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Desorption of Hydrophobic Organic Chemicals from Fragment-Type Microplastics

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Abstract - Microplastics provide an important medium for hydrophobic organic chemicals (HOCs), and the desorption of HOCs from microplastics is an important process for the dynamics of HOCs associated with microplastics. Although desorption kinetics has been studied for microplastics with ideal geometries, most of the microplastics isolated from the environment are irregular fragment-type microplastics. This study investigated the desorption of six model HOCs from polyethylene (PE) and polypropylene (PP) fragments to artificial seawater and compared the results with those predicted assuming ideal geometries (e.g., sphere and infinitely flat sheet) of microplastics. The experimental desorption was explained well by the model predictions with the characteristic radius for a sphere and the thickness for a plate estimated from visual imaging. The mass fraction remaining at the later stage of desorption was higher than the model simulation assuming a single characteristic length, likely due to the heterogeneity of the particle size distribution. Although there are inevitable uncertainties, it would be useful to assign a single length dimension in desorption modeling for even fragment-type microplastics, especially for the estimation of desorption half-life.

Keywords – plastic debris, partition coefficient, diffusion coefficient, desorption kinetics, Biot number

1. Introduction

Accumulation of marine plastic debris has been one of the most important environmental pollution issues (Derraik 2002; Moore et al. 2001, 2008; Thompson et al. 2004). Microplastics are defined as plastic particles with size less than 5 mm (Andrady 2011; Engler 2012), and they may contain hazardous organic chemicals as additives or sorbed from the environment. Among the many other potential harmful consequences of microplastics, desorption or leaching of hydrophobic organic chemicals (HOCs) has received significant attention because of the high sorptive capacity of microplastics toward HOCs (Bakir et al. 2014; Endo et al. 2013; Jang et al. 2017; Koelmans et al. 2013; Kwon et al. 2017; Lee et al. 2018). A quantitative description, using mathematical models, of the desorption of HOCs from microplastics is very important for estimating the release of organic additives to the surrounding environment (Kwon et al. 2017) as well as for the evaluation of potential trophic transfer of HOCs associated with microplastics to marine organisms at higher trophic levels.

Desorption of HOCs from plastic particles to a surrounding fluid medium such as seawater is often described by a twostep process: internal diffusion of HOCs in the plastic phase followed by convective mass transfer in the fluid medium (Endo et al. 2013; Lee et al. 2018; Seidensticker et al. 2017). For example, Endo et al. (2013) investigated desorption of polychlorinated biphenyls from polyethylene (PE) pellets and compared their experimental results with desorption models controlled by internal diffusion in plastic and by convective mass transfer in water, assuming an ideal sphere. Lee et al. (2018) measured diffusion coefficients of six persistent organic pollutants (isomers of hexachlorocyclohexanes and polychlorinated benzenes) and independently measured desorption of these pollutants from PE and polypropylene (PP) films. Experimental data, including both diffusion in

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the microplastic phase and convective mass transport in water, were successfully explained by model predictions. Seidensticker et al. (2017) showed that dissolved organic matter (DOM) changes the leaching of HOCs (e.g., phenanthrene, tonalide, and benzophenone) from pre-loaded spherical plastic particles and suggested that the leaching rate could be accelerated in the presence of DOM. The aforementioned modeling studies were conducted assuming ideal geometries of microplastics (i.e., infinitely flat plates and spheres) and the model results were compared with experiments using microplastics of ideal geometries, but the predominant shape of plastic particles isolated from the environment has been found to be irregular (Moore et al. 2001; Song et al. 2014). Because the particle size of irregular fragment-type plastic particles is often described by the pore size of the sieves used, it is useful to relate the fragment particle size with an equivalent characteristic length of an ideal geometry to describe desorption behaviors of HOCs from fragment-type microplastics.

In this study, we investigated desorption of HOCs from fragment-type microplastics into seawater using laboratory batch tests. PE and PP were chosen as model plastic materials because they are the two most widely used and thus the most abundantly found materials in plastic debris in the marine environment (Hidalgo-Ruz et al. 2012). For model HOCs, four hexachlorocyclohexanes (HCHs) (α -, β -, γ -, and δ -HCH) and two chlorinated benzenes (CBs) (pentachlorobenzene (PeCB) and hexachlorobenzene (HeCB)) were chosen because their partition coefficients and diffusion coefficients in PE and PP were studied in our previous research (Lee et al. 2014, 2018). PE/PP fragments were prepared in the laboratory using physical abrasion and sieving, and their size distribution was observed under a microscope. Experimental desorption of the selected HOCs from PE/PP fragments was compared with predictions obtained using a one-dimensional convectiondiffusion model for infinitely-flat and spherical geometries. The range of characteristic length for desorption modeling for heterogeneous PE/PP fragments was then established.

