

# Supporting Information

## **Microplastic as a Vector for Chemicals in the Aquatic Environment. Critical Review and Model-Supported Re-interpretation of Empirical Studies.**

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Supporting Information containing:

24 Pages

5 Tables

4 Figures

## Supporting Information for the Section:

### ‘Unifying empirical studies, modelling studies, and theory’.

*Modeling the uptake of HOCs from plastic by marine biota.* Uptake of HOC from plastic by marine biota was modeled using a previously published mass balance approach.<sup>1,2,3</sup> The HOC concentration in biota over time ( $dC_{B,t}/dt$ ) is quantified using:<sup>1</sup>

$$\frac{dC_{B,t}}{dt} = k_{\text{derm}}C_W + IR \times S_{\text{FOOD}}a_{\text{FOOD}}C_{\text{FOOD}} + IR \times S_{\text{PL}}C_{\text{PLR},t} - k_{\text{loss}}C_{B,t} \quad (1)$$

The first term accounts for dermal and gill uptake from water. The second term quantifies the accumulation from the diet. The third term quantifies partitioning between plastic and biota lipids during transfer of plastic in the gut. The fourth term is a loss term quantifying elimination and egestion. The rate constants  $k_{\text{derm}}$  and  $k_{\text{loss}}$  in the first and fourth term can be parameterised following traditional approaches.<sup>1,3,4</sup> In the second term, the ingestion rate  $IR$  represents the mass of particles ingested per unit of time and organism dry weight,  $a_{\text{FOOD}}$  is the absorption efficiency from the diet,  $S_{\text{FOOD}}$  is the mass fraction of food in the ingested material and  $C_{\text{FOOD}}$  is the HOC concentration in the diet. The product  $a_{\text{FOOD}} \times C_{\text{FOOD}}$  quantifies the contaminant concentration that is transferred from food, i.e. prey, to the organism during gut passage. In the third term,  $S_{\text{PL}}$  is the mass fractions of plastic in the ingested material ( $S_{\text{FOOD}} + S_{\text{PL}} = 1$ ).  $C_{\text{PLR},t}$  is the HOC concentration transferred from or to plastic during gut passage<sup>1,2,5</sup>.

$$C_{\text{PLR},t} = \frac{k_{1G}C_{\text{PL}} - k_{2G}C_{\text{L},t}}{k_{1G} + \frac{M_{\text{PL}}}{M_{\text{L}}}k_{2G}} \left( 1 - e^{-\left(k_{1G} + \frac{M_{\text{PL}}}{M_{\text{L}}}k_{2G}\right)GRT} \right) \quad (2)$$

in which  $GRT$  is gut residence time and  $M_{\text{PL}}/M_{\text{L}}$  is the ratio of plastic and lipid mass in the organism. The direction of HOC transfer depends on the gradient  $k_{1G}C_{\text{PL}} - k_{2G}C_{\text{L},t}$ . If this term is positive, HOCs are transferred from the ingested plastic to the biota lipids. If the term is negative, HOCs are transferred to the plastic leading to extra excretion. Such an increased excretion of HOCs has been modelled in a similar fashion for the effect of non-digestible lipophilic food components in the human diet.<sup>6</sup> This gradient depends on the forward and backward first order rate constants  $k_{1G}$  and  $k_{2G}$  for transport between plastic and biota lipids inside the gut, and on the concentrations in the plastic ( $C_{\text{PL}}$ ) and biota lipids ( $C_{\text{L}}$ ). In case of equilibrium, the gradient is zero and  $C_{\text{L}}/C_{\text{PL}} = k_{1G} / k_{2G}$ . This equilibrium may apply to the plastic at the moment of ingestion, when the particles already are at or close to equilibrium with seawater (see first section of this review), or equilibrium may be achieved during gut passage, in which case HOC transfer would take place only in the first part of the gut. At equilibrium, the ratio  $C_{\text{L}}/C_{\text{PL}}$  also equates to the ratio of the lipid to water ( $K_{\text{LIP}}$ ) and the plastic to water partition coefficients, which implies that  $k_{2G} = K_{\text{PL}}/K_{\text{LIP}} \times k_{1G}$ . Consequently,  $k_{2G}$  follows from  $k_{1G}$  and we can use the HOC desorption rate constant from plastic  $k_{1G}$  as primary parameter to calibrate the contribution of plastic ingestion to bioaccumulation. For further details on the sources for the parameter values the reader is referred to the Supporting Information Table SI-3.

**Calibration of the role of MP in the uptake of nonylphenol, phenanthrene, triclosan and/or PBDE-47 by the lugworm *Arenicola marina*.** The model described in the previous section (Eq. 1 and 2) was applied to data provided by Browne et al.<sup>7</sup> Non-contaminated lugworms (*A. marina*) were exposed to sand or sand mixed with 5% of 230  $\mu\text{m}$  PVC microplastic that was presorbed with nonylphenol, phenanthrene, triclosan and/or PBDE-47. Because clean worms were used, a gradient from the PVC to the organism was created and chemical transfer from the particles to the worms occurred in 10 d. No other natural pathways were accounted for, like dermal uptake or chemical uptake from digestion of ingested organic matter. Consequently, in the model calibration the first two uptake terms in Eq. 1 were omitted. Treatments with sand caused higher bioaccumulation than treatments with plastic, indicating that the uptake probably was a particle effect rather than a plastic effect. The model was parameterised using information provided by the authors<sup>7</sup> and literature data (details provided as Supporting Information Table SI-3). Using their reported nonylphenol concentrations in PVC of 692 mg/g, the specific worm weight, plastic concentration of 5% d.w. and a specific NP partition coefficient for PVC (Table SI-3), their measured tissue concentrations in body wall and gut could be reproduced by using a value for  $k_{\text{IG}}$  of 4.1 and 9.8  $\text{d}^{-1}$ , respectively (Table SI-5). For phenanthrene these values were a little lower, i.e. 0.76 and 1.6  $\text{d}^{-1}$  respectively. For triclosan and PBDE-47, the model output was insensitive to the value of  $k_{\text{IG}}$ , which implies that no parameters could be estimated.

