Relative importance of microplastics as a pathway for the transfer of hydrophobic organic chemicals to marine life

Adil Bakir [abc], *Isabel A. O'Connor* [d], *Steven J. Rowland* [b], *A. Jan Hendriks* [d] *and Richard C. Thompson* [a]

[a] *Marine Biology and Ecology Research Centre (MBERC) School of Marine Science and Engineering, Plymouth University, Drake Circus, Plymouth, Devon, PL4 8AA, United Kingdom*

[b] *Petroleum and Environmental Geochemistry Group (PEGG) School of School of Geography, Earth and Environmental sciences, Plymouth, University Drake Circus, Plymouth, Devon, PL4 8AA, United Kingdom*

[c] *School of Earth and Environmental Sciences, University of Portsmouth, Burnaby Road, Portsmouth, PO1 3QL, United Kingdom*

Radbound University Nijmegen, Institute for water and Wetland research, Department of Environmental Science, P.O. Box 9010, NL-6500 GL, Nijmegen, The Netherlands

Corresponding author: R.C.Thompson@plymouth.ac.uk

Abstract

It has been hypothesised that, if ingested, plastic debris could act as vector for the transfer of chemical contaminants from seawater to organisms, yet modelling suggest that, in the natural environment, chemical transfer would be negligible compared to other routes of uptake. However, to date, the models have not incorporated consideration of the role of gut surfactants, or the influence of pH or temperature on desorption, whilst experimental work has shown that these factors can enhance desorption of sorbed contaminants several fold. Here, we modelled the transfer of sorbed organic contaminants dichlorodiphenyltrichloroethane (DDT), phenanthrene (Phe) and bis-2-ethylhexyl phthalate (DEHP) from microscopic particles of polyvinylchloride (PVC) and polyethylene (PE) to a benthic invertebrate, a fish and a seabird using a one-compartment model OMEGA (Optimal Modeling for EcotoxicoloGical Applications) with different conditions of pH, temperature and gut surfactants. Environmental concentrations of contaminants at the bottom and the top of published ranges were considered, in combination with ingestion of either 1 or 5% by weight of plastic. For all organisms, the combined intake from food and water was the main route of exposure for Phe, DEHP and DDT with a negligible input from plastic. For the benthic invertebrate, predictions including the presence of contaminated plastic resulted in very small increases in the internal concentrations of DDT and DEHP, while the net change in the transfer of Phe was negligible. While there may be scenarios in which the presence of plastic makes a more important contribution, our modelling study suggests that ingestion of microplastic does not provide a quantitatively important additional pathway for the transfer of adsorbed chemicals from seawater to biota via the gut.

Capsule abstract: Combined intake from food and water was the main route of exposure for DEHP, DDT and Phe to the organisms under investigation with a negligible input from plastic.

Introduction

Plastics are extraordinarily useful materials, due to their low cost, high malleability and durability. However, their longevity is resulting in substantial accumulation in some environments (Browne et al., 2010). Microplastic fragments are the most numerically abundant types of plastic debris in some locations (Goldstein et al., 2012) and modelling studies suggest the global ocean may be contaminated with 93 000 to 236 000 metric tons particle of microplastic (van Sebille et al., 2015). These small pieces of debris (<5mm) can form as a result of the fragmentation of larger items or as a result of direct release of small particles, such as microbeads from cosmetics, to the environment (Napper et al., 2015). Ingestion of microplastics, has been reported for a wide range of organisms including deposit and suspension feeders (Browne et al., 2008; Graham and Thompson, 2009; Ward and Shumway, 2004), crustaceans (Murray and Cowie, 2011), fish (Boerger et al., 2010), marine mammals (Denuncio et al., 2011) and seabirds (Avery-Gomm et al., 2012; van Franeker and Bell, 1988; van Franeker et al., 2011b). Deleterious physical effects on wildlife from ingestion of macroscopic pieces of plastics are well documented and recent work suggests that microscopic particles can also have harmful physical effects (Wright et al., 2013). However, the ecotoxicological consequences of ingesting microplastics are less clear. Two

routes of exposure have been suggested: i) exposure from the release of chemical additives that were incorporated into plastics during manufacture and/or ii) the transfer and accumulation of organic or inorganic contaminants from seawater to organisms as a consequence of ingestion. This paper examines the potential for plastics to act as a vector in the transport of hydrophobic organic chemicals (HOCs) from seawater to marine organisms.

There are relatively few studies estimating the potential transfer of HOCs by microplastics (Gouin et al., 2011; Koelmans et al., 2016; Koelmans et al., 2014; Koelmans et al., 2013; Teuten et al., 2007). Using a bioavailability model, Teuten et al. (2007) showed that the addition of as little as 1 μg of contaminated PE to a gram of sediment would give a significant increase in phenanthrene (Phe) accumulation by *A. marina*. (Teuten et al., 2007). This was supported by the work of Besseling et al. (2012) who, using laboratory studies, showed an increase in bioaccumulation of polychlorinated biphenyls (PCBs) into *A. marina* when contaminated polystyrene (PS) was present in sediments (0.074 % plastic d.w.) (Besseling et al., 2012). In the natural environment, a positive correlation has also been demonstrated between the amount of ingested plastic particles and the concentrations of PCBs in the tissues of birds (Great Shearwaters; *Puffinus gravis*) (Ryan et al., 1988). Work by Tanaka et al. (2013) also provided correlative evidence for the transfer of plastic-derived polybrominated diphenyl ethers (PBDEs) from ingested particles to the short-tailed shearwaters *Puffinus tenuirostris* (Tanaka et al., 2013). It is however difficult to conclude whether PCBs accumulation in their tissues resulted from pollutant transfer from plastics as opposed to other sources, such as contaminated food. Recent analyses of seabirds contaminated with plastics suggests that exposure of the northern fulmar (*Fulmarus glacialis*) to polychlorobiphenyls (PCBs) due to ingestion of microplastic was probably negligible compared to the chemical fluxes entering the birds via their prey as internal HOC concentration was not linked to their stomach plastic concentrations (Herzke et al., 2015). In

contrast, recent laboratory work has shown that chemicals sorbed onto plastic in the marine environment can have negative effects on fish (Rochman et al., 2013). However, in this experiment, plastics were the only source of contaminants. In order to fully understand the potential for plastics to cause harm to marine life as a consequence of the transfer of contaminants from seawater to an organism it is essential to understand the relative importance of plastics compared to other pathways for chemical transfer, such as via respiration or diet.

