF), as well as between protein and SIF, were experimentally determined. Desorption of model HOCs from a polyethylene film into an artificial gut solution was measured to estimate the fraction of HOCs that can be absorbed from microplastics during their gut retention time. Monte-Carlo simulation (n = 100,000) showed that the uptake via microplastic ingestion will be negligible for HCHs as compared to uptake via other exposure routes, water ventilation and food ingestion. On the other hand, microplastic ingestion might increase the total uptake rate of PeCB and HeCB due to their accelerated desorption from microplastics into the artificial gut solution under the model scenario, assuming an extremely high intake of microplastics. However, the steady-state bioaccumulation factor was predicted to decrease with increasing ingestion of microplastics, showing a dilution effect by microplastic ingestion. Results indicate that HOCs that are close to be at phase equilibrium between microplastics and environmental media are not likely to be further accumulated via ingestion of microplastics; this is true even for cases, where ingestion of microplastics contributes significantly to the total uptake of HOCs. Therefore, future studies need to focus on hydrophobic plastic additives that may exist in microplastics at a concentration higher than their equilibrium concentration with water.

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1. Introduction

Accumulation of microplastics in marine environments is one of the most serious environmental problems in this century. Many researchers have studied the potential adverse and long-term environmental effects of microplastics (Andrady, 2011; Engler, 2012; Faggio et al., 2018; Moore, 2008). One of the central hypotheses about the adverse effects of microplastics is their role in the delivery of harmful chemicals to biota (Avio et al., 2015; Browne et al., 2013; Koelmans et al., 2013; Rochman et al., 2013a; Teuten et al., 2009). Marine microplastics may contain high concentrations of hydrophobic organic chemicals (HOCs) because they are often intentionally added to enhance the properties of plastic products (Kwon et al., 2017); alternatively, microplastics may gain HOCs from polluted environments due to their high sorption capacity for HOCs (Bakir et al., 2012; Lee et al., 2014; Liu et al., 2016; Rochman et al., 2013b; Velzeboer et al., 2014). Since the ingestion of plastic particles by marine organisms has been well-documented for decades (e.g., Choy and Drazen, 2013; Davison and Asch, 2011; Foekema et al., 2013; Jang et al., 2016; Kühn et al., 2015; Laist, 1997; Provencher et al., 2010; Remy et al., 2015; Rummel et al., 2016, Tanaka and Takada, 2016), it has created an important question of whether microplastics significantly contribute to the uptake and accumulation of HOCs through the marine food-web.

The hypothesis that microplastics play an important role as a transport vector of HOCs has been tested in two-ways: laboratory- and field-scale experimental studies and mathematical modeling studies. A few laboratory experiments indicated that plastic-bound HOCs are either transferred to test organisms (e.g., Chua et al., 2014) or substantially leached under simulated physiological conditions (Bakir et al., 2014). Chua et al. (2014) showed that polybrominated diphenyl ethers from microplastic debris can significantly assimilate in the tissues of the marine amphipod, *Allorchestes compressa*. Bakir et al. (2014) showed that the desorption rates of model HOCs (phenanthrene, di-2-ethylhexyl phthalate, perfluorooctanoic acid, and dichloro-diphenyl-trichloroethane) were faster under physiological conditions than in water without organic matter. However, these experimental studies did not reflect the actual marine environmental conditions because the number and mass concentration of plastic particles used in the experiments and the concentration of HOCs in the plastic phase were much higher than the concentrations reported for marine environments. In addition, the roles of other exposure routes (i.e., respiratory uptake and assimilation from prey) and loss processes (e.g., metabolic transformation and fecal egestion) were not quantified or compared to the contribution of microplastic-bound intake. In contrast to experimental studies, mathematical modeling studies (e.g., Gouin et al., 2011; Herzke et al., 2016; Koelmans et al., 2013) have suggested a limited role of microplastics as transport vectors for HOCs. When the distribution of HOCs between environmental media and microplastics is assumed to be in equilibrium, the fugacity of HOCs in microplastic phase is expected to be close to or lower than that in aquatic organisms at higher trophic levels. The ingestion of relatively "cleaner" microplastics may decrease the overall bioaccumulation of HOCs in piscivorous fish (Gouin et al., 2011) and in lugworm, *Arenicola marina* (Koelmans et al., 2013). Even for avians, the contribution of leachable persistent organic pollutants (POPs) from ingested microplastics to the overall bioaccumulation in northern fulmar (*Fulmarus glacialis*) was reported to be limited; ingested microplastics may act as a "passive sampler" for selected POPs in the body (Herzke et al., 2016).

The discrepancy between laboratory experimental and modeling studies is attributable to the different initial equilibrium conditions and kinetic rates. For non-additives, such as polychlorinated biphenyls, the acquired concentration in microplastic-phase from the environmental media is close to or lower than the concentration at phase equilibrium. However, chemical concentration of hydrophobic additives in microplastics could be higher than the equilibrium concentration by a few orders of magnitude. In addition, the desorption rates in the digestive tracts, where organic matter facilitates transfer of HOCs, are also much enhanced (Bakir et al., 2014). Thus, it needs to be further studied whether microplastics contribute to the bioaccumulation of plastic additives or not (Kwon et al., 2017).

The release of HOCs from microplastics has been studied and models describing the phenomenon have been developed for the cases of desorption into seawater (Endo et al., 2013; Lee et al., 2018a, 2018b) and into wastewater containing organic matter (Seidensticker et al., 2017). Studies are required on the equilibrium distribution of HOCs in digestive fluid that contains non-lipid organic matter, such as proteins and surfactants, and on the quantitative evaluation of desorption rates of HOCs to provide refined parameters for estimation of the transfer of HOCs from microplastics to organisms.

In this study, we evaluated the contribution of ingested microplastics to the overall transfer of HOCs into fish with experimentally determined parameters. Polyethylene (PE) was chosen as a model plastic material because it is most frequently used and identified as a type of microplastic (Hidalgo-Ruz et al., 2012). Five model HOCs, including three isomers of hexachlorocyclohexanes (HCHs) and two chlorinated benzenes (CBs), were chosen as model HOCs because their physico-chemical properties required for the estimation of desorption have been intensively studied in our earlier works (Lee et al., 2014, 2018a). The equilibrium partition coefficients between microplastics and simulated intestinal fluid (SIF) and also between organic matter components (surfactant and digestive enzymes) and SIF were experimentally determined. The leaching of HOCs from plastic phase into the artificial gut fluid, SIF dissolving organic matter components, were measured in a batch test; the results were compared with predictions made using a convection-diffusion model for desorption (Lee et al., 2018a). Finally, the contribution of plastic-bound intake was assessed using a one-compartment fish bioaccumulation model with an uncertainty analysis using Monte-Carlo simulation.

