Accumulation of Plastic Debris and Associated Contaminants in Aquatic Food Webs

Noël J. Diepens†‡* and Albert A. Koelmans†‡

†Aquatic Ecology and Water Quality Management Group, Department of Environmental Sciences, Wageningen University, P.O. Box 47, 6700 AA Wageningen, The Netherlands
‡Wageningen Marine Research, P.O. Box 68, 1970 AB IJmuiden, The Netherlands

Supporting Information

ABSTRACT: We present a generic theoretical model (MICROWEB) that simulates the transfer of microplastics and hydrophobic organic chemicals (HOCs) in food webs. We implemented the model for an Arctic case comprised of nine species including Atlantic cod and polar bear as top predator. We used the model to examine the effect of plastic ingestion on trophic transfer of microplastics and persistent HOCs (PCBs) and metabolizable HOCs (PAHs), spanning a wide range of hydrophobicities. In a scenario where HOCs in plastic and water are in equilibrium, PCBs biomagnify less when more microplastic is ingested, because PCBs biomagnify less well from ingested plastic than from regular food. In contrast, PAHs biomagnify more when more microplastic is ingested, because plastic reduces the fraction of PAHs available for metabolization. We also explore nonequilibrium scenarios representative of additives that are leaching out, as well as sorbing HOCs, quantitatively showing how the above trends are strengthened and weakened, respectively. The observed patterns were not very sensitive to modifications in the structure of the food web. The model can be used as a tool to assess prospective risks of exposure to microplastics and complex HOC mixtures for any food web, including those with relevance for human health.

INTRODUCTION

Plastic debris particles and hydrophobic organic chemicals (HOCs) are widely distributed in the aquatic environment1−7 and even found in remote areas such as the Arctic.8−11 The role of plastic particles as a potential vector for HOCs has received considerable attention in the recent literature.4,12−15 Although some classes of HOCs are known to biomagnify in marine food webs,16,17 the extent to which plastic debris accumulates in food webs remains unclear. Several studies showed that microplastics are readily ingested by marine organisms18−26 or that microplastics are passed on to predators in sequential dietary uptake studies.13,27−29 However, to date no studies address bioaccumulation of plastic on the level of real food webs; not from a theoretical perspective, nor in model ecosystem experiments or in food webs occurring in nature. Because of the complexity of food webs and the practical difficulty of mimicking food webs in model ecosystem experiments, prospective theoretical models constitute valuable tools to explore transfer of plastic debris in food webs, including those with relevance for human health.

Due to the high complexity of food webs and that of plastic particle and chemical behavior, the effect of food web transfer of plastic debris on the bioaccumulation and biomagnification of HOCs is difficult to predict. Until now, laboratory experiments have studied the effect of microplastic on HOC uptake on the level of individual species in simplified laboratory settings that mainly addressed HOC uptake from microplastic by clean animals (e.g. refs 30−32). Plastic-inclusive models have been applied to translate inferences from such studies to more realistic natural conditions, where for instance multiple HOC uptake pathways exist or where organisms often already are contaminated with HOCs from other sources than from microplastic.4,12,14,33,34 Until now these models were restricted to HOC transfer across one trophic level while assuming a linear and nonflexible food chain. No models exist that simulate transfer of plastic and HOCs in entire aquatic food webs, along multiple trophic levels, while accounting for multiple feeding relationships and taking all categories of plastic-chemical interactions into account.4,33 We argue that such models are urgently needed in order to provide assessments of complex plastic-HOC interactions in food webs. Simulations with such models can drive experimental or field study designs and can be used for prospective risk assessments.

Several exposure scenarios and classes of HOCs could be particularly relevant for the risks of plastic-associated chemicals in food webs. Whereas experimental and modeling studies have shown that microplastic ingestion is likely to play a minor role in the transfer of persistent global POPs and legacy compounds under natural conditions,4,12,14,15,30,33,35−45 this may be different for additives that are unique for plastic, that is, are not present in surrounding media, or for metabolizable chemicals. For such “unique additives”, plastic is the only source, maximizing the gradient for transfer and rendering other uptake pathways less...
important. Metabolizable chemicals would not be degraded when bound to plastics. These chemicals thus are preserved in ingested plastics, which maximizes the chance of transfer in the food chain. For such chemicals, the extent of food web transfer urgently needs to be assessed.4,12,13

The aim of the present work was to study the food web accumulation of plastic debris and associated contaminants from a theoretical perspective, using an integrated microplastic and contaminant food web accumulation model (MICROWEB). We postulate a set of equations that can be used as a tool for prospective risk assessments, to generate hypotheses that can be verified experimentally and that can be used to trigger the further scientific evolution of risk assessment frameworks for plastic debris. The model was implemented for an illustrative case: an Arctic food web that includes among others Atlantic cod and polar bear as top predator. Atlantic cod has a high ecological and economical value and is a central part of the Arctic food web. Moreover, cod is a key species to sustain populations of e.g. sharks, sea birds, and mammals such as seals and polar bears and is an important component of the human diet. Various studies report on microplastic presence in cod.22,24,25,46 We performed scenario analyses to investigate the effect of (a) increasing share of plastic in the organisms’ diet, (b) equilibrium versus nonequilibrium states for the dissolved and plastic-bound HOCs, and (c) chemical metabolism on food web accumulation of plastic and contaminants, as expected from existing knowledge and theory.

METHODS: AN INTEGRATED MICROPLASTIC AND CONTAMINANT FOOD WEB ACCUMULATION MODEL (MICROWEB)

The MICROWEB model predicts the steady state plastic and HOC concentration in biota for each species of a predefined food web. Bioaccumulation of HOCs and microplastic by biota can be modeled as a balance of uptake and loss processes. It is assumed that bioaccumulation of HOCs is affected by the bioaccumulation of plastic but not the other way around. Therefore, the model has two separate components: one that calculates the food web transfer of microplastic and one that uses this food web transfer of microplastic to infer the bioaccumulation and magnification of HOCs. The food web is defined by the feeding relationships reflected by diet components and their associated fractions. Below we provide the main features and equations of the model. A full description, schematic overview of main model processes (Figure S1), and list with all parameters (Table S1) are provided as Supporting Information (SI) (Section S1).