Recent models have concluded that the relative importance of plastic particles as vectors for HOCs to marine organisms is likely of limited importance when compared to other exposure pathways (Gouin et al., 2011; Koelmans et al., 2014; Koelmans et al., 2013). However, as outlined by Engler (2012), such models neglect several factors, namely: i) the role of gut retention time of ingested particles, ii) the role of physiological processes such as the presence of enzymes or gut surfactants and iii) differing physiological conditions of pH and temperature according to the type of organism with the case of a benthic invertebrate, a marine fish and a seabird. All these factors will likely influence the bioavailability of sorbed contaminants (Engler, 2012). For example, recent work has shown that Phe, DDT and bis-2-ethylhexyl phthalate (DEHP) sorbed onto PVC and PE desorbed substantially faster in the presence of surfactants and at gut pH in cold blooded organisms and were further enhanced in warm blooded organism with a combined surfactant, pH and temperature enhancement rate of over 30 times compared to in seawater alone (Bakir et al., 2014). Enhanced desorption rates might be an important factor when assessing transfer of plastic co-contaminants to organisms upon ingestion, especially if gut transit time is short i.e. faster release in the gut. Enhancement of the leaching of plastic co-contaminants, such as polybrominated diphenyl ethers (PBDEs) was also reported in seabird's stomach oil with subsequent accumulation in tissues (Tanaka et al., 2015). Over 20 times as much material was leached into stomach oil,

and over 50 times as much into fish oil (a major component of stomach oil), than in aqueous solution alone.

Previous work also indicates that sorption capacity and desorption rates are highly pollutant and polymer specific. Hence robust predictions can only be made using a systematic approach considering different HOC and plastic combinations under physiologically relevant scenarios (Bakir et al., 2014).

A range of environmentally relevant scenarios are investigated herein according to reported concentrations of HOCs in seawater and considering locations that had contrasting levels of contamination using data from low and highly polluted sites, together with either low or high quantities of ingested plastics (1 and 5% ingested plastic particles). A comparison with respiratory and dietary uptake was then used to determine the relative importance of transport by contaminated microplastics compared to other pathways. A benthic invertebrate, a pelagic fish and a seabird were selected as candidate organisms for our models as they represent both cold and warm blooded organisms. Our invertebrate example was based on *A. marina* as some work has already been done on this species in relation to uptake of HOCs (Besseling et al., 2012; Browne et al., 2013). *A. marina* is widely distributed, OSPAR (Convention for the Protection of the Marine Environment of the North-East Atlantic) approved species and forms an important component of marine food webs. This species has already been shown to ingest microplastics (Thompson et al., 2004) with some indication of bioaccumulation of PCBs sorbed onto PS present in sediments (Besseling et al., 2012). Fish were selected because microplastics have been reported in the gut of several pelagic and demersal fish species (Boerger et al., 2010; Foekema et al., 2013; Lusher et al., 2013). It is therefore highly likely that fish ingesting plastic particles are in contact with sorbed chemicals which could desorb in the gut (Bakir et al., 2014; Endo et al., 2013). However, the associated consequences are not known. Seabirds are also known to ingest plastic debris with

detrimental physical effects and concern about the transfer of harmful chemicals (Ryan, 1990). For birds, the uptake of organic compounds from seawater can only take place via ingestion of marine organisms such as fish (Walker et al., 2001). Ingestion of plastic debris by Northern fulmars has been documented in several studies at numerous locations over time (Avery-Gomm et al., 2012; Kühn and van Franeker, 2012; Mallory, 2008; Mallory et al., 2006; van Franeker, 1985) allowing them to be used as biological indicators for spatial and temporal trends of plastic pollution (Avery-Gomm et al., 2012; van Franeker, 1985; van Franeker et al., 2011a). Evidence of transfer of pollutants from ingested plastic debris could therefore be integrated into this environmental monitoring to produce a tool for the environmental risk assessment of microplastics in the marine environment required to reach Good Environmental Status (GES) as part of the quality descriptor 10 of the Marine Strategy Framework Directive (MSFD 2008/56/EC).

The main objectives of the present study were to , i) integrate previously quantified distribution coefficients and desorption rates for Phe, DDT and DEHP onto PVC and PE into bioavailability models for various scenarios of contaminant concentrations and plastic abundance in order to predict transfer to a range of marine organisms occupying different ecological niches/feeding strategies and ii) determine the relative importance of plastics compared to other routes of exposure (food, respiration) for the transport of contaminants from seawater.

Methods

Model design, description and implementation

Distribution coefficients (K_d) and desorption rates under varying gut conditions (different pH and temperatures scenarios) for the sorption/desorption of Phe, DDT and DEHP onto and

from PVC and PE (200-250 μm) were determined previously (Bakir et al., 2014). Desorption rates ranged from 1 to 10 d^{-1} in accordance with Koelmanns *et al.* (2013) and were also in agreement with the derived desorption rate of nonylpehnol (NP) from PVC upon ingestion by *Arenicola marina* of 0.5 d^{-1} as shown experimentally by Browne et al. (2013) (Browne et al., 2013).

Koelmanns et al. (2013), suggested that desorption rate constants to gut fluids are between 1 and 10 d^{-1}. However, our experimentally measured desorption rates ranged from 0.23 ± 0.08 to 12.10 ± 2.09 d^{-1} depending on the chemical-plastic combination as well as the simulated gut conditions (varying pH and temperature)(Bakir et al., 2014), indicating that some combinations of chemicals, plastic and gut conditions may lead to faster desorption of contaminants(Koelmans et al., 2014). Tanaka et al. (2015) also demonstrated considerably enhanced desorption of PBDEs from plastic under gut conditions. It is therefore important to consider such enhancement in desorption rates in a range of gut conditions (e.g. bile) for a range of plastic/co-contaminants.