**Calibration of the role of MP in the uptake of PBDEs by the amphipod *Allorchestes compressa*.** Chua et al.<sup>8</sup> fed a diet of 100% PBDE-spiked 11-700  $\mu\text{m}$  PE microplastics to amphipods (*Allorchestes compressa*). Because clean amphipods were used, a gradient from the PE to the organism was created and PBDE transfer from the PE to the worms occurred in 3 d. No natural uptake pathways were accounted for, like dermal uptake or chemical uptake from digestion of ingested organic matter and the authors claimed that ingestion of plastic was the only pathway. Consequently, uptake can be modelled with the first two uptake terms in Eq. 1 omitted. The model used the high PBDE concentrations applied in the test, species weight, and PBDE partition coefficients for PE. Details on the model parameters are provided as Supporting Information (Table SI-3). Time to reach equilibrium was calculated for all PBDEs and only those that had reached equilibrium (PBDE-28, 47, 100, 99) in 3 d were used in the calculation of one generic  $k_{\text{IG}}$  value of 0.073  $\text{d}^{-1}$  for these compounds (Table SI-5).

**Calibration of the role of MP in the uptake of PCBs and PAH by Japanese medaka; *Oryzias latipes*.** Rochman et al.<sup>9</sup> exposed fish (Japanese medaka; *Oryzias latipes*) to contaminated food, to contaminated food mixed with 10% virgin low density PE (LDPE) and to contaminated food mixed with LDPE that was pre-equilibrated in seawater. The latter treatment showed an increase in body burdens after two months of a factor of 2.4 ( $\Sigma\text{PAH}$ ), 1.2 ( $\Sigma\text{PCB}$ ) and 1.8 ( $\Sigma\text{PBDE}$ ), which was statistically significant for part of the test compounds (chrysene, PCB28 and most PBDEs). Because in this experiment also contaminated food was included, it better mimics natural conditions where the compounds tested would also be present in all environmental media. Consequently, also the second term in the model (Eq. 1) was implemented. Data on concentrations of plastic, food and chemicals in plastic and food were taken from Rochman et al.<sup>9</sup> Information on lipid content and

concentrations in water (negligible) were obtained from Chelsea Rochman (personal communication). Details on the model parameters are provided as Supporting Information (Table SI-3). Calibration of the model was performed in two steps. First, the apparent food ingestion rate (IR) was calibrated using the PCB data from the negative (i.e. no plastic) control treatment. PCB data were used for this calibration because they are best suited to act as tracer chemicals, as opposed to the less persistent PAH and PBDE. Subsequently, the plastic HOC exchange rate constant  $k_{IG}$  was optimized for PCB and PAH, using the data for the marine plastic treatment, with IR set at the value obtained from the aforementioned calibration based on the control (food-only) treatment data. This procedure thus assumes that the fish had the same food ingestion rate in both treatments. For PAH and PCB, time to reach steady state bioaccumulation (Eq. 1) was calculated to be 33 d at most (PCB187), which was less than the exposure time (two months). For the PBDEs however this was between 50 and 130 d, which is why the PBDE data could not be interpreted with the model (as it was used in steady state mode). For the PCB and PAH a fair fit to the data was obtained (Figure S2). The apparent exchange rate constants  $k_{IG}$  for PCB and PAH were calculated to be 1.89 and 1.16 d<sup>-1</sup>, respectively (Table SI-5).

***Calibration of the role of MP based on observed HOC desorption rates from plastic to artificial gut fluids.*** Teuten et al<sup>10</sup>, provided  $k_{IG}$  equivalent first order rate constants for desorption of phenanthrene from PE and PVC in artificial gut fluid (sodium taurocholate in seawater), between 2.29 and 4.09 d<sup>-1</sup>. In a follow-up study that used the same approach, Bakir et al<sup>11</sup> reported a range of 0.27 – 3 d<sup>-1</sup> for desorption of three HOCs from PE and PVC (Table SI-5). To mimic gut conditions in warm-blooded animals, the experiments were repeated at a temperature of 38°C and a lower pH, which yielded a slightly higher range of 0.54 – 12.1 d<sup>-1</sup> (Table SI-5).

Recently, Tanaka et al<sup>12</sup> reported data on the leaching of BDE209 from plastic to artificial gut fluids (fish oil at 38°C and seabird stomach oil at 38°C). However, they did not fit a desorption model to their data and therefore did not provide the kinetic desorption rate constants. Here we provide these constants by fitting a first order two compartment model to their data. The data provided by Tanaka et al<sup>12</sup> were the fractions (as percentages) of BDE209 leached from plastic to the liquid phase (fish or stomach oil) as a function of time (Figure S3). Consequently, the concentration in the liquid phase equates to:

$$C_{oil,t} = \frac{Q_T P_t}{L} \quad (3)$$

In which  $C_{oil}$  (μg/L) is the BDE209 concentration in the fish or stomach oil liquid phase,  $Q_T$  (μg) is the mass of BDE209 initially present in the plastic,  $P_t$  is the fraction of the initial quantity that has leached out after time  $t$  (d), and  $L$  (L) is the volume of the oil. Similarly, the concentration of BDE209 remaining in the plastic equates to:

$$C_{PL,t} = \frac{Q_T(1-P_t)}{S} \quad (4)$$

In which  $C_{PL,t}$  ( $\mu\text{g}/\text{kg}$ ) is the BDE209 concentration in the plastic after time  $t$ , and  $S$  ( $\text{kg}$ ) is the weight of the plastic in the system. Combination of eqs 3 and 4 yields an equation for the time dependent partition coefficient  $K_{P,t}$  ( $\text{L}/\text{kg}$ ):

$$K_{P,t} = \frac{Q_T(1-P_t)}{Q_T P_t} \times \frac{L}{S} = \frac{1-P_t}{P_t} \times \frac{L}{S} \quad (5)$$

Due to the fact that desorption is measured, initially, the experimentally determined values for this coefficient are high. When time proceeds,  $K_{P,t}$  slowly decreases to a constant value, which implies that sorption equilibrium is reached. At that time, the value of  $K_{P,t}$  is equal to the equilibrium coefficient for partitioning of BDE209 between plastic and oil. The liquid to solid ratio ( $L/S$ ) in the experiment was specified as  $1000 \text{ L}/\text{kg}$  (Tanaka et al<sup>12</sup>). Eqs 3, 4 and 5 are provided here to show how partition coefficients can be derived from the ‘percentage leached’ data provided by Tanaka et al.<sup>12</sup> These experimentally determined  $K_{P,t}$  values are plotted in Figure S4.