Food Web Accumulation of Plastic. Plastic uptake can be modeled as a mass balance of ingestion and loss processes, as was described before by Besseling et al.23 and Herzke et al.47 Here, their equations for a single diet component are extended to a generic form capable of accommodating uptake from multiple diet components in a food web. Uptake is restricted to the gastrointestinal tract, that is, transfer to the body tissues as has been hypothesized to play a role for particles smaller than 200 nm,48,49 which is not included in the present version of the model. The concentration of plastic in biota \( C_{PL,i} \) (g plastic X g\(^{-1}\) biota) can be modeled as

\[
\frac{dC_{PL,i}}{dt} = \text{IR}_i \sum_{j=1}^{n} (p_j C_{PL,j}) - k_{\text{loss,PL}} C_{PL,i}
\]  

The first term accounts for predator ‘i’ feeding on ‘n’ multiple food items ‘j’ with a predator species specific ingestion rate \( \text{IR}_i \). Following principles of chemical bioaccumulation modeling, a predator has ‘n’ food preferences ‘\( p_j \)’ \((0 < p < 1 \text{ and } \sum_j p_j = 1)\), which determine the fraction of each food item in the total diet, each of which in turn has a concentration of plastic \( C_{PL,j} \) (g plastic X g\(^{-1}\) biota). In the second term, \( k_{\text{loss,PL}} \) (d\(^{-1}\)) is the loss rate constant of plastic via egestion, and ‘t’ is time. The steady state solution to eq 1 results in the plastic concentration in species ‘i’:

\[
C_{PL,i} = \frac{\text{IR}_i \sum_{j=1}^{n} p_j (C_{PL,j})}{k_{\text{loss,PL}}}
\]  \[ (2) \]

The first order loss rate constant can be calculated as the reciprocal of the gut retention time \( (\text{GRT}_i, d) \) of the organism

\[
k_{\text{loss,PL}} = \frac{1}{\text{GRT}_i}
\]  \[ (3) \]

which implies that eq 2 can be given in an alternative form:

\[
C_{PL,i} = \text{IR}_i \times \text{GRT}_i \sum_{j=1}^{n} (p_j C_{PL,j})
\]  \[ (4) \]

We thus can model plastic uptake by an organism, once food intake rates, diet composition, plastic fraction per diet component, and either plastic loss rates (eq 2) or plastic gut retention times (eq 4) are available. In the case of a food web, implementations of eqs 2 or 4 for each of the individual species in the web are linked to each other such that ingestion rates and diet composition match the empirical data known for that food web.

The mass fraction of plastic in an organism \( (S_{PL,i}) \) can be calculated as

\[
S_{PL,i} = \frac{M_{PL,i}}{(M_{PL,i} + M_B)}
\]  \[ (5) \]

which is mathematically equivalent to \( S_{PL,i} = C_{PL,i}/(1+C_{PL,i}) \). Here, \( M_{PL,i} \) is the mass of plastic in the organism, and \( M_B \) is the mass of the organism.

Plastic-Inclusive Food Web Accumulation Modeling of HOCs. The chemical concentration in biota \( (C_{HOC,i} \text{ g g}^{-1} \text{ ww}) \) in the food web is modeled based on an earlier plastic-inclusive bioaccumulation model as introduced by Koelmans et al.33

\[
\frac{dC_{HOC,i}}{dt} = k_{\text{damp}} C_u + \text{IR}_i \sum_{j=1}^{n} (S_{FOOD,j} S_{FOOD,j} C_{HOC,j} + S_{PL,j} C_{PL,j}) - k_{\text{loss,HOC}} C_{HOC,i}
\]  \[ (6) \]

This equation quantifies how predator ‘i’ acquires HOCs from ambient water, ‘j’ prey species and/or abiotic food components.

In the first term, uptake of HOCs through dermal uptake from water is quantified with \( k_{\text{damp}} \) (L X g\(^{-1}\) X d\(^{-1}\)) the dermal absorption rate constant from water and \( C_u \) (µg L\(^{-1}\)) the HOC concentration in the water. In the second term, uptake from all ingested food components in the diet \((j = 1 \text{ to } n)\) is quantified. In the third term, loss of HOCs is quantified using an overall loss rate parameter \( k_{\text{loss,HOC}} \) (d\(^{-1}\)), accounting for elimination to water, elimination by feces, growth dilution, and metabolism.48,49 A detailed further explanation of all terms in eq 6 is provided as Supporting Information.
Here we specifically explain the second term of eq 6, which accounts for the uptake of HOCs from ingested food and plastic. Like for the plastic accumulation submodel (eqs 1−4), the predator has a preference $p_j$ for each of the food components ($0 < p_j < 1$ and $\sum p_j = 1$). However, each food component has a fraction of digestible food ($S_{\text{FOOD}}$) and a fraction of plastic ($S_{\text{PL}}$), with $S_{\text{FOOD}} + S_{\text{PL}} = 1$ and $\sum p_j S_{\text{FOOD}} + S_{\text{PL}} = 1$. Species can also ingest pure plastic either by accident e.g. when mistaking plastic debris for a food item\(^5\) or unintentionally with the water they ingest. When only pure plastic is ingested, $S_{\text{FOOD}}$ is zero and $S_{\text{PL}} = 1$. Hence, the mass fractions $p_j S_{\text{FOOD}}$ and $S_{\text{PL}}$ partition the total mass ingested per unit of time (i.e., the ingestion rate $IR_j$) into ingested mass for each of the ‘$j$’ food components. The HOC concentration absorbed from each food component ‘$j$’ is quantified by $a_{\text{FOOD}} C_{\text{HOC,j}}$ with $C_{\text{HOC,j}} (\mu g \times g^{-1} \text{ww})$ the chemical concentration and $a_{\text{FOOD,j}}$ the chemical absorption efficiency for component ‘$j$’. In contrast to chemical absorption from food, which is driven by digestion of the food, chemical uptake from the ingested plastic is assumed to depend on the concentration gradient between plastic and biota lipids. Following Koelmans et al.,\(^33,35\) the HOC concentration absorbed by species ‘$i$’ from the plastic present in the ingested food is dynamically modeled as $C_{\text{PL,j}} (\mu g \times g^{-1})$

$$C_{\text{PL,j}} = \frac{k_{1,j} C_{\text{LIP,j}} - k_2 C_{\text{LIP,j}}}{k_1 + \frac{M_{\text{PL}}}{M_{\text{lip}}} k_2}(1 - e^{-(k_1 + (M_{\text{lip}}/M_{\text{PL}})k_2)GRT,i})$$

(7)

in which $k_1 (d^{-1})$ and $k_2 (d^{-1})$ are first order rate constants for the transport from plastic to biota lipids and vice versa (assumed to depend on plastic size\(^33\)), $C_{\text{PL,j}} (\mu g \times g^{-1})$ is the average HOC concentration in all ingested plastic from all ingested food items at the moment of ingestion by predator ‘$i$’, and $C_{\text{LIP,j}} (\mu g \times g^{-1})$ is the concentration in the lipids of predator ‘$i$’ ($C_{\text{LIP,j}} = C_{\text{LIP,j}}^{\text{f,lip}}$, with $f_{\text{LIP,j}}$ being the lipid fraction of predator ‘$i$’), $M_{\text{PL}}$ is the total ingested plastic mass, and $M_{\text{lip}}$ is the lipid mass in the predator. For convenience, eq 7 can be rewritten in a simple form\(^35\)

$$C_{\text{PL,j}} = A_{\text{PL}} k_j C_{\text{PL,j}}^{\text{f,lip}} - A_{\text{PL}} k_2 C_{\text{LIP,j}}^{\text{f,lip}}$$

(8)

with

$$A_{\text{PL}} = 1 - e^{-(k_1 + (M_{\text{lip}}/M_{\text{PL}})k_2)GRT}$$

(9)

The variable $C_{\text{PL,j}}$ in eqs 7 and 8 represents the average HOC concentration in all ingested plastic originating from all ingested prey species or food items ‘$j$’ at the moment of ingestion by predator ‘$i$’. The chemical concentration in plastic in an ingested prey species ‘$j$’ depends on the time ($t < GRT$) that plastic has spent in the gut of prey species ‘$j$’ since the moment of ingestion by predator ‘$i$’. Because in a natural food web where organisms can be assumed to randomly ingest plastics, all time points $t$ with $t < GRT$, are equally likely, an average HOC concentration in plastic is calculated for each prey species ‘$j$’ by

$$C_{\text{PL,j}} = \frac{C_{\text{PL,j}} + k_2 C_{\text{LIP,j}} - k_1 C_{\text{PL,j}}^{\text{f,lip}}}{k_1 + \frac{M_{\text{PL}}}{M_{\text{lip}}} k_2}$$

(10)

Eq 10 provides the average chemical concentration in plastic present in a species, dependent on the concentration upon ingestion, kinetic constants, the plastic/lipid mass ratio, and the gut retention time of the species. A detailed explanation of eq 10 is provided as Supporting Information (SI Section S1).