Uptake and elimination rates of contaminants were estimated for three routes of exposure including uptake and elimination from/to water, food and plastic. Each prediction was carried out for Phe, DDT and DEHP onto both PVC and PE. For all the contaminants, 'low' and 'highly' polluted sites were examined according to reported aqueous concentrations for each contaminant (Table S1). In our scenarios, plastic ingestion relative to food was 1 and 5 % of diet (Carson et al., 2011). A one-compartment model was applied for an invertebrate benthic deposit feeder, a marine fish and a seabird (O'Connor et al., 2013a). Chemical concentrations in each species were calculated using three approaches (A-C): A) where concentrations of the pollutant in each species were calculated with the assumption that an individual ingested and egested sufficient plastic over its lifetime to reach equilibrium between lipid and plastics, B)

concentrations of the pollutant in individuals would achieve a lifelong plastic ingestion were calculated with the assumption that all pollutants sorbed onto plastics were transferred to the organisms and C) An OMEGA one-compartment model was used to calculate steady state internal concentrations of the chemicals. Approaches A and B represent worst-case scenarios and are shown in supplementary information while approach C, which represents the most likely environmental scenario, is presented here.

Bioaccumulation model

An OMEGA model (Optimal Modeling for EcotoxicoloGical Applications) was used to calculate the steady state internal concentrations of pollutants in individuals at different trophic levels (Hendriks et al., 2005; Hendriks et al., 2001b). For species i, the internal concentration (C_i) at steady state equals the ratio of the sum of uptake divided by the sum of elimination. In this study, we considered pollutant uptake from water ($k_{w,X,in} \cdot C_w$), from food ($k_{f,X,in} \cdot C_{food}$) and from plastic ($k_{p,X,in} \cdot C_p$) as well as elimination with water ($k_{w,X,out}$), faeces ($k_{f,X,out}$), plastic ($k_{p,X,out}$) and biomass dilution from growth or reproduction ($k_{b,X,out}$). Biotransformation can reduce the bioaccumulation potential of organic chemicals. However, rates are difficult to estimate, and therefore are not included in Eq.1. Our scenarios therefore reflect a worst-case scenario.

$$C_i = \frac{k_{w,X,in} \cdot C_w + k_{f,X,in} \cdot C_{food} + k_{p,X,in} \cdot C_p}{\Sigma k_{w,X,out} + k_{f,X,out} + k_{p,X,out} + k_{b,X,out}} \quad \textbf{(Eq. 1)}$$

Calculation of all rates and concentrations are explained in more detail in the supporting information. We treated plastic like indigestible food (Eq. 2). The term ($1\text{-}p_p$) denotes fraction of undigested plastic, and K_{pw} the plastic water partition coefficient. The denominator in the third term describes the diffusion resistances and the flow delays the pollutant experiences

during uptake: the diffusion resistance of the unstirred water layer ($\rho_{H20,f}$ in d kg^{-1}), through lipid layers ($\rho_{CH2,i}$ in d kg^{-1}) and the flow delay of the pollutant contained in the undigested food passing through the intestinal tract (1/ ($K_{pw} \cdot (1\text{-}p_p) \cdot \gamma_p \cdot q_T$)). Finally, the uptake rate scales allometrically to the species weight w (in kg) with the rate coefficient κ (-):

$$k_{p,X,in} = \frac{1}{1-p_p} \cdot \frac{1}{K_{pw}} \cdot \frac{w^{-\kappa}}{\rho_{H_2O,f} + \frac{\rho_{CH_2,i}}{K_{ow} \cdot q_T} + \frac{1}{K_{pw} \cdot (1-p_p) \cdot \gamma_p \cdot q_T}} \quad \textbf{(Eq. 2)}$$

With w = Average weight of an individual of the particular species (kg)

κ = rate coefficient (-)

p_p = Fraction of plastic assimilated (kg kg^{-1})

$P_{lip,i}$ = lipid fraction of animal (kg kg^{-1})

K_{pw} = Plastic-water partition coefficient

$\rho_{H20,f}$ = Water layer resistance from/to food (d kg^{-K})

$\rho_{CH2,i}$ = Lipid layer resistance (d kg^{-K})

K_{ow} = Octanol-water partition coefficient

q_T = Temperature correction factor (kg kg^{-1})

γ_p = Plastic ingestion coefficient (kg^{k} d^{-1})

Analogously, the elimination rate via plastic egestion was calculated using Eq. 3:

$$k_{p,X,out} = \frac{1}{p_{lip,i} \cdot (K_{ow}-1)+1} \cdot \frac{w^{-\kappa}}{\rho_{H_2O,f} + \frac{\rho_{CH_2,i}}{K_{ow} \cdot q_T} + \frac{1}{K_{pw} \cdot (1-p_p) \cdot \gamma_p \cdot q_T}} \quad \textbf{(Eq. 3)}$$

where the term $p_{lip,i} \cdot (K_{ow} - 1) + 1$ reflects the affinity of the chemical for the lipid and water compartments of the organism. The pollutant concentration in food was estimated using a standard food chain bioaccumulation model (O'Connor et al., 2013b).

Parameterisation

Model parameters are listed in Table S6 and parameters specific to each species are shown in Table S7. For the plastic ingestion rate, the assumption that no fraction of the plastic was assimilated was formulated ($p_p = 0$). The plastic ingestion coefficient (γ_p in $kg^{\kappa}\ d^{-1}$) was calculated as a fraction of the food ingestion coefficient. As there was no available reported plastic ingestion coefficient, three approaches were examined; here the plastic ingestion coefficient equaled 1% and 5% of the food ingested. At the same time, the food intake was kept constant. This scenario might not be realistic as the food intake might decrease with an increasing intake in plastic.