From these measured coefficients  $K_{P,t}$ , the ad- and desorption rate constants ( $k_u$ ,  $k_d$ ;  $\text{d}^{-1}$ ) and equilibrium partition coefficients ( $K_P$ ) were obtained using two differential equations that describe leaching as a function of time:

$$\frac{dC_{PL,t}}{dt} = k_u \frac{L}{S} C_{oil} - k_d C_{PL,t} \quad (6)$$

$$\frac{dC_{oil,t}}{dt} = k_d \frac{S}{L} C_{PL,t} - k_u C_{oil,t} \quad (7)$$

These equations describe the leaching of BDE209 from the plastic as a dynamic process, taking backward sorption from the oil to the plastic into account. The rate constant  $k_d$ , for desorption from the plastic to the artificial gut fluids ‘38°C fish- and stomach oil’ is equivalent to the desorption rate constant in the gut  $k_{IG}$  as in Eq.2.

The analytical solution to the coupled differential equations 6 and 7 was published before<sup>13,14</sup> and reads:

$$K_{P,t} = \frac{k_u + k_d e^{-(k_u + k_d)t}}{k_d (1 - e^{-(k_u + k_d)t})} \times \frac{L}{S} \quad (8)$$

At  $t=0$ , Eq. 8 predicts infinitely large  $K_{P,t}$ , which agrees to the boundary condition that at that time all BDE209 is in the plastic and the concentration in the liquid phase is zero. At infinite time, Eq. 8 reduces to:

$$\frac{k_u}{k_d} \times \frac{L}{S} = K_P \quad (9)$$

i.e. the equilibrium partition coefficient.

Eq. 8 was fitted to the raw plastic–fish oil and plastic–stomach oil partitioning data provided by Tanaka et al<sup>12</sup> using the Microsoft excel solver tool. The parameters  $k_u$  and  $k_d$  were fitted against a relative least squares criterion. The model fitted the data well (Figure S4) and parameters are provided in Table SI-4. The data in Table SI-4 represent the first published full set of sorption data for exchange of HOCs to (artificial) gut fluids, including forward, backward kinetic constants and equilibrium partition coefficients.

The values obtained from these experiments reported by Teuten et al<sup>10</sup>, Bakir et al<sup>11</sup> and those newly calculated based on data from Tanaka et al<sup>12</sup>, agree well to the value obtained with the modelling of the bioaccumulation studies (Table SI-5).

***Calibration of the role of MP based on first principles.*** The desorption of organic chemicals from porous media like polymers or sediments is well understood and can be described using diffusion theory.<sup>15,16,17</sup> For polymers (i.e. passive samplers) in surface waters usually two transfer resistances are considered: an internal resistance in the particle governed by intrapolymer diffusion and a resistance in the UBL surrounding the particle and separating it from the bulk water.<sup>17,18,19</sup> Transport rates for the UBL are determined by aqueous molecular diffusion rates, but these rates have been found to be increased due to presence of DOC,<sup>20,21</sup> proteins<sup>22</sup>, bacteria<sup>23</sup> or particle collisions.<sup>24,25</sup> Taking these processes into account, Koelmans et al<sup>1</sup> demonstrated that polymer diffusion was much slower than diffusion across the UBL, which implies that the release rates in the gut can be calculated from reported polymer diffusion coefficients. Resulting theoretical values for  $k_{IG}$  ranged 0.1 – 2 d<sup>-1</sup> for PCBs desorbing from 0.3 – 1.4 mm PS particles as used by Besseling et al<sup>1,26</sup> (Table SI-5). Here we provide new calculations showing a range of 0.9 – 9.8 d<sup>-1</sup> for the PCBs and a range of 1.67 – 53 d<sup>-1</sup> for the PAH used by Rochman et al<sup>9</sup> (Table SI-5).

**Table S1.** Calculation of the distribution of HOC across environmental media.

**A.** Calculation of the distribution of a model HOC across environmental media DOC, colloids, detritus, phytoplankton, bacteria, zooplankton, plastic and water.

Medium	g C	kg C	ton C or plastic	Koc (L/kg)	M*Koc or Vw (L)	% HOC per solid phase	% HOC all phases
DOC	1E+17	1E+14	1E+11	1,E+05	1E+19	42,8	0,7
Colloids	1E+17	1E+14	1E+11	1,E+05	1E+19	42,8	0,7
Detritus	3E+16	3E+13	3E+10	1,E+05	3E+18	12,8	0,2
Phytopl	3E+15	3E+12	3E+09	1,E+05	3E+17	1,28	0,02
Bacteria	5E+14	5E+11	5E+08	1,E+05	5E+16	0,21	0,00
Zoopl	1E+14	1E+11	1E+08	1,E+05	1E+16	0,043	0,001
<b>Plastic</b>		2,69E+08	2,69E+05	1,E+07	2,69E+15	<b>0,01151</b>	<b>0,00019</b>
Water					1,37E+21		98,3