2. Material and methods

2.1. Materials and chemicals

High purity α- (99.8%), β- (99.5%), and γ-HCH (99.8%), pentachlorobenzene (PeCB) (98%), and hexachlorobenzene (HeCB) (99%) were purchased from Sigma-Aldrich (St. Louis, MO, USA), Supelco (Bellefonte, PA, USA), or Fluka (Buch, Switzerland). All partitioning and leaching experiments were conducted using two mixtures of chemicals with similar hydrophobicity: HCHs and CBs mixtures. Simulated intestinal fluid (SIF) was prepared by dissolving 500 mM potassium dihydrogen phosphate (KH_2PO_4) and 224 mM sodium hydroxide (NaOH) in deionized water (pH 6.8).

Medical-grade polydimethylsiloxane (PDMS) sheets (thickness = 1 mm, density = 1170 kg m^{-3}) were purchased from Specialty Silicone Products, Inc. (Ballston Spa, NY, USA). The PDMS sheet was cut into disks of 6-mm diameter for partition coefficient experiments and into rectangular sheets (10 mm × 50 mm) for leaching experiments. The custom-cut PDMS disks and sheets were cleaned using *n*-hexane followed by methanol for 2 h each and stored in methanol until use.

Polyethylene (PE) films with a thickness of 75 μm and density of 940 kg m^{-3} were purchased from Goodfellow Cambridge Ltd. (Huntingdon, UK). The PE film was cut into 10 mm × 10 mm square sheets, which were cleaned using *n*-hexane followed by methanol for 2 h each and stored in methanol until use.

2.2. Determination of partition coefficients between PDMS and simulated intestinal fluid (SIF) ($K_{PDMS/SIF}$) and between polyethylene microplastics and SIF ($K_{MP/SIF}$)

Due to the low solubilities of model HOCs in the aqueous solutions, partition coefficients between microplastics and SIF ($K_{MP/SIF}$) were determined using a three-phase partitioning method (Kim et al., 2014;

Lee et al., 2012, 2014; ter Laak et al., 2005). Partition coefficients between PDMS and SIF ($K_{PDMS/SIF}$) were measured independently and were then used to obtain $K_{MP/SIF}$ with the following equation:

$$K_{MP/SIF} = \frac{K_{MPsm}}{K_{PDMSsm}} K_{PDMS/SIF} \quad (1)$$

where K_{MPsm} and K_{PDMSsm} are partition coefficients between polyethylene microplastics and the solvent mixture (methanol:water = 8:2, v/v) and between PDMS and the solvent mixture obtained in our earlier study, respectively (Lee et al., 2014).

For less hydrophobic HCHs, the $K_{PDMS/SIF}$ values were determined using the batch equilibrium method, whereas those for CBs with higher hydrophobicity were determined using the aqueous boundary permeation (ABL) method (Kwon et al., 2007). Solutions of HCHs in SIF were prepared at 4 different initial concentrations, where the maximum concentrations of HCHs were 500, 100, and 600 μg L^{-1} for α-, β-, and γ-HCH, respectively, below their water solubilities (Mackay et al., 2006). A PDMS disk was placed in a vial containing 4 mL of SIF and vials were shaken for 48 h using a shaking incubator at 150 rpm and 25 °C. After shaking, both the SIF and the PDMS disk were extracted using *n*-hexane to obtain the concentration of HOCs. Two milliliters of *n*-hexane were used to extract HOCs from 2 mL of SIF solution or a PDMS disk at 25 °C and 150 rpm. All *n*-hexane extracts were subjected to a gas chromatography analysis.

The values of the $K_{PDMS/SIF}$ for CBs were determined using the ABL permeation method. Custom-cut PDMS disks were preloaded for 24 h at 25 °C and 150 rpm using a loading solution (methanol:water = 6:4, *v*/v) containing a mixture of CBs. After loading, the initial concentrations in the preloaded PDMS were 608 μmol L_{PDMS}^{-1} for PeCB and 547 μmol L_{PDMS}^{-1} for HeCB. The preloaded donor PDMS and a clean acceptor PDMS were separated by SIF. A stainless steel stirring disk with a diameter of 5.08 mm and thickness of 0.653 mm (V&P Scientific, Inc.) was placed in the SIF solution (approximately 300 μL) and stirred at 300 rpm. After predetermined time intervals, both PDMS disks were taken, cleaned with a lint-free tissue, and extracted with 1 mL of *n*-hexane for chemical analysis. The mass transfer rate constant (k) was calculated using Eq. (2).

$$\ln\left(1 - \frac{2C_{acceptorPDMS}}{C_0}\right) = -kt \quad (2)$$

where $C_{acceptorPDMS}$ is the concentration of CBs in the acceptor PDMS disk [mg L^{-1}] and C_0 is the initial concentration of CBs in the donor PDMS disk [mg L^{-1}]. The $K_{PDMS/SIF}$ values were then calculated assuming that the overall mass transfer is dominated by the aqueous boundary layer diffusion (Kwon et al., 2007).

$$K_{PDMS/SIF} = \frac{D_{SIF}}{\delta_{SIF}} \frac{A_{PDMS}}{V_{PDMS} k} \quad (3)$$

where D_{SIF} is the molecular diffusion coefficient of CBs in SIF solution [m^2 s^{-1}], δ_{SIF} is the thickness of ABL that has previously been estimated to be 12.5×10^{-6} m in the experimental setup (Kwon et al., 2007), A_{PDMS} is the contact area of PDMS with SIF [m^2], and V_{PDMS} is the volume of PDMS [m^3]. The D_{SIF} was estimated using the Hayduk-Laudie correlation (Hayduk and Laudie, 1974). The molar liquid volume of chemicals was estimated using the LeBas method (Reid et al., 1977).

$$D_{SIF} = \frac{13.26 \times 10^{-9}}{\eta^{1.4} MLV^{0.589}} \quad (4)$$

where η is the viscosity of the SIF solution and MLV is the LeBas molar liquid volume of CBs [cm^3 mol^{-1}]. The viscosity of 3.5% (w/w) seawater (0.97 mPs at 25 °C) was used to replace η.