Evaluation of desorption: The OMEGA model is based on partition coefficients and thus assumed instantaneous equilibrium. However, our previous work has demonstrated that desorption rates were higher at higher temperature and lower pH (Bakir et al., 2014). Therefore, we evaluated the assumption of instantaneous equilibrium. Equations 4, 5 and 6 are not part of approach C but an additional analysis in order to evaluate the assumption of instantaneous equilibrium and desorption in approach C. As a first step, we calculated the uptake efficiency *E* (in %) of each pollutant from plastic in order to determine the amount of contaminant absorbed. The uptake efficiency equals the ratio of the pollutant uptake rate and the plastic ingestion rate (Eq. 4):

$$E = \frac{k_{p,X,in}}{k_{p,in}} \qquad \textbf{(Eq. 4)}$$

Plastic ingestion rate was proportional to the plastic ingestion rate coefficient and the temperature correction factor and scaled allometrically to the species mass *w* (in kg) with the coefficient κ (-) (Eq. 5):

$$k_{p,in} = \gamma_p \cdot q_t \cdot w^{-\kappa} \qquad \textbf{(Eq. 5)}$$

Subsequently, we calculated for a lugworm, a fish and a seabird whether the particle retention time (t_r [d]) was long enough to allow complete desorption of the pollutant from the plastic. We assumed a gut retention time (GRT) for food particles for *A. marina*, fish and seabird of 2h, 4-158 h and 11 h respectively (Table S2). Hence we assumed that plastic passed through the organisms at a similar rate to their typical food. We quantified the pollutant concentration onto plastic as in the high pollution scenario and calculated the fraction of the pollutant which remained sorbed to the plastic after the gut residence time of the particle (Eq. 6):

$$\frac{C_p(t=t_r)}{C_p(t=0)} = \frac{C_p(t=t_r) \cdot e^{-k_{des} \cdot t}}{C_p} \qquad \textbf{(Eq. 6)}$$

where k_{des} denotes the measured desorption rate constant (d^{-1}) as measured in Bakir et al. (2014).

Results and discussion

Model validation without plastic using reported environmental data

Model validation was carried out by comparing modelled concentrations in the organisms calculated to arise in the absence of plastics, with representative data for reported HOCs concentrations for biota in the environment.

The estimated DEHP concentration in the lugworm, excluding plastic (1055 μg kg^{-1} ww for a low polluted site), was in the same order of magnitude to reported concentrations in lugworms and was in accordance with DEHP levels reported for the sandworm *Neanthes virens* (490 μg kg^{-1} ww) (Table S5 (Ray et al., 1983a). Higher concentrations were reported for invertebrates sampled at the vicinity of a DEHP processing plant, simulated by a highly polluted site in this study (439 mg kg^{-1} ww (493,000 μg kg^{-1} ww), Table S5, with concentration of 5300 μg kg^{-1} ww for *Odonata sp*. to up to 14400 μg kg^{-1} for *Asellus*

aquaticus (Organization, 1992; Persson et al., 1978). Estimated concentrations from food excluding plastic were thus in agreement with reported HOCs concentrations for the lugworm for different locations, indicating the environmental relevance of our model (Tables S2-S4). Concentrations of DDT predicted in the tissues of the seabird from food (excluding plastic) were also in accordance with reported concentrations of DDT in low and highly polluted sites (178 μg kg^{-1} ww and 20.5 mg kg^{-1} ww respectively, Table S2). However, concentrations of DDT are highly variable, according to species. For example, reported *p,p'*-DDT concentrations ranged from 58 μg kg^{-1} ww for the black-browed albatross to 2.6 mg kg^{-1} ww for the black-footed albatross (Guruge et al., 2001). DEHP concentrations in fish are very variable depending on the habitat, feeding habits and biodegradation levels of phthalates (Chang et al., 2005; Yuan et al., 2002). High concentrations of DEHP have been widely reported in fish with concentrations up to 254 mg kg^{-1} dw (Huang et al., 2008). Data on DEHP concentrations in birds are limited. Relatively low concentrations have been reported for kittiwakes located in remote fjords of the Norwegian Arctic with concentrations up to 155 μg kg^{-1} ww (Institute, 2009). Much higher concentrations are expected to be reached in highly polluted areas (Persson et al., 1978).

Relative contribution of microplastics for the transfer of sorbed HOCs to marine organisms as compared to the contribution from food and water.

The OMEGA model permitted us to differentiate and estimate the relative contributions of microplastics from that of the combined intake from food and water to the total body burden of HOCs.

Benthic invertebrate*:* Combined intake from food and water was the main route of exposure for DEHP, DDT and Phe to the benthic invertebrate with a negligible input from plastic

(Fig.1). Predicted net transfers of HOCs to the lugworm were in the order [DEHP] > [DDT] > [Phe] in the absence of plastics and in both low HOCs-polluted and highly-HOCs polluted scenarios, with either 1% or 5% plastics (Fig.1). Estimated concentrations of DEHP transferred from PVC and PE plastics to the tissues of *A. marina* were higher than for DDT and Phe as a consequence of the high concentrations of DEHP found in the marine environment (due to its use in the manufacture of some plastics, Table S1). For DEHP sorbed onto PVC the contribution under the same conditions was less than PE, with a 2% increased contribution to the total body burden predicted by the model (Fig.1).

The contribution of the plastics to changes in the Phe body burden were negligible < 0.1% increase with both PVC and PE, while the contribution increased somewhat for DDT (max. of 2% increase for the scenario with 5% ingested plastics) with a maximum of an 11% increase in DEHP from PE in the worst case scenario (Fig.1).