**B.** As in ‘A’, however with the non-settling media water and DOC excluded.

Medium	g C	kg C	ton C or plastic	Koc (L/kg)	M*Koc or Vw (L)	% HOC per solid phase	% HOC all phases
DOC	0	0	0	1,E+05	0	0,0	0,0
Colloids	1E+17	1E+14	1E+11	1,E+05	1E+19	74,8	74,8
Detritus	3E+16	3E+13	3E+10	1,E+05	3E+18	22,5	22,5
Phytopl	3E+15	3E+12	3E+09	1,E+05	3E+17	2,25	2,25
Bacteria	5E+14	5E+11	5E+08	1,E+05	5E+16	0,37	0,37
Zoopl	1E+14	1E+11	1E+08	1,E+05	1E+16	0,075	0,075
<b>Plastic</b>		2,69E+08	2,69E+05	1,E+07	2,69E+15	<b>0,02013</b>	<b>0,02013</b>
Water					0,00E+00		0,0

**Table S2.** Overview of studies addressing the role of microplastic ingestion by marine aquatic organisms on bioaccumulation of plastic associated chemicals. General approach, type of evidence, and environmental realism are summarised.

Source	Species studied	Plastic concentration or fraction of diet	Plastic ingestion confirmed <sup>(a)</sup>	Environmentally relevant processes accounted for <sup>(b)</sup>	Comments
<b>Controlled experimental studies</b>					
Besseling et al., 2013	<i>A. Marina</i>	0, 0.073, 0.73, 7.3 % d.w. PS in sediment	Yes	1,2,3,4,5,6	Mimicked environmentally relevant exposure in the lab, including all pathways. Provided causal evidence on the treatment level.
Browne et al., 2013	<i>A. Marina</i>	5% d.w. PVC in sand, mixed with 8% d.w. Isochrysis galbana as food	No	1,(3)	Did not consider uptake from water and food, and used initially clean organisms. Considering the experimental design, uptake from water could have occurred as well. Provided causal evidence on the treatment level.
Rochman et al., 2013	<i>Oryzias latipes</i>	10% d.w. LDPE in the diet.	No	1, (2,3,5),6	Accounted realistically for uptake from plastic and food. Considering the experimental design, uptake from water could have occurred as well. Provided causal evidence on the treatment level.
Chua et al., 2014	<i>Allorchestes compressa</i>	100 g/L, 100% PE. No food.	Yes	1,(3)	Did not consider uptake from water and food, and used initially clean organisms. Considering the experimental design, uptake from water could have occurred as well. Used unrealistically high plastic concentrations. Provided causal evidence on the treatment level.
Avio et al., 2015	<i>Mytilus galloprovincialis</i>	1.5 g/L, 100% PE and PS. No food.	Yes	1,(3)	Did not consider uptake from water and food, and used initially clean



					organisms. Considering the experimental design, uptake from water could have occurred as well. Used very high plastic concentrations. Provided causal evidence on the treatment level.
<b>Modeling studies</b>					
Gouin et al., 2011	Fish	PE as 10% of diet	Modeled	1,2,3,4,5,6	Considered all known accumulation pathways in order to quantitatively assess the relative importance of plastic ingestion to total bioaccumulation. A worst case was considered by assuming concentrations in plastic and tissue to be at steady state. Provided mechanistic evidence based on first principles.
Koelmans et al., 2013	<i>A. Marina</i>	PS 0, 0.073, 0.73, 7.3 % d.w. in sediment. PE as 0, 0.1, 1, 10% d.w. in sediment	Modeled	1,2,3,4,5,6	Considered all known accumulation pathways in order to quantitatively assess the relative importance of plastic ingestion to total bioaccumulation. The 7.3% and 10% scenarios can be considered unrealistic or worst case. Provided mechanistic evidence based on first principles, as well as causal evidence on the treatment level by comparison with empirical data.
Koelmans et al., 2014	Lugworm, cod	Lugworm: continuous between 0 and 10% of diet. Cod: 2.34E-5 % of diet based on North Sea field data.	Modeled	1,2	Compared bioaccumulation due to plastic ingestion only, with total observed bioaccumulation in the field. Provided mechanistic evidence based on first principles.

Bakir et al., 2014	Lugworm, fish, seabird	1, 5, 50% of PVC or PE in diet	Modeled	1,2,3,4,5,6	Considered all known accumulation pathways in order to quantitatively assess the relative importance of plastic ingestion to total bioaccumulation. The 50% scenario can be considered unrealistic or worst case. Provided mechanistic evidence based on first principles.
Present study (Table SI-5)	<i>A. Marina</i>	5% d.w. PVC	Modeled	1,2	Simulated an experimental set up using spiked plastic and clean organisms. Provided mechanistic validation based on first principles, modeling and empirical data.
Present study (Table SI-5)	<i>Oryzias latipes</i>	10% d.w. LDPE	Modeled	1,2,6	Simulated an experimental set up that accounted for uptake from plastic and food. Neglected uptake from water. Provided mechanistic validation based on first principles, modeling and empirical data.
Present study (Table SI-5)	<i>Allorchestes compressa</i>	100 g/L, 100% PE	Modeled	1,2	Simulated an experimental set up using spiked plastic and clean organisms. Provided mechanistic validation based on first principles, modeling and empirical data.
<b>Field studies</b>					
Fossi et al, 2012	<i>Balaenoptera physalus</i>	0.00013 – 0.00094 #/L Corresponds to: 8.5 – 62 µg/L <sup>(c)</sup>	No	1,2,3,4,5,6 (no process can be ruled out)	Effect of microplastic ingestion on bioaccumulation of phthalates was speculated from detection of plastic in plankton samples, and phthalates detected in the same plankton samples and in fin whale. The plankton to