**Averaging Chemical Concentrations in Plastic in the Food Items of the Diet.** Over time, the predator ingests a mixture of ‘$n$’ food items, each having an individual $C_{\text{PL,j}}$ (from eq 10). These individual $C_{\text{PL,j}}$ values are used to calculate one average value with the mass fractions of the ingested prey species as weighting factors. Our rationale for this approach is as follows. First, it is likely that chemical transfer among the plastic particles originating from different prey species ‘$j$’ in the gut of predator ‘$i$’ is faster than the lipids of the predator. After all, the plastics are mixed together in the same gut fluid with surfactants and micelles enhancing chemical solubility and exchange,\(^33\) whereas transport to lipids requires transfer across multiple cell membranes and layers.\(^12,50\) This would effectively lead to fast equilibration among the plastic particles in the gut, leading to one average concentration with masses of contributing plastics as weighting factors. Second, even if this transport were to be slow, a weighted average would best represent the net effect of all individual transfers from ingested plastics with their individual $C_{\text{PL,j}}$ values to the lipids of the predator. Consequently, the effective HOC concentration in the total mass of plastic ingested by predator ‘$i$’ (\$C_{\text{PL,i}}\$), which is required as input for eqs 7−9, is calculated as

$$\bar{C}_{\text{PL,i}} = \sum_{j=1}^{n} p_j S_{\text{PL,j}} C_{\text{PL,j}}$$

(11)

As our model application does not require that time trends in HOC and plastic abundance exposures are simulated, a steady state solution of eq 6 is used ($d C_{\text{HOC,i}} / dt = 0$), as was argued before by Koelmans et al.\(^34,35\) The model needs HOC concentrations in water and in free floating plastics as input variables. Depending on the exposure scenario to be modeled, (part of) these values can either be taken from measurements, can be set to zero, or can be inferred from partitioning relationships such as the plastic-water partitioning coefficient (\$K_{\text{PL,j}} = L \times kg^{-1})\$.

Combination of the above eqs 6−9, 10, and 11 leads to the following steady state solution for bioaccumulation of HOCs by a species from multiple food items containing microplastics:

$$C_{\text{HOC,i}} = \frac{k_{\text{i,m}} C_{\text{w}} + IR_{\text{i}} \sum_{j=1}^{n} p_j (S_{\text{FOOD,j}} A_{\text{FOOD,j}} C_{\text{HOC,j}} + S_{\text{PL,j}} A_{\text{PL,j}} k_j C_{\text{PL,j}})}{IR_{\text{i}} \sum_{j=1}^{n} p_j (S_{\text{PL,j}} A_{\text{PL,j}} k_j C_{\text{PL,j}})} + k_{\text{i,m}}$$

(12)

By using the outcomes of eq 12 for each of the species, as input to all other species that consume them, transfer of microplastic and HOCs across trophic levels can be simulated. As a result of this linking, subscripts ‘$i$’ apply to a species as long as it acts as a consumer in the web, whereas the same species receives.
Food Web Accumulation of Microplastic and Associated PCBs and PAHs in an Arctic Food Web

We applied the MICROWEB model to simulate transfer of microplastic along a marine Arctic food web (Figure S2). This food web is defined using biological data previously reported by De Laender et al.70 and reaches up to the polar bear as top predator. This food web can be seen as a spatiotemporal average of a sub-Arctic and an Arctic food web and is suggested to become more relevant as a result of climate change.53 Five trophic levels are included: phytoplankton (TL = 1), zooplankton (TL = 2), polar arctic cod (Boreogadus saida) (TL = 3.4) hereafter referred to as polar cod, Northern shrimp (Pandalus borealis) (TL = 3.5), capelin (Mallotus villosus) (TL = 3.5), Atlantic herring (Clupea harengus) (TL = 3.6), Atlantic cod (Gadus morhua) (TL = 3.7), seal (Phoca spp.) (TL = 4.0), and polar bear (Ursus maritimus) (TL = 5.1) (Figure S2, Tables S2–S5). Phytoplankton as well as zooplankton were modeled as functional groups, without further speciation within the groups. Characteristics for the functional group “seal” (Phoca spp.) were based on combined data for Harbour, Ringed, and Harp seal. Trophic levels were obtained from the literature describing Arctic food webs and a fish database71 (Table S3). To illustrate the sensitivity of the model output to food web structure, also a fully Arctic food web was modeled, omitting the sub-Arctic species, which assumes a linear feeding structure of phytoplankton, zooplankton, polar cod, seal, and polar bear.55,56

Ten PCBs (CB 2, 8, 28, 52, 101, 118, 153, 180, 206, 209) and ten PAHs [acenaphthene (Ace), anthracene (Ant), phenanthrene (Phe), pyrene (Pyr), fluoranthene (Flu), benzo[a]-anthracene (BaA), benzo[b]fluoranthene (BbF), benzo[g,h,i]-perylene (BghiPer), indeno[1,2,3-cd]pyrene (Ind123P), and coronene (Cor)] were modeled, spanning a wide range of logK_{ow} values (from 4.60 to 8.27 for PCBs and from 3.98 to 7.64 for PAHs; Tables S6, S7).

Model Parameters. Most biological parameters were taken from the literature, and references therein (Tables S2–S5). Ingestion rate and GRT were based on literature values (Table S4). When more than one value for a parameter was found, the average (in case of 2 or 3 data points) or the geometric mean (in case of >3 data points) was used, and in all cases the range is provided (Table S4). Sometimes unit conversions were required (Table S4). Because for capelin no value for the GRT could be found, we used the value for Atlantic herring, which is a species similar in weight. For phytoplankton, chemical uptake and elimination occur via transfer across cell walls; they do not “ingest” particles. The plastic water partition coefficients (K_{PL}) for high density polyethylene (HDPE) were used, as provided by Endo and Koelmans.56

Chemical concentrations in Arctic water were available for five PCBs (9.23 × 10^{-8}–2.13 × 10^{-7} μg L^{-1}) and seven PAHs (9.11 × 10^{-8}–2.37 × 10^{-5} μg L^{-1}).57,58 For concentrations in plastic, global concentration data were available for nine PCBs (0.079–1.08 μg kg^{-1}) and ten PAHs (0.179–23.8 μg kg^{-1}) (Tables S6, S7). For HOC concentration in plastic, geometric means were used, and concentrations in water were calculated with K_{PL}. Negative values were omitted, and zero values were replaced by 50% of the detection limit or the minimal value reported, in case a detection limit was not reported (e.g., as in Hiri et al.59). When concentration data for chemicals were not available, values from the chemicals with similar logK_{ow} were used (e.g., for PCB 2 the value for PCB 8 was used).