***Marine fish*:** Combined intake from food and water was the main route of exposure for DEHP, DDT and Phe to the marine fish with a negligible input from plastic (Fig.2). Predicted net transfers of contaminants to a marine fish were again dependent on the chemical contaminant (HOC) (Fig.2) and also followed the order [DEHP] > [DDT] > [Phe] in the absence of plastics and in both low HOCs-polluted and highly-HOCs polluted scenarios, and with either 1% or 5% plastics. Whilst a decrease in bioaccumulation of each of the HOCs was predicted following plastics ingestion (Fig.2), for Phe and DEHP this was very small for both plastics, for DDT a decrease in body burden of 4% was predicted for PE and PVC for a worst case scenario (5 % ingested plastic) (Fig. S2b).

***Seabird*:** Combined intake from food and water was the main route of exposure for DEHP, DDT and Phe for the seabird with a negligible input from plastic (Fig.3). Predicted net transfers of contaminants to seabirds were also highly dependent on the chemical (Fig.3) and

once more followed the order [DEHP] > [DDT] > [Phe] in the absence of plastics and in both low HOCs-polluted and highly-HOCs polluted scenarios, and with either 1% or 5% plastics.

Concentrations of p,p'-DEE (as the major DDT) reported in muscle and liver tissues in 75 Northern Fulmars (Herzke et al., 2015) were in agreement with the internal DDT concentration predicted in the present study with concentrations of 178, 176 and 166 μg kg^{-1} ww for no plastic, 1 and 5% ingested PE respectively for a low polluted site (Figs. 3 and S10).

Tanaka et al. (2015) also investigated the accumulation of PBDEs from ingested plastics in the tissues of 18 wild seabirds which contained on average 22.5 plastic particles in either their gizzard or in their proventriculus (average weight of plastic 0.31 g per bird). This was in the range of the amount of plastic reported in the gut of seabirds, including Northern fulmars (Table S2A) (Avery-Gomm et al., 2012; Blight and Burger, 1997; van Franeker et al., 2011a). PBDEs were detected in all birds in both the liver and abdominal adipose tissue suggesting strong correlation between HOCs in ingested plastics and internal concentration for seabirds. However a recent study suggested that HOC concentration was more representative of intake via prey than from transfer from plastic which is consistent with this study (Herzke et al., 2015).

Tanaka et al. (2015) also estimated the relative importance of the ingestion of plastic particles contaminated with PBDEs for transfer to bird tissues as compared from food alone. By using the measured desorption rate of PBDEs in stomach oil of 15% and the initial concentration of PBDE in plastic (5080 ng of BDE209) they estimated a leaching of 762 ng into the digestive fluid within the bird during 15 days, The relative contribution from the food source was estimated to be 11 ng of BDE209 and 164 ng of BDE47 through prey over 15 days. They concluded that for the congener BDE209, the relative contribution was more substantial from plastic ingestion (762 ng) than from the food source (11 ng). However the opposite trend was

estimated for the congener BDE47 (Tanaka et al., 2015). This was in agreement with our finding that relative contribution of plastic ingestion for the transfer of sorbed co-contaminants was plastic and pollutant specific.

The model used in the present study predicted a decrease in bioaccumulation due to the presence of plastics, which was very small, except for with Phe, where a 5% to 45% decrease was predicted with ingestion of PVC and PE respectively (Fig. 3). This result is in accordance with Koelmans et al. (2013) who also suggested that the decrease in bioaccumulation would be more substantial for plastics which had a high affinity for HOCs, such as PE (Koelmans et al., 2013). Such effect was also suggested by Herzke et al. (2015) indicating that microplastics can act as "negligible depletion" passive samplers for HOCs originating from ingested food.

Role of gut retention time for desorption of sorbed contaminants

The impact of particle retention time in the gut of the different organisms on the concentrations of desorbed contaminants was estimated using Eq.6. The ratio between contaminant concentrations on the plastic at the end of the particle retention time and the initial concentrations, are listed in Table 1. It is clear from the data shown in Table 1 that t_r was not long enough in all cases to allow complete desorption from plastics, even for a t_r of 158 hours for the fish. In other cases, a substantial amount could potentially desorb from the plastic particles. However, approach C, which is based on the instantaneous equilibrium assumption, indicated that other uptake and elimination routes were still more important in determining the body burdens of the organisms than the ingestion of plastic particles. Therefore, varying gut retention time appears to be of minor relevance in this context. However, microplastic particles have been shown to translocate from the gut and hence could

accumulate or persist in locations other than the gut. The potential toxicological effects of prolonged retention and associated desorption in other tissues, together with the potential for antagonistic effects caused by the physical presence of plastic particles may therefore warrant further investigation.

Predicted influence of plastic ingestion on internal concentration of sorbed contaminants

Predicted concentrations of contaminants transferred from plastics to marine organisms using the OMEGA model indicated a range from no impact through to increasing internal concentrations of contaminants in the lugworm, or a decrease for the fish and the seabird (Figs. 1-3, Tables S8-S10). These findings indicate that the body burden changes with plastic ingestion are partly dependent on the type of organism. The lack of effect may be more certain for higher trophic species, but due to the small impact of plastic observed in this study, it also depends on the model parameterization and the related uncertainties, and should therefore be interpreted with care.

Generally, an increase of the internal concentration occurred if the contribution of the uptake via plastic ingestion relative to other uptake routes was larger than the contribution of elimination through plastic egestion relative to other elimination routes. In the opposite direction the same holds for the predicted decreases in internal concentrations. The major (but not the only) factor explaining the differences between the lugworm and the other species was the difference in feeding strategy and in modelled food assimilation efficiencies (Hendriks et al., 2001a). Both Koelmans et al. (2013) and Gouin et al. (2011) predicted a small impact of plastic ingestion on the overall body burden of chemical (Gouin et al., 2011; Koelmans et al., 2013). Both studies predicted a decrease in bioaccumulation due to the counteraction of the biomagnification mechanism by the attenuation of the gradient between

plastics and lipids. However, predictions from our study have shown that any increase of decrease in internal concentration was small and should not be regarded as important given the intrinsic uncertainties of such modelling approaches. Herzke et al. (2015) reported a strong correlation between the PCBs and DDTs in ingested plastics and concentrations found in muscle tissue, considered as reflecting a long-term HOC exposure via food uptake. However bioaccumulation of HOCs was not found to be proportional to the quantity of plastic ingested, thus not supporting the suggestion that the presence of plastics in the environment might increase the accumulation of contaminants in marine organisms postulated by some other studies e.g. (Browne et al., 2013; Rochman et al., 2013; Teuten et al., 2007). This conclusion was also consistent with the present study which suggested negligible uptake of plastic co-contaminants from ingestion of plastic alone.