2.3. Determination of partition coefficients between organic matter components and SIF

The partition coefficients between organic matter components (surfactants and proteins) and water were also determined. To function as organic matter surrogates in the fish gut conditions, 10,000 mg L^{-1} bovine serum albumin (BSA) and 8000 mg L^{-1} sodium taurocholate (NaT) were chosen (critical micelle concentration of 1500–5700 mg L^{-1} (Volparil and Mayer, 2004)). The partition coefficients between BSA and SIF ($K_{BSA/SIF}$), between sodium taurocholate and SIF ($K_{NaT/SIF}$), and between both organic matter surrogates and SIF ($K_{BSA+NaT/SIF}$) were determined by the three-phase partitioning method with PDMS as the third phase. Test chemicals were loaded into a PDMS disk using methanol:water (6:4, v/v) loading solution. A loaded PDMS disk was placed in a vial containing 6 different volumes of SIF containing organic matter; the vials were shaken for 7 days in a shaking incubator at 150 rpm and 25 °C. Once apparent equilibrium had been attained in 7 days, the PDMS disk was removed from the solution, rinsed twice with 2 mL of deionized water, and extracted using 1 mL of *n*-hexane. The *n*-hexane extracts were subjected to gas chromatography – electron capture detector to obtain concentrations of test chemicals in PDMS. The experimentally-determined relative concentration of test chemicals in PDMS ($C_{PDMS}/C_{PDMS,0}$) is expressed as follows (Kim et al., 2010;Kwon et al., 2009):

$$\frac{C_{PDMS}}{C_{PDMS,0}} = \frac{1}{1 + \frac{(V_{SIFom}/V_{PDMS})}{K_{PDMS/SIFom}}} \quad (5)$$

where C_{PDMS} is the concentration in PDMS at phase equilibrium [mg L^{-1}], $C_{PDMS,0}$ is the initial concentration in PDMS [mg L^{-1}], V_{SIFom} and V_{PDMS} are the volume of the SIF containing organic matter and the volume of PDMS [L], respectively, and $K_{PDMS/SIFom}$ is the partition coefficient between PDMS and the SIF with organic matter. The $K_{PDMS/SIFom}$ values were obtained by a nonlinear regression using Eq. (5) using $C_{PDMS}/C_{PDMS,0}$ vs. V_{SIFom}/V_{PDMS}. The value of $K_{om/SIF}$ was then calculated using $K_{PDMS/SIF}$ by:

$$K_{om/SIF} = \frac{\left(\frac{K_{PDMS/SIF}}{K_{PDMS/SIFom}}\right) - 1}{m_{om}/10^6} \quad (6)$$

where m_{om} is the concentration of organic matter components in the SIF in mg L^{-1}.

2.4. Leaching of HOCs from plastic phase to the artificial gut fluid

Leaching of the model HOCs from PE sheet into the artificial gut fluid (1000 mg L^{-1} BSA and 800 mg L^{-1} NaT in SIF) was evaluated and modeled with a one-dimensional convection-diffusion model (Lee et al., 2018a) because a one-dimensional film model was found to provide a good approximation for the desorption from plastic fragments with arbitrary shapes (Lee et al., 2018b). Detailed explanation of the model is provided in the Supplementary material. The model HOCs were loaded into the PE sheet using *n*-hexane for 24 h at 25 °C and 150 rpm. The PE sheet was then separated and washed with 4 mL of methanol:water (8:2, v/v) solution to remove residual *n*-hexane from the surface. After loading, the initial concentrations in the preloaded PE sheet ranged from 15 mmol m^{-3} (for α-HCH) to 600 mmol m^{-3} (for HeCB).

The experimental setup used for a batch leaching test was similar to that used in our earlier study (Lee et al., 2018a). In short, the preloaded PE sheet was placed in a vial containing 20 mL of artificial gut solution, which provides an infinite sink condition due to the presence of sufficiently large volume of PDMS. The vials were shaken up to 360 h, considering the typical gut retention times for fish species: 10–158 h for

carnivorous and 3–10 h for herbivorous fish (Clements, 1997), to measure the concentration of remaining HOCs in the PE sheet. After the designated time interval, the PE sheet was removed from the solution and extracted using 1 mL of *n*-hexane. The SIF with its organic matter components and PDMS were also extracted using *n*-hexane to obtain the remaining mass of HOCs and to ensure the conservation of mass. All *n*-hexane extracts were agitated in a shaking incubator for 24 h at 25 °C and 150 rpm and then subjected to gas chromatography analysis.

2.5. Bioaccumulation model for fish to account for ingestion of microplastics

To simulate the transfer of desorbed HOCs from ingested microplastics by fish, a one-compartment bioaccumulation model (Hauck et al., 2011; Koelmans et al., 2013; Thomann et al., 1992) was used.

$$\frac{dC_{fish}}{dt} = k_1 C_{sw}^{free} + IR_{food}\alpha_{food}C_{food} + IR_{MP}\alpha_{MP}C_{MP} - k_{loss}C_{fish} \quad (7)$$

where C_{fish} is the HOC concentration in fish [μg kg_{fish}^{-1}], C_{sw}^{free} is the free concentration of HOC in seawater [μg L^{-1}], k_1 is the gill uptake rate constant [L kg_{fish}^{-1} d^{-1}], IR_{food} and IR_{MP} are intake rates of food and microplastic particles containing HOC [g kg_{fish}^{-1}], α_{food} and α_{MP} are absorption fraction of HOC [−], C_{food} and C_{MP} are the concentration of HOC in food and microplastics [μg g^{-1}], and k_{loss} is the overall elimination rate constant [d^{-1}], which is estimated by the linear sum of dilution rate constant by growth (k_g), metabolic degradation rate constant (k_m), fecal excretion rate constant (k_e), excretion of microplastics (k_{MP}), and gill elimination rate constant (k_2).

$$k_{loss} = k_g + k_m + k_e + k_{MP} + k_2 \quad (8)$$

where k_g and k_m were assumed to be zero. The values of k_1, k_2, and k_e were obtained using empirical equations. These values are related to body weight (W), lipid content (f_{lip}), temperature (T), and K_{ow} (used as a surrogate for biopartitioning coefficient) (Arnot and Gobas, 2003).

$$k_1 = \frac{1}{\left(\left(0.01 + \frac{1}{K_{ow}}\right) \times W^{0.4}\right)} \quad (9)$$

$$k_2 = \frac{k_1}{f_{lip}K_{ow}} \quad (10)$$

$$k_e = 0.125 \times \left(\frac{0.02 \times W^{-0.15} \times e^{0.06T}}{5.1 \times 10^{-8}K_{ow} + 2}\right) \quad (11)$$