Metabolization rates for PCBs are known to be small and therefore were assumed negligible.60 For PAHs, metabolization rates for phytoplankton also were assumed to be negligible. PAH metabolization rates for zooplankton and shrimp were calculated according to Berrojalbiz et al.,65 and for fish and mammals PAH metabolization rates were calculated according to Arnot et al.66 and Moermond et al.67 (see eq S38 for details).

Scenarios. Various scenarios were simulated in order to answer our research questions regarding trophic transfer of microplastics and associated HOCs. The scenarios are briefly described below.

Trophic Transfer of Microplastic as a Function of Ingested Plastic Mass. Concentrations of microplastics in all the components of the food web were modeled for scenarios with 0, 1, 3, 10, 30, and 99% microplastic in the ingested diet. The 99% scenario complies to the high plastic-biomass ratios reported recently by Chen et al.61 as well as to possibly high doses of microplastics that would occur in hot spots as emissions of plastic to the oceans increase in the future.62 Using 99% or 100% was arbitrary; there were no discernible differences between model outputs for these percentages.

Effects of Microplastic on Bioaccumulation of PCBs and PAH. Two types of scenario calculations were used to investigate the effect of plastic on bioaccumulation of HOCs, for two contrasting groups of HOCs: PCBs, with zero or low metabolism, and for PAHs, with more substantial metabolization. First, the effects of increasing plastic abundance were assessed by varying the fraction of microplastic in the organisms’ diets between 0 and 99%, assuming chemical equilibrium between microplastic and seawater. The plausibility of sorption (near-)equilibrium for microplastics has been motivated before55,61,69 and complies to the common equilibrium partitioning approach in chemical risk assessment of contaminates.
nated solids.\textsuperscript{70,71} The age of microplastics in the oceans is at least several years,\textsuperscript{4} whereas (de)sorption times scales are less.\textsuperscript{72} Second, this sorption equilibrium scenario was compared to scenarios with chemical concentrations in microplastic at higher as well as at lower than their aqueous phase equilibrium concentrations, to assess implications of the nonequilibrium behavior of additives and sorbing chemicals, respectively.

**Sensitivity Analyses.** A local sensitivity analysis was performed using the “Morris method”\textsuperscript{73} for the parameters: $K_{PL}$, $K_{W}$, $K_{M}$, GRT, $f_{LIP}$, and IR. These default scenario parameter values were varied by ±10%, and the effects in predicted chemical concentrations in each species’ lipids were recorded.

### RESULTS AND DISCUSSION

**Accumulation of Microplastic in an Arctic Food Web.** 
Steady state microplastic concentrations vary with trophic level and with abundance of microplastic in the diet (Figure 1A). Concentrations are lowest for Atlantic herring and highest for seal. This trend among species in the food web is the same for all ingested plastic fractions (1 to 99%). Our theoretical calculation shows that concentrations can reach high levels, i.e. approaching 1 g × g\textsuperscript{-1} biota for seal, when very high plastic to biomass ratios would be present in their habitat and in habitats of the species they feed on. When the fraction of plastic in the diet exceeds 30%, the resultant microplastic mass fraction exceeds the lipid fraction for zooplankton and Atlantic cod. At 99% this is also the case for the Northern shrimp, polar cod, and seal.

For all scenarios with plastic ingestion, BMFs were higher than 1 for Atlantic cod and seal indicating accumulation between these specific predators and their preys (Table S12). BMFs were highest for seals, up to 8.0. This is explained from the relatively high ingestion rate and gut residence time of the seals. Data compiled by Bakir et al.\textsuperscript{12} also showed the highest plastic content in the stomach for seals. Interestingly, the BMF for polar bear is almost negligible (<0.1).

TMF values are below one and decreased from 0.88 to 0.75 when the fraction of plastic in the diets is increased from 1 to 99% (Figure 1B). This indicates that on the level of the entire food chain, trophic dilution rather than magnification of microplastic concentrations is expected. We performed additional calculations where microplastic was only included in the diet of zooplankton, as an entry point at the base of the food chain. This calculation thus shows the transfer of microplastic when no parallel uptake of pure microplastic at the higher trophic levels would occur. This more pure measure of biomagnification yielded TMF values below 0.05 (Figure S5D), clearly indicating that theory predicts trophic dilution and not biomagnification.

The results from these microplastic scenarios can be understood by the mechanisms of microplastic uptake. Whereas for POPs, biomagnification involves the digestion of food, leading to higher than equilibrium fugacity and transfer to lipid reservoirs,\textsuperscript{74} concentrations for microplastics are thought to merely reflect the balance between ingestion, gut retention, and egestion. This does not imply that magnification cannot exist between specific combinations of predator and prey, as was demonstrated here most clearly for seals. However, using biologically realistic ingestion and egestion data for the species expected in this Arctic food web revealed that no magnification occurs on the level of the entire food chain.

We performed additional calculations where the food web was defined to represent a fully Arctic food web, omitting the sub-

---

**Figure 1.** Concentration of plastic in biota (g plastic × g\textsuperscript{-1} biota) (A) and the trophic magnification factor (TMF) (B) for percentages of microplastic in the diet of 0, 1, 3, 10, and 99% by species in an Arctic food web.
Arctic species. For this web, very similar levels of accumulation of microplastic were obtained (Figure S6A). Food web accumulation of microplastic thus is not very sensitive to food web structure, which is explained by the same mechanisms of microplastic uptake mentioned above.

**Effects of Microplastic on Trophic Transfer and Biomagnification of HOCs.** Accumulation of HOCs in an Arctic Food Web Contaminated with Microplastics. Like for the microplastic concentrations (Figure 1), steady state HOC concentrations vary with trophic level and with abundance of microplastic in the diet (Figure 2 “Equilibrium” scenario, Figure S4, Tables S13, S14). In the absence of microplastic (0% plastic), PCB concentrations in biota lipids increase along the chain for all species in Figure S4B, S4E, Table S13. The model thus predicts biomagnification of PCBs, just like traditional bioaccumulations models do. When higher percentages of microplastic are present in the diet and assuming chemical equilibrium between water and ingested microplastic, this biomagnification is attenuated until it is fully absent at the highest simulated plastic concentrations (99%) (Figure 2A, 2B, 2C “Equilibrium” scenario, Figure S4B, S4E). The explanation for this “cleaning” mechanism has been provided in earlier studies and is 2-fold. First, biomagnified PCBs in biota have a higher fugacity than those in ingested microplastics, leading to overall less magnification when more plastic is present. Second, PCBs are rapidly biomagnified from prey by digestion, whereas they are only absorbed from microplastics by retarded polymer diffusion, which is far less efficient. This difference between the bioavailability of HOCs in natural organic matter and polymers has been experimentally verified by Beckingham and Ghosh. Hence, replacing part of the diet by microplastics thus results in lower biomagnification.