					microplastic number concentration ratio was 1600 (Ligurian sea) to 18000 (Sardinian sea). Given these ratios and aging of plastic in the oceans (Figure 2B) causing chemical desorption and uptake by plankton, process 5 (plankton pathway) may be more likely to occur than process 1 (plastic ingestion pathway).
Gassel et al., 2013	<i>Seriola lalandi</i>		Yes <sup>(d)</sup>	1,2,3,4,5,6 (no process can be ruled out)	Ingestion of plastic was speculated to best explain the detection of nonylphenol in 6 out of 19 fish individuals, given the detection of two plastic particles in two out of the 19 fish individuals. Given the data provided, the study shows that plastic may have been the source of the nonylphenol. However, fish (n=19) and plastic (n=2) sample sizes were very low and any of the simultaneous pathways 1 – 6 may have contributed to the uptake.
Rochman et al., 2014	<i>Myctophidae</i>	~ 14,000 – 200,000 items/km <sup>2</sup>	No	1,2,3,4,5,6 (no process can be ruled out)	Myctophid sampled at stations with greater plastic densities had larger concentrations of BDE#s 183 –209 in their tissues suggesting that these chemicals are indicative of plastic contamination in the marine environment. Plastic was not measured in the fish and the BDEs might as well have accumulated from water or the plankton diet. No strong conclusion on the role of ingestion was drawn.

Fossi et al, 2014	<i>Balaenoptera physalus</i> , <i>Cetorhinus maximus</i>	Not measured. Data from Fossi et al. (2012) were used. From the data provided it can be calculated that plastic made up 0.0262 % and 2.8 % of the plankton diet for the two species <sup>(c)</sup> .	No	1,2,3,4,5,6 (no process can be ruled out)	Effect of microplastic ingestion on bioaccumulation of phthalates was speculated from detection of phthalates in a zooplankton species and in the sharks and whales. Given the low plastic fractions in the diet and aging of plastic in the oceans (Figure 2B) causing chemical desorption and uptake by plankton, process 5 (plankton pathway) is more likely to occur than process 1 (plastic ingestion pathway).
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<sup>(a)</sup> Empirical studies may have concluded on the effect of plastic ingestion on transfer of HOCs without actually demonstrating that plastic has been ingested. ‘Yes’ means that plastic ingestion has been confirmed by detection of plastic in either the gastro-intestinal tract, body tissues or faeces.

<sup>(b)</sup> Processes explicitly accounted for in the design and interpretation of the study. Numbers relate to the processes summarized in **Figure 3** of the main paper. Numbers in parenthesis relate to processes likely to have occurred in the experiment yet that not have been specifically accounted for. For instance, use of spiked plastics leads to chemical desorption to water followed by dermal / body wall uptake. In order to separate the contributions of ingestion and dermal uptake to total bioaccumulation, the fluxes (i.e. mass accumulated per unit of surface area per unit of time) per separate pathway need to be assessed. This requires measurement of exposure concentration in water as well as in the ingested plastic and/or food.

<sup>(c)</sup> Highest estimate, based on (i) the reported number concentration (#/L) (ii) the definition of microplastic that specifies a maximum diameter of 5 mm, and (iii) a density of 1. Range relates to Sardinian sea (lowest) and Ligurian sea (highest).

<sup>(d)</sup> Two particles were detected in two out of 19 individuals, and the particles were presumed to be plastic. No IR or other polymer identification performed.

**Table S3.** Definition and units of model parameters for the plastic-inclusive bioaccumulation model (SI Eq. 1 and 2).

<b>Parameter</b>	<b>Definition and units</b>	<b><u>Lugworm</u></b>	<b><u>Japanese Medaka</u></b>	<b><u>Marine Amphipod</u></b>
		Simulation of experiments by Browne <i>et al.</i> , 2013.  <b>Value and source of model input.</b>	Simulation of experiments by Rochman <i>et al.</i> , 2013  <b>Value and source of model input.</b>	Simulation of experiments by Chua <i>et al.</i> , 2013  <b>Value and source of model input.</b>
$a_{\text{FOOD}}$	Absorption efficiency from food (-)	Only uptake through plastic was assessed $a_{\text{FOOD}}=0$	0.8 Hendriks <i>et al</i> , 2001	Only uptake through plastic was assessed $a_{\text{FOOD}}=0$
$C_{\text{B,t}}$	Chemical concentration in biota ( $\mu\text{g/g}$ )	SI Equation 1	SI Equation 1	SI Equation 1
$C_{\text{PL}}$	concentration in plastic ( $\mu\text{g/g}$ )	Browne <i>et al.</i> 2013 (SI)	Rochman <i>et al</i> , 2013 (SI) marine LPDE (ng/g)	0.5 Chua <i>et al</i> , 2014
$C_{\text{FOOD}}$	Concentration in food ( $\mu\text{g/g}$ ).	$C_{\text{FOOD}}=0$ . Only uptake through plastic was assessed	Rochman <i>et al</i> , 2013 (SI) marine plastic diet (ng/g)	$C_{\text{FOOD}}=0$ . Only uptake through plastic was assessed
$C_{\text{w}}$	Concentration in water ( $\mu\text{g/L}$ ).	$C_{\text{w}}=0$ . Only uptake through plastic was assessed	$C_{\text{w}}=0$ . Only uptake through plastic was assessed	$C_{\text{w}}=0$ . Only uptake through plastic was assessed
$dw$	Dry weight fraction (-)	0.146 Besseling <i>et al</i> , 2013	Estimated 0.15	0.23 Amanda A Fay <i>et al</i> , 2000
$f_{\text{lip}}$	Lipid fraction (-) of organism.	$0.052 \pm 0.012$ Hauck <i>et al</i> , 2007	0.043 Takuya Kundo <i>et al</i> , 2005	0.104 Amanda A Fay <i>et al</i> , 2000
$\text{GRT}_t$	Gut retention time (d)	0.135 (0.1 – 0.25) d Koelmans <i>et al</i> , 2013	1 day	12 h or 16 h Chua <i>et al</i> , 2014
$\text{IR}_t$	Food ingestion rate (g/g per day).	60 (47 – 74) g/g DW $\times$ d <sup>-1</sup> Browne <i>et al.</i> 2013	Fitted from food-only data	$0.006336$ g/g DW $\times$ d <sup>-1</sup> Harley <i>et al</i> , 1994
$k_{1\text{G}}$	constant for plastic to lipid	Fit	Fit	Fit