The loss rate constant via excretion of microplastics (k_{MP}) was estimated by assuming that an instantaneous equilibrium was reached between the fish and the microplastics as estimated by the following equation:

$$k_{MP} = IR_{MP}\frac{K_{MPw}}{f_{lip}K_{ow}} \quad (12)$$

Assuming that seawater, food, and microplastics are all at the partitioning equilibrium, the fractions of water ventilation (f_w), food ingestion (f_{food}), and microplastic-bound intake (f_{MP}) for the overall intake were estimated as:

$$f_w = \frac{k_1 C_{sw}^{free}}{k_1 C_{sw}^{free} + IR_{food}\alpha_{food}C_{food} + IR_{MP}\alpha_{MP}C_{MP}} = \frac{k_1}{k_1 + IR_{food}\alpha_{food}f_{lip,food}K_{ow} + IR_{MP}\alpha_{MP}K_{MPw}} \quad (13)$$

$$f_{food} = \frac{IR_{food}\alpha_{food}C_{food}}{k_1 C_{sw}^{free} + IR_{food}\alpha_{food}C_{food} + IR_{MP}\alpha_{MP}C_{MP}} = \frac{IR_{food}\alpha_{food}f_{lip,food}K_{ow}}{k_1 + IR_{food}\alpha_{food}f_{lip,food}K_{ow} + IR_{MP}\alpha_{MP}K_{MPw}} \quad (14)$$

$$f_{MP} = \frac{IR_{MP}\alpha_{MP}C_{MP}}{k_1 C_{sw}^{free} + IR_{food}\alpha_{food}C_{food} + IR_{MP}\alpha_{MP}C_{MP}} = \frac{IR_{MP}\alpha_{MP}K_{MPw}}{k_1 + IR_{food}\alpha_{food}f_{lip,food}K_{ow} + IR_{MP}\alpha_{MP}K_{MPw}} \quad (15)$$

where $f_{lip,food}$ is the lipid content of food.

The bioaccumulation factor (BAF) was then calculated as:

$$BAF = \frac{k_1 + IR_{food}\alpha_{food}f_{lip,food}K_{ow} + IR_{MP}\alpha_{MP}K_{MPw}}{k_{loss}} \quad (16)$$

For the Monte-Carlo simulation (n = 100,000), a 500-g adult fish with 5% lipid content living at an average temperature of 15 °C was assumed. Log-uniform distributions were assumed for $f_{lip,food}$ (between 0.01 and 0.10) and IR_{food} (between 0.005 and 0.025 kg_{food} kg_{fish}^{-1} d^{-1}) based on the typical food ingestion rate of fish (Craig and Helfrich, 2002), as well as IR_{MP} (between 0.0001 and 0.001 kg_{MP} kg_{fish}^{-1} d^{-1}). A uniform distribution was assumed for α_{MP} (between 0.7 and 1.0) based on the results of laboratory leaching tests explained later in Section 3.3. All input values are summarized in Table S1 (Supplementary material). The BAF values were also obtained from the Monte-Carlo simulation.

2.6. Instrumental analyses

The concentration of the 5 HOCs was quantified using a gas chromatographic (Hewlett-Packard 5890 Series II) system equipped with an electron capture detector (ECD). Three micrometers of *n*-hexane extract were injected in the splitless mode at 200 °C and separated on an HP-5 column (30 m × 0.25 mm i.d., 0.25 μm film thickness, Agilent J&W Scientific, Folsom, CA). The column oven temperature was held at 120 °C for 3 min, and subsequently, increased to 200 °C at a rate of 5 °C min^{-1} and held for 5 min. The temperature was finally increased to 280 °C at a rate of 45 °C min^{-1} and held for 1 min. The ECD temperature was 320 °C.

3. Results and discussion

3.1. Determination of $K_{PDMS/SIF}$ and $K_{MP/SIF}$

Fig. 1a shows the determination of $K_{PDMS/SIF}$ for HCHs using the batch equilibrium method. Fig. 1b shows the determination of the mass transfer rate (k) for PeCB and HeCB according to Eq. (2) using the ABL permeation method. The experimental data fitted well ($r^2 > 0.94$) with Eq. (2). All measured values of the $K_{PDMS/SIF}$ for the 5 selected HOCs are listed in Table 1. The measured values of log $K_{PDMS/SIF}$ ranged from 1.84 to 5.10 and were not much different from the values measured for K_{PDMSsw} in our previous study (Lee et al., 2014). The effects of dissolved inorganic salt were not expected to be different because the calculated ionic strength of the SIF was 0.948 M, which is not significantly different from 0.75 M, a typical ionic strength of seawater.

Using values of log K_{PEsm} and log K_{PDMSsm} reported in our previous study (Table 1) (Lee et al., 2014), the partition coefficient between polyethylene microplastic and SIF ($K_{MP/SIF}$) was calculated and is listed in Table 1.

3.2. Determination of $K_{BSA/SIF}$, $K_{NaT/SIF}$, and $K_{BSA+NaT/SIF}$

Fig. 2 shows plots of $C_{PDMS}/C_{PDMS,0}$ vs. V_{SIFom}/V_{PDMS} for the determination of $K_{BSA/SIF}$, $K_{NaT/SIF}$, and $K_{BSA+NaT/SIF}$ using Eq. (5) for (a) α-HCH

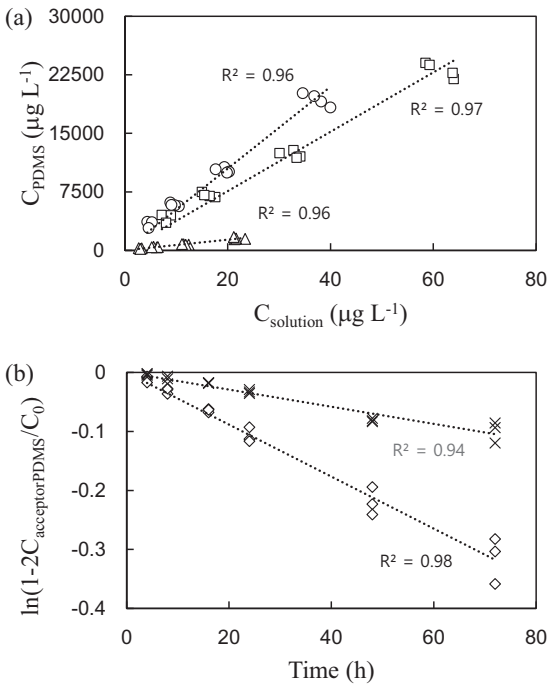

Fig. 1. Determination of partition coefficients between PDMS and SIF using the batch equilibrium method for (a) mixture of HCHs and determination of mass transfer rate constant (k) using ABL permeation method for (b) mixture of CBs: ○, α-HCH; □, β-HCH; △, γ-HCH; ◇, PeCB; ×, HeCB.