2. Materials and Methods

Materials and chemicals

The selected model HOCs were of high purity (\geq 98%) and were purchased from Sigma-Aldrich (St. Louis, MO, USA), Supelco (Bellefonte, PA, USA), or Fluka (Buch, Switzerland). The desorption experiments were conducted using two mixtures: an HCHs mixture (α -, β -, γ -, and δ -HCH) and a CBs mixture (PeCB and HeCB). Artificial seawater was prepared at 3.50% (w/w) by dissolving artificial sea salt purchased from Sigma-Aldrich in deionized water. The chemical properties of all the HOCs are listed in Table 1.

Medical-grade polydimethylsiloxane (PDMS) sheets (thickness of 1.0 mm, density of 1.17 g cm⁻³, purchased from Specialty Silicone Products, Inc. (Ballston Spa, NY, USA)) were used as a medium to satisfy the infinite sink condition for the desorption experiments. The PDMS was cut into 10 mm × 50 mm rectangular sheets before use, and each custom-cut sheet was cleaned using a sufficient volume of *n*-hexane and methanol for 2 h. After the cleaning, the PDMS sheets were stored in methanol until use.

For the evaluation of desorption from fragment-type plastic particles, microplastic was prepared from PE and PP labware using a based-cut hand file, as described in our previous study (Lee et al. 2014). To separate large particles, the microplastic obtained through this process was sieved using a sieve with a mesh size of 250 μ m. To quantify the size distribution of the plastic fragments in suspension, visual images of more than 10 μ m were taken for each plastic material using an inverted microscope (IX71, Olympus, Tokyo, Japan). More than 100 particles were examined for statistical analysis of

Table 1. Values of log K_{ow} , log K_{PDMSsw} , log K_{PEsw} , and log K_{PPsw} for the selected HOCs along with their diffusion coefficient values for PE and PP reported in the literature

Chemicals	log K _{ow}	log K _{PDMSsw} ^c	log K _{PEsw} ^c	log K _{PPsw} ^c	$\mathrm{D}_{\mathrm{PE}}^{}\mathrm{d}}$	$\mathrm{D_{PP}}^{d}$
α-HCH	3.79 ^a	2.77 (2.74, 2.80)	2.41 (2.36, 2.46)	2.69 (2.64, 2.75)	1.38 (± 0.10)	5.31 (± 0.42)
β-нсн	3.88 ^a	1.81 (1.77, 1.84)	2.04 (1.99, 2.09)	2.18 (2.08, 2.28)	1.43 (± 0.06)	4.57 (± 0.23)
ү-НСН	3.72 ^a	2.62 (2.58, 2.65)	2.33 (2.28, 2.38)	2.58 (2.52, 2.64)	$1.00 (\pm 0.07)$	5.44 (± 0.38)
δ-нсн	4.17 ^a	2.17 (2.13, 2.20)	2.08 (2.03, 2.12)	2.23 (2.13, 2.34)	$1.08 (\pm 0.07)$	7.26 (± 0.35)
PeCB	4.94 ^b	4.51 (4.40, 4.60)	4.63 (4.49, 4.75)	4.50 (4.39, 4.59)	5.54 (± 0.53)	8.01 (± 0.55)
HeCB	5.46 ^b	4.90 (4.79, 4.99)	5.22 (5.08, 5.34)*	5.01 (4.89, 5.10)	2.76 (± 0.17)	6.01 (± 0.35)

*Value might be underestimated due to limited equilibration time. Partition coefficient values in parentheses are the lower and upper 95% confidence limits. Diffusion coefficient values in parentheses represent standard error. ^aPaschke and Schüürmann (1998). ^bBahadur et al. (1997). ^cLee et al. (2014). ^dLee et al. (2018). α -, β -, γ -, and δ - HCH = α -, β -, γ -, and δ -hexachlorocyclohexanes, PeCB = pentachlorobenzene, and HeCB = hexachlorobenzene

the plastic fragments using ImageJ software (NIH, Bethesda, MD, USA). The area of each fragment was measured by using the 'particle analysis' option in ImageJ software.