Several recent studies provide support for the approach and trends predicted here. The model has been applied to laboratory bioaccumulation data, and fitted model parameters were consistent with predictions based on polymer diffusion first-principles and kinetic experimental data. A recent experimental study reported less uptake of PCBs by lobster in the presence of clean microplastics, which is consistent with the PCB...
simulations presented. Seemingly in contrast, another recent study could not detect a “cleaning” effect for PCBs when 40% of PE microplastics was included in the diet of rainbow trout. However, confidence intervals in the rate constants spanned 1–2 orders of magnitude, probably preventing the statistical rigor required to detect the effect.

Interestingly, model results for PAHs show trends opposite to those obtained for PCBs (Figure 2D, 2E, 2F “Equilibrium” scenario, Figure S4C, S4F; Table S14). Without microplastics in the diet, PAH concentrations generally decrease, a process referred to as ‘biodilution’. For all species except for phytoplankton, increasing the fraction of ingested microplastic increases bioaccumulation of PAHs up to 1 order of magnitude. As the hydrophobicity range largely overlaps with those for the PCBs and modeling principles are the same except for the metabolization which for PAHs is included, the latter process explains the difference. Metabolization causes the fugacities in lipids to be lower than those in ingested plastic, causing transfer of PAHs to biota lipids. When more microplastic is ingested, a higher proportion of ingested PAHs is transferred via the plastic, whereas a smaller proportion of PAHs is available for metabolism. Phytoplankton was assumed not to be able to metabolize PAHs substantially; therefore concentrations were higher compared to the other species.

The patterns described above are also reflected in the bioaccumulation metrics BMF and TMF. For PCB congeners, BMF values ranged between 0.9 and 170 without microplastic in the diet and between 0.8 and 45 with microplastics in the diet (Table S15). PCB BMF values for polar cod, feeding on Calanus for an Arctic food web in the North of Alaska, were between 1.1 and 4.5, which agrees well to the here estimated values for polar cod feeding on zooplankton (BMF = 1.08–63.4). For PAHs congeners, BMF values ranged between 9.6 × 10\(^{-4}\) and 37 without microplastic in the diet and between 2.4 × 10\(^{-3}\) and 170 with microplastics in the diet (Table S16).

For PCB congeners, TMFs ranged between 5 and 37 when microplastic was not included in the diet and increased with congenor hydrophobicity (Figure S5B). Also these TMFs for PCBs overlap with the TMF values between 1.29 and 6.69 for a similar Arctic food web in the North of Alaska. Already with 1% microplastic in the diet, TMFs decrease to a range between 4 and 9 and have a maximum at intermediate PCBs; 52, 101, and 118. This maximum becomes less clear at higher percentages of microplastic with TMF values for the lower congeners becoming less than one when >99% of the diet is microplastic.

For PAHs, on the compound level, TMFs were low, ranging from 0.1 to 2.1 when microplastic was not included in the diet (Figure S5C). With 1% microplastic in the diet, TMFs are between 0.2 and 2.7 with the highest values for PAHs with highest log\(K_{OW}\). The relatively high TMF for coronene can be explained by the high log\(K_{OW}\) value and low metabolization rate. Note however that this TMF is much lower than PCBs with similar high log\(K_{OW}\) values. With increasing fraction of plastic in the diet, TMFs for low molecular weight PAHs increase, whereas TMFs for high molecular weight PAHs slightly decrease.

Like for food web transfer of microplastics as such, we also performed additional HOC scenario studies assuming microplastic was only included in the diet of zooplankton, as an entry point at the base of the food chain. The results indicate that excluding direct ingestion by species higher in the web attenuates the effects for PCBs (less “cleaning”) and low molecular weight PAH but generally increases the food web effects for high molecular weight PAH. Details are provided as S1 (Section S3, Figure S4E, S4F, SSE, SSF).

Finally, we also performed additional calculations where the food web was defined to represent a linear fully Arctic food web (Figure S6). It appears that the increasing and decreasing trends in HOC bioaccumulation with increasing plastic dose for PCBs and PAHs as described above do not change (Figure S6B,C). However, for PCBs, the linear food web (Figure S6) shows less bioaccumulation for the highest trophic level than for the default food web, which is explained from the shorter length of the linear Arctic food chain.

Implications of Nonequilibrium between HOC Concentrations in Microplastics and Water. The simulations discussed in the previous section assumed equilibrium partitioning (EqP) between HOCs in water and microplastics. EqP is widely accepted as the default condition used in the ecological risk assessment of HOCs. For oceanic microplastic, it has been argued that EqP can be assumed as well and that PCB and PAH concentrations showed little variation among pellets in the North Pacific Subtropical Gyre. Nevertheless, here we present simulations where the aqueous phase concentration was kept constant, and the HOC concentration in the microplastic was set at 0 (clean plastic), 0.01, 0.1, 1, 10, 100, and 1000 times the value of the EqP scenario (Figure 2). Scenarios 0, 0.01, and 0.1 thus represent cases where the plastic is in “underequilibrium”, implying that HOCs are in the process of rate-limited sorption to the plastic. Scenario 1 represents the previously discussed default EqP scenario, and scenarios 10, 100, and 1000 explore the cases when HOCs in plastic are in “overequilibrium”, which could be representative for some additives which are still in the process of rate-limited desorption from microplastics.

Compared to the EqP scenario, “underequilibrium” decreases chemical concentrations in lipids, whereas “overequilibrium” increases the chemical concentrations (Figures 2, S7). Furthermore, for PCBs the decrease in bioaccumulation with increasing plastic dose is more pronounced at “underequilibrium and less pronounced at “overequilibrium”. For Northern shrimp, Atlantic cod, seal, and polar bear, overequilibrium causes ingested microplastic to act as a carrier of contaminants, yet the extra bioaccumulation becomes less with increasing plastic dose. The latter “secondary cleaning effect” however pertains even when the PCB concentrations in microplastic are 1000 times higher than in the EqP scenario. For zooplankton and other species, this secondary cleaning effect does not occur, and the net vector effect increases with increasing overequilibrium factor as well as with increasing plastic dose. When cleaning of organism lipids by ingestion of microplastic is defined as the reduction of bioaccumulation compared to a scenario without plastic, a “cleaning” break-even point (CBP) can be defined. The CBP then is the point where the decrease in bioaccumulation due to the cleaning effect is exactly compensated for by overequilibrium. Although CBP values essentially relate to the specific combination of a species, chemical and plastic dose, here we present CBP values for sum PCBs. They were obtained by fitting the nonequilibrium factor until the calculated bioaccumulation was identical to the bioaccumulation calculated for the zero plastic scenario, using the Excel solver tool. For sum PCBs, the CBP values range from 1.4 to 3 × 10\(^{5}\) among species, and generally increase with trophic level and microplastic fraction in the diet (Figures S7, S8).