and (b) HeCB using 8000 mg L^{-1} NaT, (c) α-HCH and (d) HeCB using 10,000 mg L^{-1} BSA, and (e) α-HCH and (f) HeCB using both 8000 mg L^{-1} NaT and 10,000 mg L^{-1} BSA in SIF. For all other chemicals, plots are shown in Fig. S1–3 (Supplementary material). As shown, the experimental data fitted very well with the equations. Obtained values of $K_{BSA/SIF}$, $K_{NaT/SIF}$, and $K_{BSA+NaT/SIF}$ are listed in Table 2. The log $K_{NaT/SIF}$ values for β- and γ-HCHs were not determined because they were too low to be measured. The log $K_{NaT/SIF}$ values for HCHs were lower than expected as compared to their log K_{ow} values, which may be because of the molar volume of HCHs being much larger as compared to the size of the inner cavity of sodium taurocholate micelles; however, further studies are needed on this front. Values of log $K_{BSA/SIF}$ were lower than log K_{ow} values by 0.74 (HeCB) to 1.65 (α-HCH). These differences agree well with the reported differences between log K_{ow} and log K_{BSAw} (deBruyn and Gobas, 2007). The value of log $K_{BSA/SIF}$ for γ-HCH (1.94) was slightly lower than the logarithm partition coefficient between BSA and water (2.46) reported by Endo and Goss (2011).

Interestingly, experimentally determined values of log $K_{NaT+BSA/SIF}$ were lower than expected from the individually determined log $K_{NaT/SIF}$ and log $K_{BSA/SIF}$ values (Table 2). Except for the values for α-HCH, the differences were between 0.17 and 0.36 log units, indicating that the association of BSA with taurocholate surfactants slightly decreases the sorption capacity of mixed organic matter. Shielding hydrophobic surfaces of BSA by taurocholate might decrease available sorption sites for HOCs and/or the sites for association with micelles, and thus, lowers the sorption capacity in both cases. The hypothesis for the decreased sorption capacity of both the organic matters warrants further investigations.

3.3. Leaching of HOCs from plastic phase into the artificial gut solution

Fig. 3 shows the desorption curves of model HOCs from the PE sheet into the artificial gut solution. Model predictions using the convection-diffusion model (Lee et al., 2018a, 2018b), while assuming the thickness of aqueous boundary layer (δ_w) to be 100 (dashed lines), 300 (solid lines), and 1000 μm (dotted lines), are presented in Fig. 3 with the mass transfer Biot numbers (Bi) estimated at $\delta_w = 300$ μm. As shown, experimental observations agreed well with the model's predictions. The Bi values were >1.0 for all selected HOCs, suggesting that the overall desorption of HOCs from PE sheet into the artificial gut solution was controlled by their internal diffusion in the plastic phase. There are quite significant changes, especially for more hydrophobic PeCB and HeCB, because their desorption into seawater is characterized by Bi numbers <1.0, meaning that the overall desorption is dominated by internal diffusion in the aqueous boundary layer (Lee et al., 2018a). This tendency was also observed by Seidensticker et al. (2017), where desorption of HOCs (e.g., phenanthrene, tonalide, and benzophenone) from PE sphere was accelerated in the presence of dissolved organic matter (DOM) in water. Therefore, the increased DOM concentration in gut solution may enhance the bioavailability of HOCs from ingested microplastics by accelerating transfer kinetics; however, this process also depends on the size of the ingested plastic particles as well.

3.4. Transfer of HOCs to model fish with polyethylene microplastics

As shown in Fig. 3, the desorption of all selected HOCs was controlled by internal diffusion in the plastic particles. In terms of desorption, ideally, a 75-μm infinitely flat PE sheet as that used in this study corresponds to approximately 100 μm of an arbitrary fragment (Lee et al., 2018b). The absorbed fraction (α_{MP}) was assumed to be uniform between 0.7 and 1.0, considering gut retention time to be 10–158 h for carnivorous fish (Clements, 1997).

Fig. 4 shows the fractional contributions of the three major uptake routes (i.e., water ventilation, food ingestion, and microplastic intake)

Table 1
Summary of log K_{ow}, log K_{PDMSsw}, log K_{PDMSsm}, log K_{PEsm}, log K_{PEsw}, and log D_{PE} from literature and log $K_{PDMS/SIF}$ and log $K_{MP/SIF}$ obtained in this study.

Chemicals	log K_{ow} [b]	log K_{PDMSsw} [c]	log K_{PDMSsm} [c]	log K_{PEsm} [c]	log K_{PEsw} [c]	D_{PE} [d] ($\times 10^{-14}$ m^2 s^{-1})	log $K_{PDMS/SIF}$	log $K_{MP/SIF}$
α-HCH	3.80	2.77 (2.74, 2.80)	1.01 (1.00, 1.02)	0.65 (0.62, 0.68)	2.41 (2.36, 2.46)	1.38 (1.28, 1.40)	2.72 (2.70, 2.74)	2.36 (2.32, 2.40)
β-HCH	3.81	1.81 (1.77, 1.84)	0.20 (0.18, 0.22)	0.43 (0.40, 0.46)	2.04 (1.99, 2.09)	1.43 (1.37, 1.49)	1.84 (1.82, 1.86)	2.07 (2.03, 2.11)
γ-HCH	3.55	2.62 (2.58, 2.65)	0.88 (0.87. 0.89)	0.59 (0.55, 0.63)	2.33 (2.28, 2.38)	1.00 (0.93. 1.07)	2.58 (2.56, 2.60)	2.30 (2.03, 2.12)
PeCB	5.17	4.51 (4.40, 4.60)	1.84 (1.83, 1.85)	1.96 (1.95, 1.97)	4.63 (4.49, 4.75)	5.54 (5.01, 6.07)	4.64 (4.62, 4.66)	4.76 (4.67, 4.66)
HeCB	5.31	4.90 (4.79, 4.99)	2.07 (2.06, 2.08)	2.39[a] (2.31, 2.47)	5.22[a] (5.08, 5.34)	2.76 (2.59, 2.93)	5.10 (5.07, 5.14)	5.10 (5.07, 5.14)

[a] Value might be underestimated due to limited equilibration time. Partition coefficient values in parentheses represent the lower and upper 95% confidence limits, respectively. Diffusion coefficient values in parentheses represent standard error.
[b] Value suggested by Sangster Research Laboratory (2013).
[c] Values taken from Lee et al. (2014).
[d] Values taken from Lee et al. (2018a). α-, β-, and γ-HCH = α-, β-, and γ-hexachlorocyclohexanes, PeCB = pentachlorobenzene, and HeCB = hexachlorobenzene.