	transport ( $d^{-1}$ )			
$k_{2G}$	rate constant for lipid to plastic transport ( $d^{-1}$ )	Estimated as $k_{1G}/K_{PLIP}$ .	Estimated as $k_{1G}/K_{PLIP}$ .	Estimated as $k_{1G}/K_{PLIP}$ .
$k_{derm}$	Rate constant for uptake from water (L/g DW $d^{-1}$ )	$k_{derm} = 0$ . Only uptake through plastic was assessed	$k_{derm} = 0$ . Only uptake through plastic and food was assessed	$k_{derm} = 0$ . Only uptake through plastic was assessed
$k_{loss}$	Loss rate constant (g/g DW $d^{-1}$ )	Hendriks <i>et al</i> , 2001	Hendriks <i>et al</i> , 2001	Hendriks <i>et al</i> , 2001
$LogK_{OW}$	Octanol water partition coefficient (-)	Brooke and Thursby, 2005	Miller <i>et al</i> , 1985 and de Maagd <i>et al</i> , 1998	Braekvelt <i>et al</i> . 2002
$K_{LIP}$	Lipid-water partition coefficient (-)	Approximated as $K_{OW}$	Approximated as $K_{OW}$	Approximated as $K_{OW}$
$K_{PL}$	polyethylene - water partition coefficient (-)	Lohman <i>et al</i> , 2012 (SI)	Atkinson and Duffull, 1991	Atkinson and Duffull, 1991
$K_{PLIP}$	Lipid - plastic equilibrium partition coefficient (-)	Ratio between the lipid water partition coefficient ( $K_{LIP}$ ) and plastic - water partition coefficient ( $K_{PL}$ ).	Ratio between the lipid water partition coefficient ( $K_{LIP}$ ) and plastic - water partition coefficient ( $K_{PL}$ ).	Ratio between the lipid water partition coefficient ( $K_{LIP}$ ) and plastic - water partition coefficient ( $K_{PL}$ )
$S_{FOOD}$	Mass fraction of food ingested (-)	0.95 (5% plastic in sediment)	Set to one for the IR-calibration and set to 0.9 for the plastic-calibration	Term set to zero in the 'exposure-by-ingested-plastic-only' scenarios
$S_{PL}$	Mass fraction of plastic particles ingested (-)	0.05 (5% plastic in sediment) Browne <i>et al</i> , 2013	0.1 (10% plastic in diet) Rochman <i>et al</i> , 2013	1 (100% plastic)
W	Wet weight of organism (g).	4.0 g Browne <i>et al</i> , 2013	0.3 g Rochman <i>et al</i> , 2013	0.000476 g Amanda A Fay <i>et al</i> , 2000

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**Table S4:** Parameters for BDE209 leaching from HDPE to artificial gut fluids (fish oil, stomach oil), obtained by fitting SI Eq. 8 to data provided by Tanaka et al.<sup>12</sup> See SI Eq. 2 to 9 for detailed explanation of the calculation of these parameters.

	<b>Desorption rate constant <math>k_d</math> (<math>d^{-1}</math>)</b>	<b>Adsorption rate constant <math>k_u</math> (<math>d^{-1}</math>)</b>	<b>Plastic - gut fluid Equilibrium partitioning constant (<math>\text{Log}K_p</math>) (<math>L/kg</math>)</b>
<b>Fish oil</b>	0.81	1.16	3.16
<b>Stomach oil</b>	0.085	0.51	3.78



**Table S5:** Summary of HOC desorption rate constant  $k_{IG}$  ( $d^{-1}$ ) from plastic in the gut of marine biota, obtained from (a) bioaccumulation studies, (b) desorption studies, (c) first principles<sup>a)</sup>.

Value or range	Chemical/Plastic	Organism or condition	Plastic	Comment	Source
<b>Laboratory bioaccumulation studies (modeled with SI Eqs. 1 and 2)</b>					
4.1 – 9.8	Nonylphenol	Lugworm <i>A. marina</i>	PVC 230 $\mu$ m	Body wall - gut	This study, based on Browne et al 2013
0.76 – 1.6	Phenanthrene	Lugworm <i>A. marina</i>	PVC 230 $\mu$ m	Body wall - gut	This study, based on Browne et al 2013
2.5 → '> 50'	PBDE-47/PVC	Lugworm <i>A. marina</i>	PVC 230 $\mu$ m	Body wall - gut	This study, based on Browne et al 2013
0.073	PBDE-28, 47, 100, 99	Amphipod, <i>Allorchestes compressa</i>	PE 11-700 $\mu$ m	Chemicals at equilibrium only	This study, based on Chua et al, 2014
1.89	PCBs	Japanese Medaka. <i>Oryzias latipes</i>	PE < 500 $\mu$ m		This study, based on Rochman et al, 2013
1.16	PAH	Japanese Medaka. <i>Oryzias latipes</i>	PE < 500 $\mu$ m		This study, based on Rochman et al, 2013
<b>Desorption studies using artificial gut fluids</b>					
4.09	Phenanthrene	15.5 mM STC <sup>b</sup> 18°C	PE 200-250 $\mu$ m		Teuten et al, 2007
9.7	Phenanthrene	15.5 mM STC <sup>b</sup> 18°C	PP 200-250 $\mu$ m		Teuten et al, 2007
2.29	Phenanthrene	15.5 mM STC <sup>b</sup> 18°C	PVC 200-250 $\mu$ m		Teuten et al, 2007
2.3	Phenanthrene	15.5 mM STC <sup>b</sup> 18°C	PVC 130 $\mu$ m		Teuten et al, 2007
3	Phenanthrene	15 mM STC <sup>b</sup> 18°C, pH=7.5-8.4	PE		Bakir et al, 2014
1.68	DDT	15 mM STC <sup>b</sup> 18°C, pH=7.5-8.4	PE		Bakir et al, 2014
0.27	DEHP	15 mM STC <sup>b</sup> 18°C, pH=7.5-8.4	PE		Bakir et al, 2014
12.10	Phenanthrene	15 mM STC <sup>b</sup> 38°C, pH=4	PE		Bakir et al, 2014
7.2	DDT	15 mM STC <sup>b</sup> 38°C, pH=4	PE		Bakir et al, 2014
3.89	DEHP	15 mM STC <sup>b</sup> 38°C, pH=4	PE		Bakir et al, 2014