For sum PAHs (Figures 2D, 2E, 2F, S7), underequilibrium leads to a decrease in PAH concentration with increasing plastic
ingestion at higher trophic levels (e.g., Figure 2; panel E, scenario 0 and 0.01, and panel F scenario 0), whereas for the other species, PAH concentration increases with increasing plastic ingestion. Also here the CBP concept applies, that is, when nonequilibrium is higher than the CBP, ingested microplastic acts as a vector for PAHs. In contrast to PCBs where CBP values are higher than 1, for PAHs the values are lower than 1, i.e. between 0.03 and 0.29 depending on the species and plastic digestion dose (Figures S7, S8). Secondary cleaning does not occur for PAHs (Figure S7). These calculations show how the MICROWEB model can be used to assess prospective risks for POPs and additives under a variety of (non)equilibrium scenarios, despite the inherent complexity of chemical-microplastic interactions in food webs.

Sensitivity Analyses. We performed a sensitivity analysis emphasizing the novel parameters that drive the role of microplastic in the food web accumulation of HOCs (Table S17). In general, $C_{LIP}$ was more influenced at higher trophic levels. The model was most sensitive to $K_{PL}$ (up to 250), which can be explained from the fact that this parameter determines the HOC concentrations in the water. For sum PCBs, IR and $f_{LIP}$ were the next sensitive parameters, with a sensitivity of 2 to 3. For sum PAHs, the sensitivities to $IR, f_{LIP}, K_M$, and $K_{OW}$ were all around 1, indicating low sensitivity. Interestingly, model output was not very sensitive to the HOC kinetics in the organisms’ gut ($k_i$ and GRT, Table S17).

**GENERAL DISCUSSION**

We presented a theoretical model that simulates trophic transfer of microplastics and associated chemicals in biologically realistic food webs. Model analysis showed that although biomagnification of microplastic is not predicted, the trophic transfer as such has profound effects on the biomagnification of HOCs like PCBs and PAH. Because the effect is opposite for nonmetabolizable versus metabolizable HOCs, the net effect on ecological risks quotients for chemical mixtures depends on the relative concentrations of the components of the mixture and their interactions. In our case, PCB concentrations in biota lipids were 2–3 orders of magnitude higher than PAH concentrations. However, this may differ among locations and emphasizes the need to consider chemical risk on a case by case basis. We showed how the MICROWEB model can be used to assess prospective risks for POPs and additives, given the complexity of chemical-microplastic interactions inherent to food webs. Effects of microplastics on HOC accumulation generally were small when microplastic made up less than 3% of the diet, a condition which probably is met for many habitats. However, fractions of microplastics in biota diets may increase in the future and may differ locally. Recent data showed a plastic to biomass ratio of 180 for the North Pacific Subtropical Gyre,\(^{51}\) which would validate the relevance of the highest microplastic diet fractions modeled here. This does not imply that the exposure to some plastic-associated HOCs would be of primary concern. After all, the low food quality of a 99% plastic diet would have a far more direct and fast impact, i.e. via starvation, than increased exposure to HOCs.

As mentioned above, the concepts behind the MICROWEB model have been used and validated in earlier papers for a limited number (i.e. typically one) trophic level.\(^{4,12,14,47,78}\) Although consistent with current bioaccumulation theory, our model also has several limitations.

Microplastic was modeled as a conservative material flowing in and out of an organisms’ body without taking translocation and subsequent toxicodynamic processes into account. This ‘what goes in, must go out’ assumption is likely correct for a wide size range of particles but not necessarily for submicron or nanoscale particles.\(^{96}\) Also the chemical carrier or “Trojan horse” function of these submicron particles is not covered by the present implementation, whereas at the small scale, chemical sorption and release processes are known to differ from those for the bulk polymer material.\(^{79}\)

We assumed a food web structure based on the literature\(^{10}\) and showed that the general conclusions also hold for a differently defined food web.\(^{53,55}\) Still, we acknowledge that other food webs may give different results. We thus emphasize that the general model tool is the main merit of this work, rather than the specific simulation results for the case studies provided here. Food webs can be defined in different ways, i.e. modeled dynamically\(^{80}\) or assuming a steady state with fixed diet matrices based on best available information.

We assumed certain fractions of microplastics in the organisms’ diets; however, we have no information yet on how to accurately translate environmental concentrations to such fractions. For zooplankton, the diet fraction probably can be equated to the microplastic fraction in the mixture with phytoplankton; however, for higher trophic levels it is not that clear how much free-floating microplastic would be ingested besides that incorporated in the diet. We used a scenario approach that considered a wide range of diet fractions, however being the same for all species. In reality these fractions are supposed to be more variable across species, individuals of a species, locations, and time. This environmental variability is however not known. Nevertheless, it will not affect the trends reported here, whereas simulations may be updated once more specific information becomes available.

**ASSOCIATED CONTENT**

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.8b02515.

Additional information, including Tables S1–S17 and Figures S1–S8 (PDF)

**AUTHOR INFORMATION**

**Corresponding Author**

*Phone: +31 317 489701. E-mail: noel.diepens@wur.nl.

**ORCID**

Noël J. Diepens: 0000-0001-9159-7276

Albert A. Koelmans: 0000-0001-7176-4356

**Notes**

The authors declare no competing financial interest.

**ACKNOWLEDGMENTS**

This work is part of the project ‘Digestion and maternal/paternal transfer of microplastic contaminants in the Atlantic cod (Gadus morhua) food web’, funded by the Norwegian Research Council (project number 255267).

**REFERENCES**

1. Cole, M.; Lindeque, P.; Halsband, C.; Galloway, T. S. Microplastics as contaminants in the marine environment: A review. Mar. Pollut. Bull. 2011, 62 (12), 2588–2597.

2. Law, K. L. Plastics in the Marine Environment. Annu. Rev. Mar. Sci. 2017, 9 (1), 205–229.
(3) Engler, R. E. The Complex Interaction between Marine Debris and Toxic Chemicals in the Ocean. *Environ. Sci. Technol.* 2012, 46 (22), 12302–12315.

(4) Koelmans, A. A.; Bakir, A.; Burton, G. A.; Jannsen, C. R. Microplastic as a Vector for Chemicals in the Aquatic Environment: Critical Review and Model-Supported Reinterpretation of Empirical Studies. *Environ. Sci. Technol.* 2016, 50 (7), 3315–3326.

(5) Koelmans, A. A.; Besseling, E.; Shim, W. J. Nanoplastics in the Aquatic Environment: Critical Review. In *Marine Anthropogenic Litter*; Springer International Publishing: Cham, 2015; pp 325–340, DOI: 10.1007/978-3-319-16510-3_12.

(6) Bergmann, M.; Gutow, L.; Klages, M. *Marine Anthropogenic Litter*; Springer: Berlin, 2013.

(7) Johnston, E. L.; Mayer-Pinto, M.; Crowe, T. P. REVIEW: Chemical contaminant effects on marine ecosystem functioning. *J. Appl. Ecol.* 2015, 52 (1), 140–149.

(8) Zarfl, C.; Matthies, M. Are marine plastic particles transport vectors for organic pollutants to the Arctic? *Mar. Pollut. Bull.* 2010, 60 (10), 1810–1814.