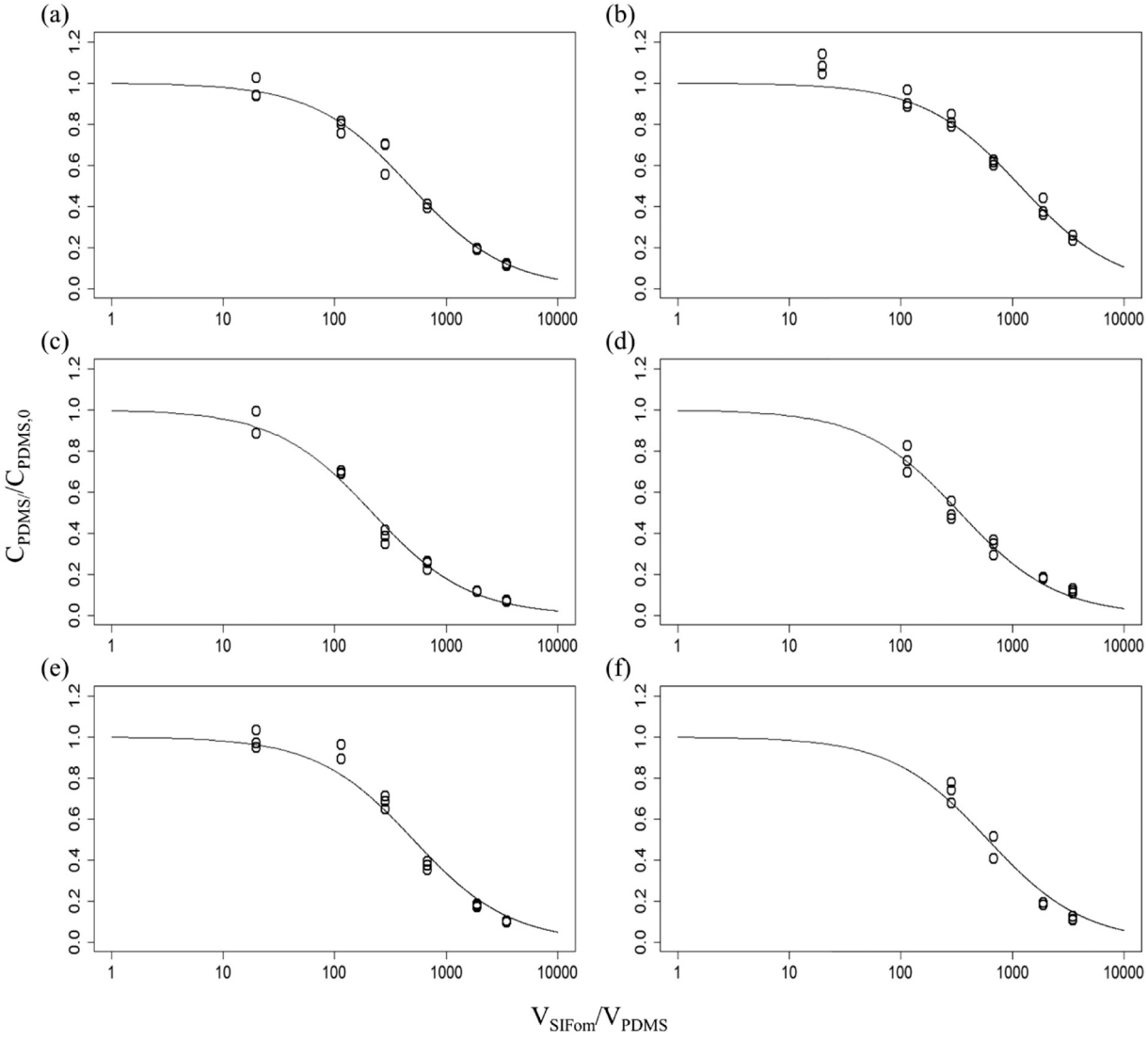

Fig. 2. Determination of partition coefficients between PDMS and dissolved organic matter solutions for (a) α-HCH and (b) HeCB using 8000 mg L^{-1} NaT, (c) α-HCH and (d) HeCB using 10,000 mg L^{-1} BSA, and (e) α-HCH and (f) HeCB using both 8000 mg L^{-1} NaT and 10,000 mg L^{-1} BSA in SIF.

for (a) α-HCH, (b) β-HCH, (c) γ-HCH, (d) PeCB, and (e) HeCB. For less hydrophobic HCHs, water ventilation is dominant over food ingestion and microplastic-bound uptake due to its low partition coefficients. Microplastic-bound uptake contributed <3% to all iterations. However, microplastics might be an important player in the uptake of more hydrophobic PeCB (Fig. 4d) and HeCB (Fig. 4e). For HeCB, contribution of microplastic-bound uptake may be higher than 10%. This is because of the enhanced desorption due to the presence of a high concentration of organic matter in the gut fluid as demonstrated in this study. However, it should be noted that the assumed intake rate of microplastics by fish was exaggerated as compared to the food intake (IR_{MP} between 0.0001 and 0.001 kg_{MP} kg_{fish}^{-1} d^{-1} and IR_{food} between 0.005 and 0.025 kg_{food} kg_{fish}^{-1} d^{-1}). Under more environmentally relevant conditions, microplastic-bound uptake would contribute less than what has been modeled.

Fig. 5 shows the resulting bioaccumulation factor ($BAF = C_{fish}/C_{sw}^{free}$) of HeCB with the increasing contribution of microplastic-bound uptake. The value of BAF was observed to be the highest at the lowest fractional contribution by microplastics to the overall uptake, while it decreased with the increasing fractional contribution. This "dilution" effect agrees

Table 2
Partition coefficients between surrogate organic matter and simulated intestinal fluid (SIF).