Determination of the characteristic length for plastic fragments is visualized in Figure A1 (Appendix).

Desorption of HOCs from microplastic fragments

Desorption of the selected HOCs from the heterogeneous PE or PP microplastics was evaluated as shown schematically in Fig. 1. The model HOCs were loaded into the microplastic by placing approximately 10 mg of ground microplastic fragments into a vial containing 4 mL of *n*-hexane solution with the test chemical mixture and then agitating the vial at 150 rpm in a shaking incubator for 24 h. After shaking, the *n*-hexane solution was filtered through a glass fiber filter (pore size of 1.2 μ m) to separate the microplastic fragments loaded with HOCs from the solution. To remove residual *n*-hexane on the particle surfaces, the filtered microplastic fragments were washed by passing 2 mL of methanol–water mixture (methanol:water = 8:2, v/v) and then distilled water through the glass fiber filter.

An aqueous concentration of model HOCs of nearly zero (i.e., infinite sink condition) was maintained in each test vial by placing a sufficiently large volume of PDMS into the vial (Fig. 1). The vials were shaken for predetermined time intervals to measure the remaining HOCs in the plastic fragments. After the designated number of days, the microplastics were removed from the suspension using a spatula and placed on a glass fiber filter. After removing the water using a suction pump, the mass of the remaining dried microplastics was measured. Immediately after the mass measurement, the microplastics were placed in an extraction vial containing 1 mL of *n*-hexane and the vial was shaken at 25°C and 150 rpm in a shaking incubator for one day. Both the artificial seawater and PDMS were extracted to obtain the remaining mass of HOCs in the seawater and the PDMS. In short, 10 mL



Fig. 1. Schematic diagram of the experimental system for measuring the desorption kinetics of the analytes from plastic debris

of seawater and PDMS samples were extracted using 10 mL of *n*-hexane for 24 h at 25°C and 150 rpm. All extraction samples were concentrated to 200 μ L under a gentle stream of nitrogen gas and then subjected to gas chromatography–electron capture detector analysis.

Desorption models

To simulate desorption from fragmental microplastics into the surrounding medium, two ideal geometries, a sphere and an infinitely flat sheet, were assumed. Using the experimentally determined diffusion coefficients and partition coefficients, the remaining fraction (M/M_0) was calculated following Crank (1975):

$$\frac{M}{M_0} = \sum_{n=1}^{\infty} \frac{6\mathrm{Bi}^2 \exp\left(-\frac{\beta_n^2 Dt}{r^2}\right)}{\beta_n^2 \{\beta_n^2 + \mathrm{Bi}(\mathrm{Bi} - 1)\}} \quad \text{(for a sphere)} \quad (1)$$

$$\frac{M}{M_0} = \sum_{n=1}^{\infty} \frac{2\mathrm{Bi}^2 \exp\left(-\frac{\beta_n^2 Dt}{L^2}\right)}{\beta_n^2 \{\beta_n^2 + \mathrm{Bi}(\mathrm{Bi}+1)\}} \quad \text{(for a sheet)}$$
(2)

where the β_n 's are the roots of

$$\beta_n \cot \beta_n + \operatorname{Bi} - 1 = 0$$
 (for a sphere) (3)

$$\beta_n \tan \beta_n = \operatorname{Bi} \quad \text{(for a sheet)}$$
 (4)

The mass transfer Biot numbers are given as

$$Bi = \frac{kr}{D} \text{ (for a sphere) or } Bi = \frac{kL}{D} \text{ (for a sheet)}$$
(5)

where *r* is the radius of the sphere (m), 2*L* is the thickness of the sheet, and *D* is the diffusion coefficient in the microplastic phase (m² s⁻¹). Figure A2 (Appendix) shows the range of the radii of the microplastics estimated from the microscopic images. The mass transfer coefficient (*k*) in water, including the instantaneous partitioning between plastic phase and seawater, is estimated as

$$k = \frac{D_w}{K_{pw}\delta_w} \tag{6}$$

where D_w is the molecular diffusion coefficient in artificial seawater (m² s⁻¹), K_{pw} is the plastic–water partition coefficient (-), and δ_w is the thickness of the aqueous boundary layer (m). For the spherical geometry, δ_w is not the actual thickness of the aqueous boundary layer, but the length corresponding to the thickness of a flat geometry. The D_w was estimated using the Hayduk-Laudie correlation (Hayduk and Laudie 1974). The molar liquid volume of chemicals was estimated using the LeBas methods (Reid et al. 1977):

$$D_w = \frac{13.26 \times 10^{-9}}{\eta^{1.4} ML V^{0.589}}$$
(7)

where η is the viscosity of artificial seawater (0.97 mPs at 25°C) and MLV is the LeBas molar liquid volume (cm³ mol⁻¹).