(9) Lusher, A. L.; Tirelli, V.; O’Connor, I.; Officer, R. Microplastics in Arctic polar waters: the first reported values of particles in surface and subsurface samples. *Sci. Rep.* 2015, 5, 14947.

(10) Laender, F. De; Hammer, J.; Hendriks, A. J.; Soetaert, K.; Janssen, C. R. Combining Monitoring Data and Modeling Identifies PAHs as Emerging Contaminants in the Arctic. *Environ. Sci. Technol.* 2011, 45 (20), 9024–9029.

(11) Oubbard, R. W.; Sadri, S.; Wong, Y. Q.; Khtian, A. A.; Baker, I.; Thompson, R. C. Global warming releases microplastic legacy frozen in Arctic Sea ice. *Earth’s Future* 2014, 2 (6), 315–320.

(12) Bakir, A.; O’Connor, I. A.; Rowland, S. J.; Hendriks, A. J.; Thompson, R. C. Relative importance of microplastics as a pathway for the transfer of hydrophobic organic chemicals to marine life. *Environ. Pollut.* 2016, 219, 56–65.

(13) Teuten, E. L.; Saqing, J. M.; U Knappe, D. R.; Barlaz, M. A.; Jonsson, S.; Bjorn, A.; Rowland, S. J.; Thompson, R. C.; Galloway, T. S.; Yamashita, R.; et al. Transport and Release of Chemicals from Plastics to the Environment and to Wildlife. *Philos. Trans. R. Soc., B* 2009, 364 (1526), 2027–2045.

(14) Gouin, T.; Roche, N.; Lohmann, R.; Hodges, G. A Thermodynamic Approach for Assessing the Environmental Exposure of Chemicals Absorbed to Microplastic. *Environ. Sci. Technol.* 2011, 45 (4), 1466–1472.

(15) Ziccardi, L. M.; Edgington, A.; Hentz, K.; Kulacki, K. J.; Kane Driscoll, S. Microplastics as vectors for bioaccumulation of hydrophobic organic chemicals in the marine environment: A state-of-the-science review. *Environ. Toxicol. Chem.* 2016, 35 (7), 1667–1676.

(16) Amor, I. J.; Gobas, F. A. A review of bioconcentration factor (BCF) and bioaccumulation factor (BAF) assessments for organic chemicals in aquatic organisms. *Environ. Res.* 2006, 14 (4), 257–297.

(17) Walters, D. W.; Jardine, T. D.; Cade, B. S.; Kidd, K. A.; Muir, D. C. G.; Leipzig-Scott, P. Trophic Magnification of Organic Chemicals: A Global Synthesis. *Environ. Sci. Technol.* 2016, 50 (9), 4650–4658.

(18) Wright, S. L.; Thompson, R. C.; Galloway, T. S. The physical impacts of microplastics on marine organisms: A review. *Environ. Pollut.* 2013, 178, 483–492.

(19) Cole, M.; Lindeque, P.; Fileman, E.; Halsband, C.; Goodhead, R.; Moger, J.; Galloway, T. S. Microplastic Ingestion by Zooplankton. *Environ. Sci. Technol.* 2013, 47 (12), 6646–6655.

(20) Bravo Rebollo, E. L.; Van Franeker, J. A.; Janssen, O. E.; Brasseur, S. M. J. M. Plastic ingestion by harbour seals (Phoca vitulina) in The Netherlands. *Mar. Pollut. Bull.* 2013, 67, 200–202.

(21) Briste, I. L. N.; Eidsvoll, D. P.; Steindal, C. C.; Thomas, K. V. Plastic ingestion by Atlantic cod (Gadus morhua) from Newfoundland destined for human consumption collected through citizen science methods. *Environ. Sci. Technol*. 2017, 51, 7987–7995.

(22) Moger, J.; Galloway, T. S. Microplastic Ingestion by Zooplankton. *Environ. Sci. Technol.* 2015, 49 (7), 4037–4044.

(23) Koelmans, A. A.; Besseling, E.; Wegner, A.; Foekema, E. M. Plastic as a Carrier of POPs to Aquatic Organisms: A Model Analysis. *Environ. Sci. Technol.* 2013, 47 (4), 7812–7820.

(24) Van Cauwenbergh, L. Occurrence, effects and risks of marine microplastics, Ghent University, Belgium, 2015.

(25) Koelmans, A. A.; Besseling, E.; Foekema, E. M. Leaching of plastic additives to marine organisms. *Environ. Pollut.* 2014, 187, 49–54.

(26) Beckingham, B.; Ghosh, U. Differential bioavailability of polychlorinated biphenyls associated with environmental particles: Microplastic in comparison to wood, coal and biochar. *Environ. Pollut.* 2017, 220, 150–158.

(27) Paul-Pont, I.; Lacroix, C.; Gonzalez Fernández, C.; Hégaret, H.; Lambert, C.; Le Goic, N.; Frère, L.; Cassone, A.-L.; Sussarellu, R.; Fabioux, C.; et al. Exposure of marine mussels Mytilus spp. to polystyrene microplastics: Toxicity and influence on fluoranthene bioaccumulation. *Environ. Pollut.* 2016, 216, 724–737.

(28) GESAMP. Sources, Fate and Effects of Microplastics in the Marine Environment: A Global Assessment. Rep. Stud. GESAMP No. 90; 2015.

(29) Besseling, E.; Foekema, E. M.; Van den Heuvel-Greve, M. J.; Koelmans, A. A. The effect of microplastic on chemical uptake by the lugworm Arenicola marina (L.) under environmentally relevant conditions. *Environ. Sci. Technol.* 2017, 51, 8795.
(42) EPA. State of the Science White Paper: A Summary of Literature on the Chemical Toxicity of Plastics Pollution to Aquatic Life and Aquatic-Dependent Wildlife; EPA-822-R-16-009; 2016.

(43) Hartmann, N. B.; Rist, S.; Bodin, J.; Jensen, L. H.; Schmidt, S. N.; Mayer, P.; Meibom, A.; Baun, A. Microplastics as vectors for environmental contaminants: Exploring sorption, desorption, and transfer to biota. Int. J. Environ. Res. Public Health 2017, 14 (3), 488–493.

(44) Devriese, L. I.; De Witte, B.; Vethaak, A. D.; Hostens, K.; Leslie, H. A. Bioaccumulation of PCBs from microplastics in Norway lobster (Nephrops norvegicus): An experimental study. Chemosphere 2017, 186, 10–16.

(45) Rehse, S.; Klosa, W.; Zarfl, C. Microplastics Reduce Short-Term Effects of Environmental Contaminants. Part I: Effects of Bisphenol A on Freshwater Zooplankton Are Lower in Presence of Polymide Particles. Int. J. Environ. Res. Public Health 2018, 15 (2), 280.

(46) Bräte, I. L. N.; Eidsvoll, D. P.; Steindal, C. C.; Thomas, K. V. Plastic ingestion by Atlantic cod (Gadus morhua) from the Norwegian coast. Mar. Pollut. Bull. 2016, 112 (1), 105–110.