Chemicals	log $K_{NaT/SIF}$	log $K_{BSA/SIF}$	log $K_{NaT+BSA/SIF}$ (experimental)	log $K_{NaT+BSA/SIF}$ (estimated)
α-HCH	1.11 (1.06, 1.17)	2.15 (2.09, 2.20)	0.34 (0.26, 0.41)	1.93 (1.87, 1.98)
β-HCH	–	2.61 (2.53, 2.67)	2.18 (2.15, 2.22)	2.35 (2.27, 2.41)
γ-HCH	–	1.94 (1.89. 1.98)	1.35 (1.29, 1.40)	1.68 (1.63, 1.72)
PeCB	3.61 (3.54, 3.68)	4.25 (4.19, 4.30)	3.77 (3.72, 3.82)	4.07 (4.01, 4.12)
HeCB	4.11 (4.03, 4.18)	4.57 (4.50, 4.63)	4.06 (3.97, 4.13)	4.42 (4.35, 4.48)

*Values in parentheses represent the lower and the upper confidence limits, respectively.

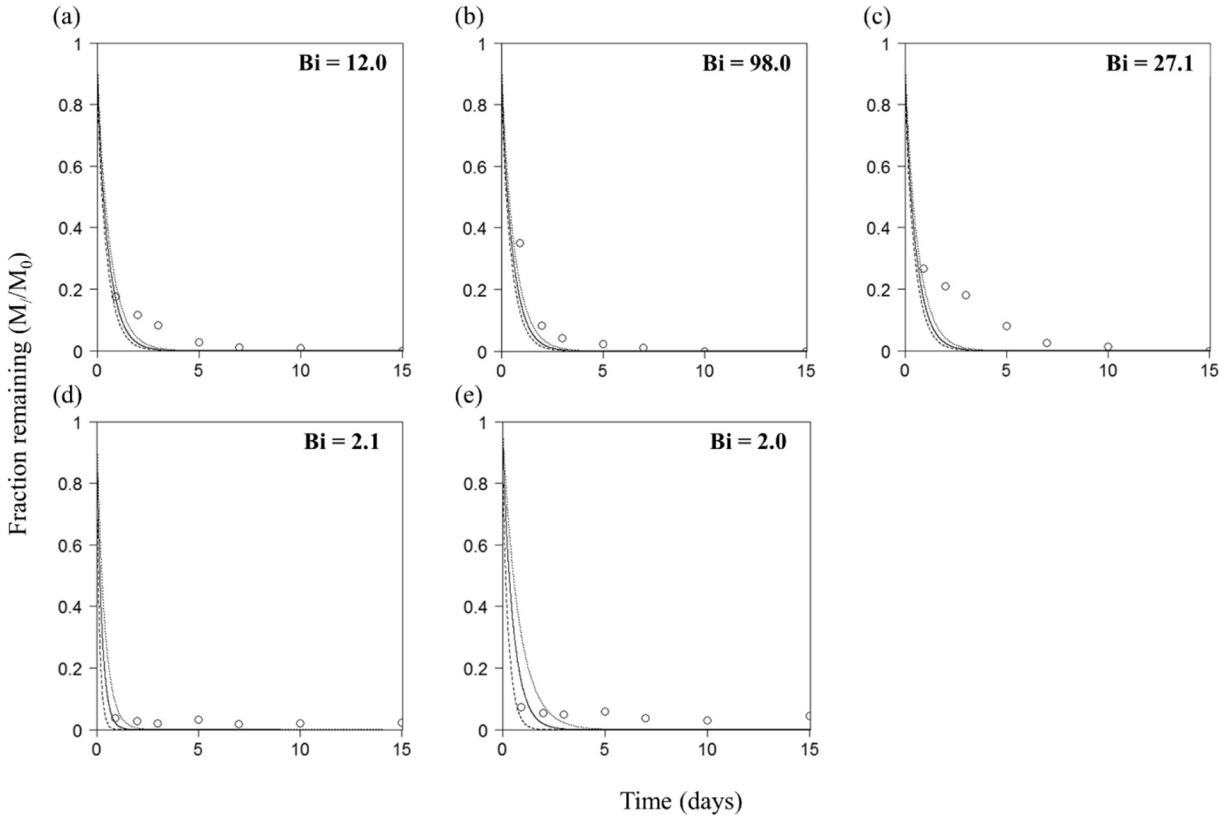

Fig. 3. Fraction of HOCs remaining in the polyethylene sheet (M/M_0) after the designated desorption time (t) for (a) α-HCH, (b) β-HCH, (c) γ-HCH, (d) PeCB, and (e) HeCB. Lines are values predicted using the convection-diffusion model, where thickness of the aqueous boundary layer (δ_w) is: 100 (dashed lines), 300 (solid lines), and 1000 μm (dotted lines). Biot numbers (Bi) shown were calculated at $\delta_w = 300$ μm.

well with previous modeling studies (Gouin et al., 2011; Koelmans et al., 2013), as well as the reported loss of body-burden of lipophilic contaminants by ingestion of non-digestible lipids (Moser and McLachlan, 1999).

3.5. Implications for hydrophobic plastic additives

As demonstrated in Section 3.4, microplastic-bound uptake of HOCs that are present in equilibrium between marine microplastics and seawater is not likely to alter bioaccumulation; it should be noted that this process may, however, shorten the time required to reach a steady-state by increasing the overall uptake rate, when the intake of microplastics is comparable to food ingestion. However, this might not be the same for hydrophobic plastic additives that leach from plastic particles very slowly and are found in microplastic particles at concentrations much higher than the equilibrium concentrations with surrounding water. Hexabromocyclododecanes in expanded polystyrenes and microplastics are an example (Jang et al., 2016, 2017).