1.67	Phenanthrene	15 mM STC <sup>b</sup> 18°C, pH=7.5-8.4	PVC		Bakir et al, 2014
0.31	DDT	15 mM STC <sup>b</sup> 18°C, pH=7.5-8.4	PVC		Bakir et al, 2014
1.37	DEHP	15 mM STC <sup>b</sup> 18°C, pH=7.5-8.4	PVC		Bakir et al, 2014
4.8	Phenanthrene	15 mM STC <sup>a</sup> 38°C, pH=4	PVC		Bakir et al, 2014
0.54	DDT	15 mM STC <sup>b</sup> 38°C, pH=4	PVC		Bakir et al, 2014
4.86	DEHP	15 mM STC <sup>b</sup> 38°C, pH=4	PVC		Bakir et al, 2014
0.81	BDE209	Fish oil, 38°C	HDPE	See Table SI-4, calculated with SI Eqs. 3 to 9.	<i>This study</i>
0.085	BDE209	Stomach oil, 38°C	HDPE		
<b><i>Calculated from first principles<sup>c</sup></i></b>					
0.1	PCB	Polymer diffusion	1.3 mm PS	Besseling et al, 2013	Koelmans et al, 2013
2	PCB	Polymer diffusion	0.4 mm PS	Besseling et al, 2013	Koelmans et al, 2013
>100	PCB	Polymer diffusion	0.1 – 1 µm PS	Besseling et al, 2013	Koelmans et al, 2013
0.9 - 9.8	PCB (28, 52, 101, 153, 180)	Polymer diffusion	PE 0.5 mm	As used by Rochman 2013 (< 0.5 mm)	This study
1.67 - 53	PAH (BAA, CHRYS, PHEN, ANT, FLU)	Polymer diffusion	PE 0.5 mm	As used by Rochman 2013 (< 0.5 mm)	This study

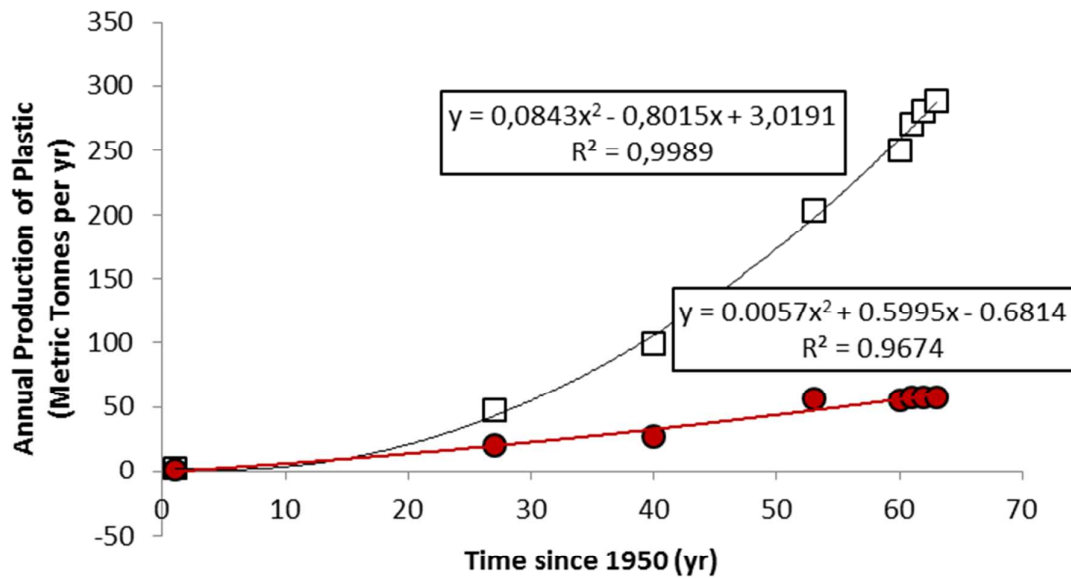
<sup>a</sup> The 10-90% inter quantile range  $PR_{10-90\%}$  for all data is 0.3 – 9.8 d<sup>-1</sup> with a median of 2.1 d<sup>-1</sup>. For the separate categories of studies,  $PR_{10-90\%}$  overlaps with ranges of 0.6 – 17.8; 0.3 – 8.0; and 0.7 – 27.8 for modeling studies, desorption studies and first principle calculations, respectively. The ranges are not affected by the uncertainty in the outlying values (indicated with '>').