Instrumental analyses

The concentrations of the six HOCs were quantified using a Hewlett-Packard 5890 Series II gas chromatograph system equipped with an electron capture detector (ECD). Three microliters of the hexane extract containing HCHs or CBs was injected and separated on an HP-5 column ($30 \text{ m} \times 0.25 \text{ mm}$ 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, subsequently increased to 200°C at 5°C min⁻¹, held for 5 min, increased to 280°C at 45°C min⁻¹, and held for 1 min. The injector and ECD temperature were 200 and 320°C, respectively.

The concentration of HCHs and CBs in the extract was quantified using a calibration curve obtained using external standards at five different concentrations over a linear range of $12.5-100 \text{ nmol } \text{L}^{-1}$ for CBs and $5-40 \text{ nmol } \text{L}^{-1}$ for HCHs.

The coefficient of linear regression (r^2) was at least 0.98 for all analytes. *n*-Hexane extract of cleaned PDMS was used as a reagent blank and all analytes were not detected in the reagent blanks.

3. Results and Discussion

Characterization of PE and PP particles using visual analyses

The characteristic radius (*r*) of the fragments was determined by visual analysis assuming a spherical particle with the same projected area. Figure A2 (Appendix) shows the size distributions of 526 PE and 538 PP particles with 15 μ m intervals. The number-weighted average radii (25 and 75 percentile values and mode) for PE and PP particles were 73 (25, 105, 23) μ m and 72 (40, 100, 37) mm, respectively. The projected area-weighted average radii were 167 μ m for PE and 123 μ m for PP particles. These values are approximately two times higher than the number-weighted average radii.

Desorption of HOCs from fragment-type microplastics and prediction of desorption using ideal geometries

Figures 2 and A3 (Appendix) show the desorption of the selected HOCs from PE/PP fragments as mass of HOCs remaining per initial mass of HOCs in the plastic fragments (M/M_0). As shown, the desorption of HCHs was much faster than the desorption of CBs because HCHs have lower plastic/water partition coefficients than CBs. This tendency agrees with our earlier observation using PE/PP films (Lee



Fig. 2. Fraction of HOCs remaining in the plastic debris (M/M_0) with desorption time (*t*) for (a) α -HCH from PE particles and (b) from PP particles and (c) PeCB from PE particles and (d) from PP particles. Lines are values predicted using Equation 1 where the thickness of the aqueous boundary layer (δ_w) is 300 µm. Solid, dashed, and dotted lines represent the number-weighted average, mode, and projected area-weighted average values of estimated radius assuming spherical particles

et al. 2018). The experimental measurements were compared with the theoretical predictions made by assuming δ_w of 300 µm (Lee et al. 2018) and ideal spherical particles with three representative diameters, the mode, the number-weighted average, and the projected area-weighted average values from the visual analyses. For all of the HOCs, the mass fraction remaining (M/M_0) agreed well with the range of the theoretical predictions, especially in the middle of the desorption stage, although the model parameters were not adjusted for the experimental data. However, because desorption occurs more slowly from larger fragments, the experimental M/M_0 was slightly lower than predicted by the model at the earlier stage of desorption, and the experimental M/M_0 was slightly greater than predicted at the later stage. For the prediction of desorption half-lives, a single parameter desorption model assuming spherical geometry provides reasonable estimates despite the heterogeneity of plastic particles.

Figures 3 and 4 show the desorption of the HOCs (i.e., (a) α -HCH, (b) β -HCH, (c) γ -HCH, (d) δ -HCH, (e) PeCB, and (f) HeCB) from PE (Fig. 3) and PP (Fig. 4) microplastic fragments along with model predictions assuming infinitely

flat sheets. The five lines in Figures 3 and 4 represent model predictions assuming a sheet half-thickness of 10 (dashed lines), 30 (solid lines), 50 (dotted lines), 100 (dot-dash lines), and 200 (long-dash lines) μ m, respectively. The experimental M/M_0 values agreed well with the model predictions assuming sheet half-thickness of 30 to 100 μ m for PE and 10 to 30 μ m for PP. The half-thickness values from the desorption fraction were generally thicker for PE than for PP. The thicker and wider thickness range in PE might be due to the various characteristic lengths of irregular-fragment particles. Because irregular-fragment particles have various cross-sectional areas, the characteristic length also has various values. Nevertheless, modeling to infinitely flat geometry and to characteristic lengths would be useful for estimating the half-lives for HOCs from irregular fragment microplastics.