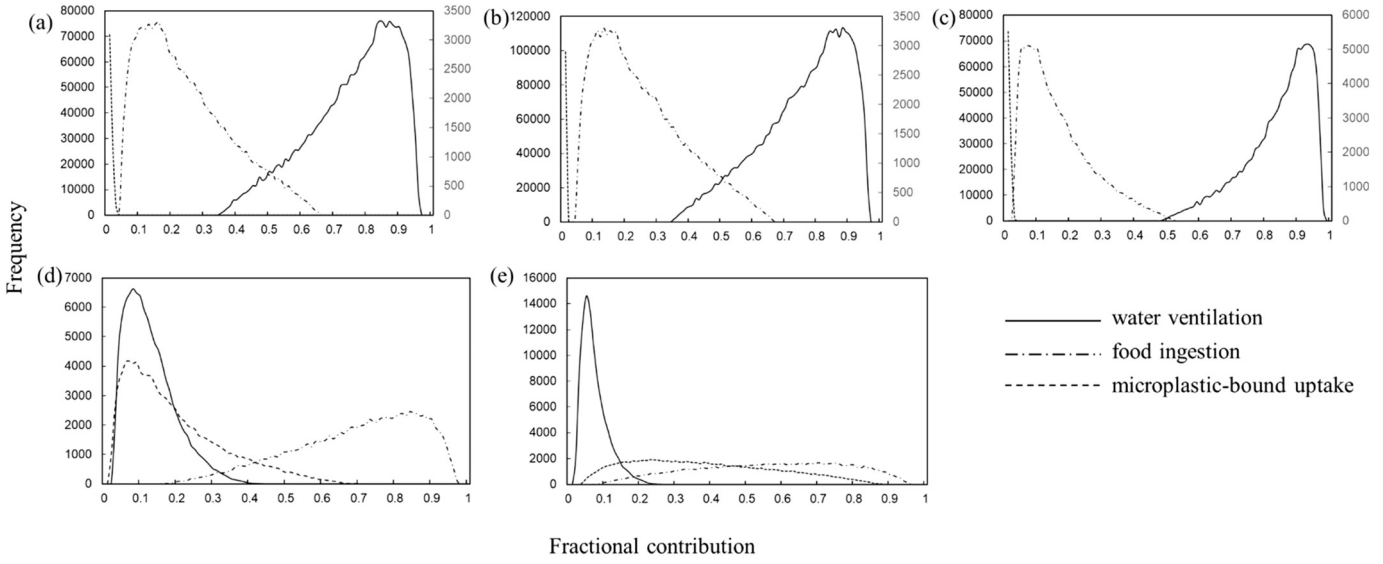

Fig. 4. Histograms showing the contribution of three exposure routes: water ventilation (solid line), food ingestion (dashed line), microplastic-bound uptake (dot-dashed line), to the overall uptake of (a) α-HCH, (b) β-HCH, (c) γ-HCH, (d) PeCB, and (e) HeCB by model fish. For HCHs, contributions by food ingestion and microplastic-bound uptake are shown on the alternative axis. The number of iterations was 100,000.

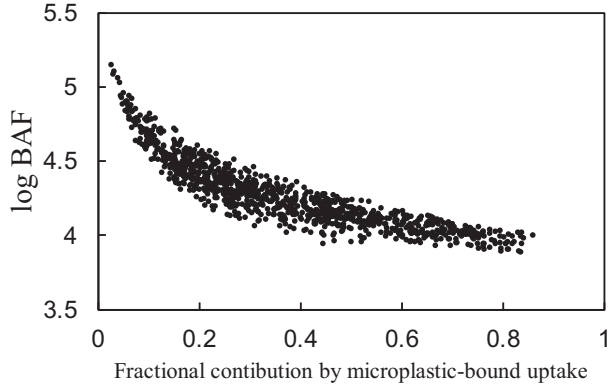

Fig. 5. The relationship between bioaccumulation factor (BAF) and the fractional contribution of microplastic-bound intake of HeCB by model fish. The number of iterations was 100,000.

For such hydrophobic additives, the term, $IR_{MP}\alpha_{MP}C_{MP}$, in Eq. (7) may dominate over the other two uptake terms, even if IR_{MP} is not as high as simulated in this study because C_{MP} could be much higher than the equilibrium concentration, $K_{MPsw}C_{sw}$. The value of k_{MP} cannot be estimated assuming equilibrium (Eq. (12)). Instead, it was estimated using a mass balance equation of

$$IR_{MP}\alpha_{MP}C_{MP} + k_{MP}C_{fish} = IR_{MP}C_{MP} \quad (17)$$

$$k_{MP} = (1-\alpha)IR_{MP}\frac{K_{MPw}}{BAF} \quad (18)$$

where k_{MP} was obtained by iterations.

For a hypothetical additive having $\log K_{ow} = 6.0$ and $\log K_{PEsw} = 6.0$, the fractional contribution by microplastic-bound uptake will be as high as 0.93, when $C_{MP}/C_w = 10^8$, $K_{MPsw}C_{sw}$, $f_{lip,\ food} = 0.05$, $IR_{food} = 0.01\ kg_{food}\ kg^{-1}d^{-1}$, $\alpha_{MP} = 0.7$, and $IR_{MP} = 0.0001\ kg_{MP}\ kg^{-1}d^{-1}$. The resulting BAF in the fish assumed in this study was 3.3×10^5, a value lower than 3.8×10^5 without ingestion of microplastics, showing a slight dilution effect. However, this BAF is still higher than the partitioning-based bioconcentration factor, $f_{lip}K_{ow} = 5.0 \times 10^4$, because seawater concentration is much lower than the equilibrium concentration in microplastics. Because microplastic-bound uptake is a dominant process, the time required to reach the steady-state will be shortened by ingestion of microplastics. Further studies on the refinement of model parameters for hydrophobic additives, such as partition coefficients and leachable fraction in the digestive tracts, are needed with appropriate validation of models that describe the contribution of microplastic-bound uptake of plastic additives.

4. Conclusions

The contribution of microplastic ingestion to the overall uptake of HOCs that are present in equilibrium with seawater would be limited even for highly hydrophobic chemicals, such as HeCB, for which desorption in artificial gut solution is generally much faster than in seawater. The steady-state BAF was predicted to decrease with increasing ingestion of microplastics, exhibiting a dilution effect. However, the model shows that the ingestion of microplastics containing hydrophobic additives, not likely in their phase-equilibrium states, with similar partitioning properties may lead to increased accumulation of these additives. Future research on the roles of microplastics as transport vectors for HOCs should focus on such hydrophobic additives.

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Appendix A. Supplementary data

Short description of the desorption model is presented in Text S1. All input parameters for the Monte-Carlo simulation are shown in Table S1. Plots of $C_{PDMS}/C_{PDMS,0}$ vs. V_{SIFom}/V_{PDMS} for the determination of $K_{BSA/SIF}$, $K_{NaT/SIF}$, and $K_{BSA+NaT/SIF}$ using Eq. (5) for β-HCH, γ-HCH, and PeCB are shown in Fig. S1. Supplementary data associated with this article can be found in the online version, at doi: https://doi.org/10.1016/j.scitotenv.2018.09.068.

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