Environmental significance

Recent experimental data from laboratory studies has indicated that some sorbed chemicals can be transferred from microplastics to organisms at high concentrations and some biological effects have also been demonstrated (Browne et al., 2013; Rochman et al., 2013; Wright et al., 2013). However, the concentrations of plastics used in these experiments were typically high compared to those typically reported in the natural environment and hence there is uncertainty as to whether transfer of sorbed chemicals by microplastics is a quantitatively important route when compared to other pathways, such as respiratory or uptake from food. Calculations using mathematical models suggest that transfer of some sorbed hydrophobic organic pollutants (HOCs) from plastics is of limited importance compared to other routes of exposure Gouin et al. (2011; Koelmans et al. (2013). The present study also indicates that the predicted contribution of desorption from plastics to the overall

body burdens of sorbed HOCs in three marine organisms is probably small and that accumulation is dominated by uptake and elimination processes other than the presence of plastic. It has been estimated elsewhere that the flux of HOCs by ingestion of natural prey would be at least 21000 times higher than the flux of HOCs following plastic ingestion (Herzke et al., 2015); and that ingested plastic particles act as "passive samplers" due to their lipophilic behaviour explaining the correlation between concentrations of HOCs found in plastic and tissues of organisms (Herzke et al., 2015).

From the perspective of risk assessment for plastic debris, for example in the context of assessing harm associated with plastic debris for policy such as the MSFD, it may therefore be beneficial to focus on scenarios other than transport of HOCs. By contrast, the potential desorption from microplastics and subsequent bioavailability of chemicals which have been incorporated as additives during plastics manufacture, sometimes at high percentage concentrations (~ 80 % by weight in some polymers (Di Gangi, 1999; Kavlock et al., 2002)), has received much less attention in environmentally-relevant scenarios. Commonly used additives, such phthalate esters are found in many plastic products. These items are frequently reported in marine litter; however, it is unclear whether any substantive release of such additives from ingested plastic could occur into marine organisms. Therefore, bioaccumulation models such as that used herein, should also be applied to relate release rates of plastic additives to the tissues of organisms. This might permit predictions of additive concentrations in the tissues of organisms following ingestion of microplastics and allow investigation of any related toxicological effects in a similar manner to that conducted here for sorbed organic pollutants.

In addition to the potential for release of additive chemicals there is already some evidence that ingestion of relatively small quantities of microplastics (1% by weight in sediment) can cause physical harm by compromising the ability of deposit feeding worms to store energy.

There is also evidence that small particles (> 9.6 μm) may be able to pass from the digestive tract into the circulatory system, although it is not clear what the subsequent fate of these particles might be (Browne et al., 2008). If they accumulate in tissues or organs they may present as yet undescribed types of hazard. Moreover, there may be cumulative effects if plastic particles accumulate in tissues and subsequently release chemicals over longer timescales than would occur during gut transit. Further experimental work on very small particles including those into the nano-size range are therefore needed (GESAMP, 2015). Due to their smaller size and larger surface areas, desorption rates are expected to be much faster than for mm size particles (Koelmans et al., 2013).

Acknowledgements

A. Bakir was supported by the Department for Environment, Food and Rural Affairs (DEFRA) in the United Kingdom as part of a larger project investigating microplastics in the marine environment (Grant number ME 5416). We also gratefully acknowledge the financial support of the European Union to I. O'Connor through the Environmental ChemOinformatics (ECO) project (FP7-PEOPLE-ITN-2008, no.238701).

References

Avery-Gomm, S., O'Hara, P.D., Kleine, L., Bowes, V., Wilson, L.K., Barry, K.L., 2012. Northern fulmars as biological monitors of trends of plastic pollution in the eastern North Pacific. Marine Pollution Bulletin 64, 1776-1781.

Bakir, A., Rowland, S.J., Thompson, R.C., 2014. Enhanced desorption of persistent organic pollutants from microplastics under simulated physiological conditions. Environmental Pollution 185, 16-23.