(47) Herzez, D.; Anker-Nilssen, T.; Nost, T. H.; Götsch, A.; Christensen-Dalsgaard, S.; Langset, M.; Fangøl, K.; Koelmans, A. A. Negligible Impact of Ingested Microplastics on Tissue Concentrations of Persistent Organic Pollutants in Northern Fulmars off Coastal Norwegian. Envir. Sci. Technol. 2016, 50 (4), 1924–1933.

(48) Bergami, E.; Pugnalini, S.; Vannuccini, M. L.; Manfra, L.; Faleri, C.; Savorelli, F.; Dawson, K. A.; Corsi, I. Long-term toxicity of surface-Dunaliella tertiolecta and Artemia franciscana. Int. J. Environ. Assess. Manage. 2017, 11 (3), 460–465.

(51) Ku, H.; Koelmans, A. A. Bioaccumulation of PCBs from microplastics in Norway lobster (Nephrops norvegicus): An experimental study. Environ. Toxicol. Chem. 2008, 27 (11), 2263–2270.

(56) Endo, S.; Koelmans, A. A. Sorption of Hydrophobic Organic Materials and metabolic transformation. Environ. Sci. Technol. 2017, 51 (2), 318–324.

(61) Chen, Q.; Reisser, J.; Cunsolo, S.; Kwadijk, C.; Otterman, M.; Proietti, M.; Slat, B.; Ferrari, F.; Schwarz, A.; Levivier, A.; et al. Pollutants in Plastics within the North Pacific Subtropical Gyre. Environ. Sci. Technol. 2018, 52, 446–456.

(62) Fisner, M.; Taniguchi, S.; Moreira, F.; Bicego, M. C.; Turra, A. Polycyclic aromatic hydrocarbons (PAHs) in plastic pellets: Variability in the concentration and composition at different sediment depths in a sandy beach. Mar. Pollut. Bull. 2013, 70 (1), 219–226.

(63) Fisner, M.; Taniguchi, S.; Mayer, A. P.; Bicego, M. C.; Turra, A. Concentration and composition of polycyclic aromatic hydrocarbons (PAHs) in plastic pellets: implications for small-scale diagnostic and environmental monitoring. Mar. Pollut. Bull. 2013, 76 (1–2), 349–354.

(64) van der Oost, R.; Beyer, J.; Vermeulen, N. P. E. Fish bioaccumulation and biomarkers in environmental risk assessment: a review. Environ. Toxicol. Pharmacol. 2003, 13 (2), 57–149.

(65) Berrojalbiz, N.; Lacorte, S.; Calbet, A.; Saiz, E.; Barata, C.; Dachs, J. Accumulation and cycling of polycyclic aromatic hydrocarbons in zooplankton. Environ. Sci. Technol. 2009, 43 (7), 2295–2301.

(66) Arnot, J. A.; Mackay, D.; Parkerton, T. F.; Bonnell, M. A database of fish bioconcentration rates for organic chemicals. Environ. Toxicol. Chem. 2008, 27 (11), 2263–2270.

(67) Meinicke, C. T. A.; Traas, T. P.; Roessink, I.; Veltman, K.; Hendriks, A. J.; Koelmans, A. A. Modeling decreased food chain accumulation of PAHs due to strong sorption to carbonaceous materials and metabolic transformation. Environ. Sci. Technol. 2007, 41 (17), 6185–6191.

(68) Koelmans, A. A.; Kooi, M.; Law, K. L.; van Sebille, E. All is not lost: deriving a top-down mass budget of plastic at sea. Environ. Res. Lett. 2012, 7 (11), 114028.

(69) Lohmann, R. Microplastics are not important for the cycling and bioaccumulation of organic pollutants in the oceans—but should microplastics be considered POPs themselves? Integr. Environ. Assess. Manage. 2017, 13 (3), 460–465.

(70) Diepens, N. J.; Koelmans, A. A.; Baveco, H.; van Den Brink, P. J.; van Den Heuvel-Greve, M. J.; Brock, T. C. M. Prospective environmental risk assessment for sediment-bound organic chemicals: A proposal for tiered effect assessment. Rev. Environ. Contam. Toxicol. 2017, 239, 1–77.

(71) ECHA. Principles for environmental risk assessment of the sediment compartment: Proceedings of the topical scientific workshop, 7–8 May 2013; Tarazona, J. V., Verssonen, B., Janssen, C., De Laender, F., Vangheluwe, M., Knight, D., Eds.; European Chemicals Agency: Helsinki, Finland, 2014; Vol. ECHA-14-R-13-EN.

(72) Seidensticker, S.; Zarfl, C.; Cirpka, O. A.; Fellenberg, G.; Grathwohl, P. Shift in Mass Transfer of Wastewater Contaminants from Materials and metabolic transformation. Environ. Sci. Technol. 2017, 51 (21), 12254–12263.

(73) Norris, M. D. Factorial sampling plans for preliminary experimental investigations. Technometrics 1991, 33 (2), 161–174.

(74) Macdonald, R. W.; Mackay, D.; Hickie, B. Contaminant amplification in the environment: revealing the fundamental mechanisms. Environ. Sci. Technol. 2006, 36, 456A–462A.

(75) Di Paolo, C.; Gandhi, N.; Bhavsar, S. P.; Van Den Heuvel-Greve, M.; Koelmans, A. A. Black carbon inclusive multichemical modeling of PBDE and PCB biomagnification and -transformation in estuarine food webs. Environ. Sci. Technol. 2010, 44 (19), 7548–7554.

(76) Rummel, C. D.; Adolphsson-Enrici, M.; Jahne, A.; MacLeod, M. No measurable “cleaning” of polychlorinated biphenyls from Rainbow Trout in a 9 week depuration study with dietary exposure to 40% polyethylene microspheres. Environ. Sci. Process. Impacts 2016, 18 (7), 788–795.

(77) Hoekstra, P. F.; O’hara, T. M.; Fisk, A. T.; Borgå, K.; Solomon, K. R.; Muir, D. C. G. Trophic transfer of persistent organochlorine contaminants (OCs) within an Arctic marine food web from the southern Beaufort–Chukchi Seas. Environ. Pollut. 2003, 124 (3), 509–522.
(78) Rochman, C. M.; Parnis, J. M.; Browne, M. A.; Serrato, S.; Reiner, E. J.; Robson, M.; Young, T.; Diamond, M. L.; Teh, S. J. Direct and indirect effects of different types of microplastics on freshwater prey (Corbicula fluminea) and their predator (Acipenser transmontanus). PLoS One 2017, 12 (11), e0187664.

(79) Velzeboer, I.; Kwadijk, C. J. A. F.; Koelmans, A. A. Strong sorption of PCBs to nanoparticles, microplastics, carbon nanotubes, and fullerenes. Environ. Sci. Technol. 2014, 48 (9), 4869–4876.

(80) Koelmans, A. A.; Van der Heijde, A.; Knijff, L. M.; Aalderink, R. H. Integrated modelling of eutrophication and organic contaminant fate and effects in aquatic ecosystems. A Review. Water Res. 2001, 35 (15), 3517–3536.