<sup>b</sup> Sodium taurocholate

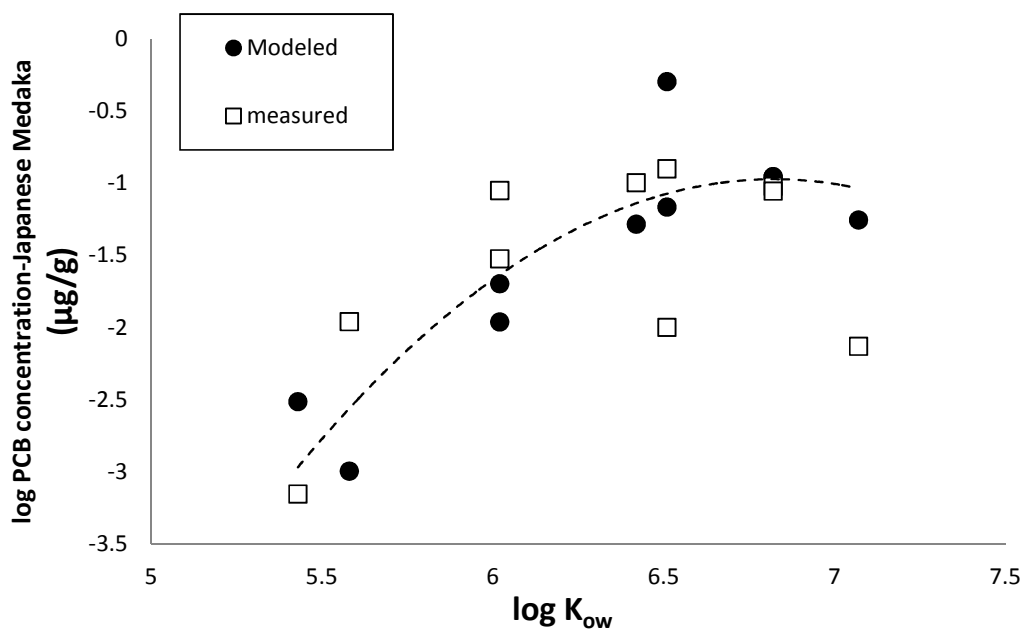
<sup>c</sup> First order rate constants based on  $k_{1G} \approx 23D/r^2$  with D is the polymer diffusion coefficient and r is the (average) radius of the MP particle.

Koelmans, A.A., E. Besseling, A. Wegner, E.M. Foekema. (2013). Plastic as a carrier of POPs to aquatic organisms. A model analysis. *Environ. Sci. Technol.* 47, 7812–7820.

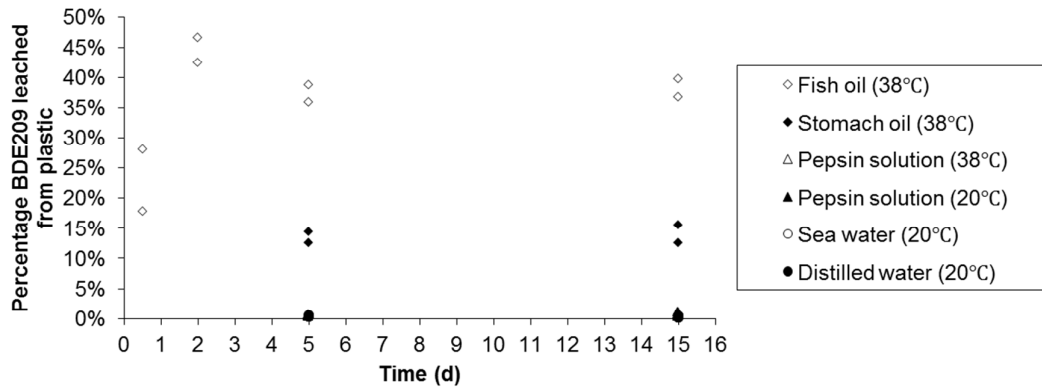
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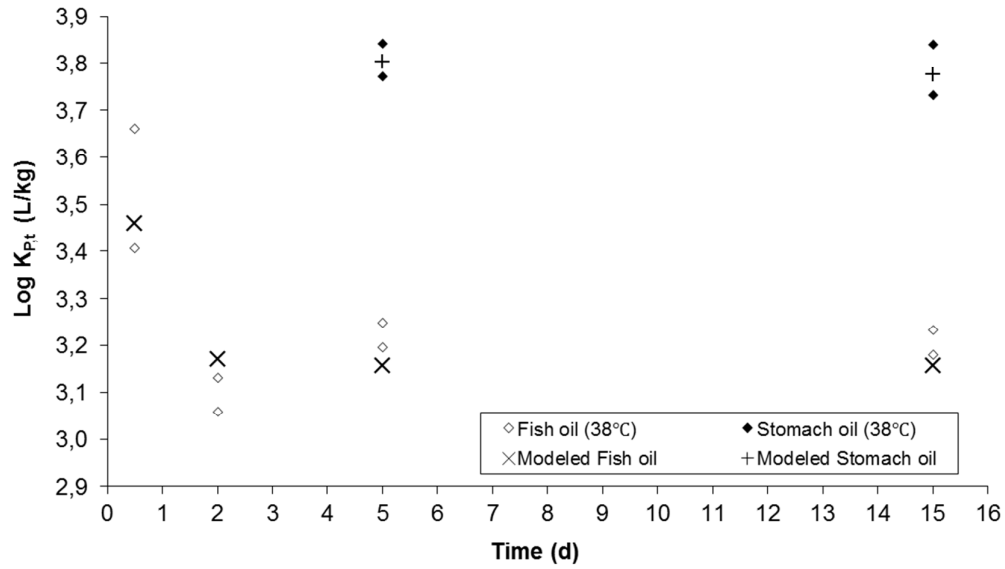
**Figure S1.** World annual production of plastic as a function of time since 1950, smoothed using a 2<sup>nd</sup> order polynomial (-□-). Red curve and markers (-●-) relate to plastic production from Europe.



**Figure S2.** Bioaccumulation of PCB by Japanese medaka from PCB contaminated plastic and food. **Error! Bookmark not defined.** The modelled data points were obtained using a bioaccumulation model that includes plastic as a component of the diet, **Error! Bookmark not defined.** with a fitted PCB desorption rate constant of  $1.89 \text{ d}^{-1}$  (this study).



**Figure S3.** Percentage BDE209 leached from plastic to various liquids. Fish and stomach oil at °C were taken to represent gut fluids of seabirds. Figure drawn based on original data<sup>12</sup> provided by Hideshige Takada.



**Figure S4.** Ratios of the concentration of BDE209 in plastic and in artificial gut fluids ( $K_{P,t}$ ) over time, as calculated from the data provided by Tanaka et al<sup>12</sup> (Figure S1) using SI Eq. 5, for Fish oil and Stomach oil at 38 °C, together with modeled  $K_{P,t}$  values (SI Eq. 8).

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