Besseling, E., Wegner, A., Foekema, E.M., van den Heuvel-Greve, M.J., Koelmans, A.A., 2012. Effects of Microplastic on Fitness and PCB Bioaccumulation by the Lugworm Arenicola marina (L.). Environmental Science & Technology 47, 593-600.
Blight, L.K., Burger, A.E., 1997. Occurrence of plastic particles in seabirds from the eastern North Pacific. Marine Pollution Bulletin 34, 323-325.
Boerger, C.M., Lattin, G.L., Moore, S.L., Moore, C.J., 2010. Plastic ingestion by planktivorous fishes in the North Pacific Central Gyre. Marine Pollution Bulletin 60, 2275-2278.
Browne, M.A., Dissanyake, A., Galloway, T.S., Lowe, D., Thompson, R.C., 2008. Ingested microplastic translocates to the circulatory system of the mussel, Mytilus edulis (L.). Environ.Sci.Technol. 42, 5026-5031.
Browne, M.A., Galloway, T.S., Thompson, R.C., 2010. Spatial patterns of plastic debris along estuarine shorelines. Environmental Science & Technology 44, 3404-3409.
Browne, Mark A., Niven, Stewart J., Galloway, Tamara S., Rowland, Steve J., Thompson, Richard C., 2013. Microplastic Moves Pollutants and Additives to Worms, Reducing Functions Linked to Health and Biodiversity. Current Biology 23, 2388-2392.
Carson, H.S., Colbert, S.L., Kaylor, M.J., McDermid, K.J., 2011. Small plastic debris changes water movement and heat transfer through beach sediments. Marine Pollution Bulletin 62, 1708-1713.
Chang, B.V., Liao, C.S., Yuan, S.Y., 2005. Anaerobic degradation of diethyl phthalate, di-n-butyl phthalate, and di-(2-ethylhexyl) phthalate from river sediment in Taiwan. Chemosphere 58, 1601-1607.
Denuncio, P., Bastida, R., Dassis, M., Giardino, G., Gerpe, M., Rodríguez, D., 2011. Plastic ingestion in Franciscana dolphins, Pontoporia blainvillei (Gervais and d'Orbigny, 1844), from Argentina. Marine Pollution Bulletin 62, 1836-1841.
Di Gangi, J., 1999. Phthalates in PVC medical products from 12 countries. Washington, DC: Greenpeace.
Endo, S., Yuyama, M., Takada, H., 2013. Desorption kinetics of hydrophobic organic contaminants from marine plastic pellets. Marine Pollution Bulletin 74, 125-131.
Engler, R.E., 2012. The Complex Interaction between Marine Debris and Toxic Chemicals in the Ocean. Environmental Science & Technology 46, 12302-12315.
Foekema, E.M., De Gruijter, C., Mergia, M.T., van Franeker, J.A., Murk, A.J., Koelmans, A.A., 2013. Plastic in North Sea Fish. Environmental Science & Technology 47, 8818-8824.
GESAMP, 2015. Sources, fate and effects of microplastics in the marine environment: a global assessment, in: Kershaw, P.J. (Ed.), p. 96 p.
Goldstein, M.C., Rosenberg, M., Cheng, L., 2012. Increased oceanic microplastic debris enhances oviposition in an endemic pelagic insect. Biology Letters.
Gouin, T., Roche, N., Lohmann, R., Hodges, G., 2011. A thermodynamic approach for assessing the environmental exposure of chemicals absorbed to microplastic. Environmental Science & Technology 45, 1466-1472.
Graham, E.R., Thompson, J.T., 2009. Deposit- and suspension-feeding sea cucumbers (Echinodermata) ingest plastic fragments. Journal of Experimental Marine Biology and Ecology 368, 22-29.
Guruge, K.S., Watanabe, M., Tanaka, H., Tanabe, S., 2001. Accumulation status of persistent organochlorines in albatrosses from the North Pacific and the Southern Ocean. Environmental Pollution 114, 389-398.
Hendriks, A.J., Traas, T.P., Huijbregts, M.A.J., 2005. Critical body residues linked to octanol - Water partitioning, organism composition, and LC50 QSARs: Meta-analysis and model. Environmental Science and Technology 39, 3226-3236.
Hendriks, A.J., van der Linde, A., Cornelissen, G., Sijm, D.T., 2001a. The power of size. 1. Rate constants and equilibrium ratios for accumulation of organic substances related to octanol-water partition ratio and species weight. Environmental Toxicology and Chemistry 20, 1399-1420.

Hendriks, A.J., Van der Linde, A., Cornelissen, G., Sijm, D.T.H.M., 2001b. The power of size. 1. Rate constants and equilibrium ratios for accumulation of organic substances related to octanol-water partition ratio and species weight. Environmental Toxicology and Chemistry 20, 1399-1420.
Herzke, D., Anker-Nilssen, T., Nøst, T.H., Götsch, A., Christensen-Dalsgaard, S., Langset, M., Fangel, K., Koelmans, A.A., 2015. Negligible Impact of Ingested Microplastics on Tissue Concentrations of Persistent Organic Pollutants in Northern Fulmars off Coastal Norway. Environmental Science & Technology.
Huang, P.-C., Tien, C.-J., Sun, Y.-M., Hsieh, C.-Y., Lee, C.-C., 2008. Occurrence of phthalates in sediment and biota: Relationship to aquatic factors and the biota-sediment accumulation factor. Chemosphere 73, 539-544.
Institute, S.E.R., 2009. Screening of new contaminants in samples from the Norwegian Arctic. Silver, Platinum, Sucralose, Bisphenol A, Tetrabrombisphenol A, Siloxanes, Phtalates (DEHP) and Phosphororganic flame retardants.
Kavlock, R., Boekelheide, K., Chapin, R., Cunningham, M., Faustman, E., Foster, P., Golub, M., Henderson, R., Hinberg, I., Little, R., 2002. NTP center for the evaluation of risks to human reproduction: phthalates expert panel report on the reproductive and developmental toxicity of di (2-ethylhexyl) phthalate. Reproductive toxicology 16, 529-653.
Koelmans, A.A., Bakir, A., Burton, G.A., Janssen, C.R., 2016. Microplastic as a vector for chemicals in the aquatic environment: critical review and model-supported reinterpretation of empirical studies. Environmental Science & Technology 50, 3315-3326.
Koelmans, A.A., Besseling, E., Foekema, E.M., 2014. Leaching of plastic additives to marine organisms. Environmental Pollution 187, 49-54.
Koelmans, A.A., Besseling, E., Wegner, A., Foekema, E.M., 2013. Plastic as a carrier of POPs to aquatic organisms: a model analysis. Environmental Science & Technology 47, 7812-7820.
Kühn, S., van Franeker, J.A., 2012. Plastic ingestion by the northern fulmar (< i> Fulmarus glacialis</i>) in Iceland. Marine Pollution Bulletin 64, 1252-1254.
Lusher, A.L., McHugh, M., Thompson, R.C., 2013. Occurrence of microplastics in the gastrointestinal tract of pelagic and demersal fish from the English Channel. Marine Pollution Bulletin 67, 94-99.
Mallory, M.L., 2008. Marine plastic debris in northern fulmars from the Canadian high Arctic. Marine Pollution Bulletin 56, 1501-1504.
Mallory, M.L., Roberston, G.J., Moenting, A., 2006. Marine plastic debris in northern fulmars from Davis Strait, Nunavut, Canada. Marine Pollution Bulletin 52, 813-815.
Murray, F., Cowie, P.R., 2011. Plastic contamination in the decapod crustacean Nephrops norvegicus (Linnaeus, 1758). Marine Pollution Bulletin 62, 1207-1217.
Napper, I.E., Bakir, A., Rowland, S.J., Thompson, R.C., 2015. Characterisation, quantity and sorptive properties of microplastics extracted from cosmetics. Marine Pollution Bulletin 99, 178-185.
O'Connor, I.A., Huijbregts, M.A., Ragas, A.M., Hendriks, A.J., 2013a. Predicting the oral uptake efficiency of chemicals in mammals: Combining the hydrophilic and lipophilic range. Toxicology and applied pharmacology 266, 150-156.
O'Connor, I.A., Huijbregts, M.A.J., Ragas, A.M.J., Hendriks, A.J., 2013b. Predicting the oral uptake efficiency of chemicals in mammals: Combining the hydrophilic and lipophilic range. Toxicology and Applied Pharmacology 266, 150-156.
Organization, W.H., 1992. International programme on chemical safety "diethylhexyl phthalate", Environmental Health Criteria 131, Geneva.
Persson, P.-E., Penttinen, H., Nuorteva, P., 1978. DEHP in the vicinity of an industrial area in Finland. Environmental Pollution (1970) 16, 163-166.
Ray, L.E., Murray, H., Giam, C., 1983. Organic pollutants in marine samples from Portland, Maine. Chemosphere 12, 1031-1038.