Implications of the study for estimating organic chemicals leaching from microplastics in the environment

Leaching of organic chemicals from microplastics is important in evaluating risks of microplastics (e.g., Koelmans et al. 2013; Rochman et al. 2013; Teuten et al. 2009). A general



Fig. 3. Fraction of HOCs remaining in the plastic debris (M/M_0) with desorption time (t) for (a) α-HCH, (b) β-HCH, (c) γ-HCH, (d) δ-HCH, (e) PeCB, and (f) HeCB from PE particles. Lines are values predicted using Equation 2 where the thickness of the aqueous boundary layer (δ_w) is 300 µm. Dashed, solid, dotted, dot-dash, and long-dash lines represent 10, 30, 50, 100, and 200 µm half-thickness of sheet (L)



Fig. 4. Fraction of HOCs remaining in the plastic debris (M/M_0) with desorption time (t) for (a) α-HCH, (b) β-HCH, (c) γ-HCH, (d) δ-HCH, (e) PeCB, and (f) HeCB from PP particles. Lines are values predicted using Equation 2 where the thickness of the aqueous boundary layer (δ_w) is 300 µm. Dashed, solid, dotted, dot-dash, and long-dash lines represent 10, 30, 50, 100, and 200 µm half-thickness of sheet (L)

one-dimensional convection-diffusion model assuming ideal geometry was found to be useful for describing the leaching of hydrophobic organic chemicals from arbitrary fragmenttype microplastics. Because the shape of isolated microplastics from environmental samples can be easily determined under a microscope, the characteristic length of microplastics can be derived and desorption or leaching of organic contaminants from microplastics can be modeled using the convectiondiffusion model if partition coefficients (K_{pw}) and diffusion coefficients in plastics of chemicals are known or estimated. The applicability of the model is not limited to chemicals sorbed to microplastics from the environment, such as HCHs and CBs in this study. There are many hydrophobic additives with a wide range of physico-chemical properties in use (Kwon et al. 2017; Murphy 2001). Leaching of those additives from weathered microplastics would be important as well to assess the roles of microplastics as sources of contaminants.

Although an initial uniform distribution of hydrophobic organic chemicals in plastic phase was assumed in this study for simplicity, chemical concentration in real microplastics might not satisfy this assumption. Chemical concentration profiles in microplastics would vary due to the combination of various aging/fragmentation processes and leaching or absorption of chemicals. Further research is needed to test whether the general model is applicable for HOCs in microplastics isolated from field samples.

4. Conclusions

The present study suggests that the experimental desorption of HOCs from PE/PP fragments can be modeled assuming ideal geometries, spheres or infinitely flat sheets, with characteristic length obtained via visual analysis. Although the experimental results deviated from the model predictions at the earlier and later stages of the desorption tests due to the heterogeneity of the particle size distribution, desorption half-lives were shown to be estimated with relatively low uncertainties. Onedimensional desorption models with ideal geometry were found to be also useful for microplastic particles classified as irregular fragments after visual analysis.

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Appendix



Fig. A1. Schematic diagram of the determination of the characteristic length for fragment particle debris using image analysis



Fig. A2. Size distribution in abundance of (a) PE and (b) PP debris. The abundance by size categories has been analyzed with a pool of 526 (for PE) and 538 (for PP) plastic fragments

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Fig. A3. Fraction of HOCs remaining in the plastic fragments (M/M₀) with desorption time (*t*) for (a) β -HCH from PE particles and (b) from PP particles, (c) γ -HCH from PE particles and (d) from PP particles, (e) δ -HCH from PE particles and (f) from PP particles, and (g) HeCB from PE particles and (h) from PP particles. Lines are predicted values using equation 1 at thickness of the aqueous boundary layer (δ_w) of 300 µm. Solid, dashed, and dotted lines number-weighted average, mode, and projected area-weighted average of estimated radius assuming spherical particle