Rochman, C.M., Hoh, E., Kurobe, T., Teh, S.J., 2013. Ingested plastic transfers hazardous chemicals to fish and induces hepatic stress. Scientific reports 3.
Ryan, P.G., 1990. The effects of ingested plastic and other marine debris on seabirds. NOAA Technical Memorandum.
Ryan, P.G., Connell, A.D., Gardner, B.D., 1988. Plastic ingestion and PCBs in seabirds: Is there a relationship? Marine Pollution Bulletin 19, 174-176.
Tanaka, K., Takada, H., Yamashita, R., Mizukawa, K., Fukuwaka, M.-a., Watanuki, Y., 2013. Accumulation of plastic-derived chemicals in tissues of seabirds ingesting marine plastics. Marine Pollution Bulletin.
Tanaka, K., Takada, H., Yamashita, R., Mizukawa, K., Fukuwaka, M.-a., Watanuki, Y., 2015. Facilitated Leaching of Additive-Derived PBDEs from Plastic by Seabirds' Stomach Oil and Accumulation in Tissues. Environmental Science & Technology 49, 11799-11807.
Teuten, E.L., Rowland, S.J., Galloway, T.S., Thompson, R.C., 2007. Potential for Plastics to Transport Hydrophobic Contaminants. Environmental Science & Technology 41, 7759-7764.
Thompson, R.C., Olsen, Y., Mitchell, R.P., Davis, A., Rowland, S.J., John, A.W., McGonigle, D., Russell, A.E., 2004. Lost at sea: where is all the plastic? Science 304, 838-838.
van Franeker, J.A., 1985. Plastic ingestion in the North Atlantic fulmar. Marine Pollution Bulletin 16, 367-369.
van Franeker, J.A., Bell, P.J., 1988. Plastic ingestion by petrels breeding in Antarctica. Marine Pollution Bulletin 19, 672-674.
van Franeker, J.A., Blaize, C., Danielsen, J., Fairclough, K., Gollan, J., Guse, N., Hansen, P.-L., Heubeck, M., Jensen, J.-K., Le Guillou, G., 2011a. Monitoring plastic ingestion by the northern fulmar< i> Fulmarus glacialis</i> in the North Sea. Environmental Pollution 159, 2609-2615.
van Franeker, J.A., Blaize, C., Danielsen, J., Fairclough, K., Gollan, J., Guse, N., Hansen, P.-L., Heubeck, M., Jensen, J.-K., Le Guillou, G., Olsen, B., Olsen, K.-O., Pedersen, J., Stienen, E.W.M., Turner, D.M., 2011b. Monitoring plastic ingestion by the northern fulmar Fulmarus glacialis in the North Sea. Environmental Pollution 159, 2609-2615.
van Sebille, E., Wilcox, C., Lebreton, L., Maximenko, N., Hardesty, B.D., van Franeker, J., Eriksen, M., Siegel, D., Galgani, F., Lavender, L.K., 2015. A global inventory of small floating plastic debris. Environmental Research Letters 10, 124006.
Walker, C., Hopkin, S., Sibly, R., Peakall, D., 2001. Toxicity testing. Principles of ecotoxicology. 2nd edition. Taylor & Francis, 93-118.
Ward, E.J., Shumway, S.E., 2004. Separating the grain from the chaff: particle selection in suspension- and deposit-feeding bivalves. Journal of Experimental Marine Biology and Ecology 300, 83-130.
Wright, S.L., Rowe, D., Thompson, R.C., Galloway, T.S., 2013. Microplastic ingestion decreases energy reserves in marine worms. Current Biology 23, R1031-R1033.
Yuan, S.Y., Liu, C., Liao, C.S., Chang, B.V., 2002. Occurrence and microbial degradation of phthalate esters in Taiwan river sediments. Chemosphere 49, 1295-1299.

Table 1. Evaluation of particle retention time (tr) on desorption of sorbed contaminants estimated using Eq.6

organism	POP	Cx (ng/g), plastic (t=tr)/Cx (ng/g), plastic (t=0)	
		PE	PVC
Lugworm - tr 2 hours	DDT	0.9	1.0
	Phe	0.8	0.9
	DEHP	1.0	0.9
Fish - tr 4 hours	DDT	0.8	0.9
	Phe	0.6	0.8
	DEHP	1.0	0.8
Fish tr 158 hours	DDT	1.6E-05	0.13
	Phe	2.6E-09	1.7E-05
	DEHP	1.7E-01	1.2E-04
Seabird - tr 11 hours	DDT	0.04	0.8
	Phe	0.004	0.1
	DEHP	0.2	0.1

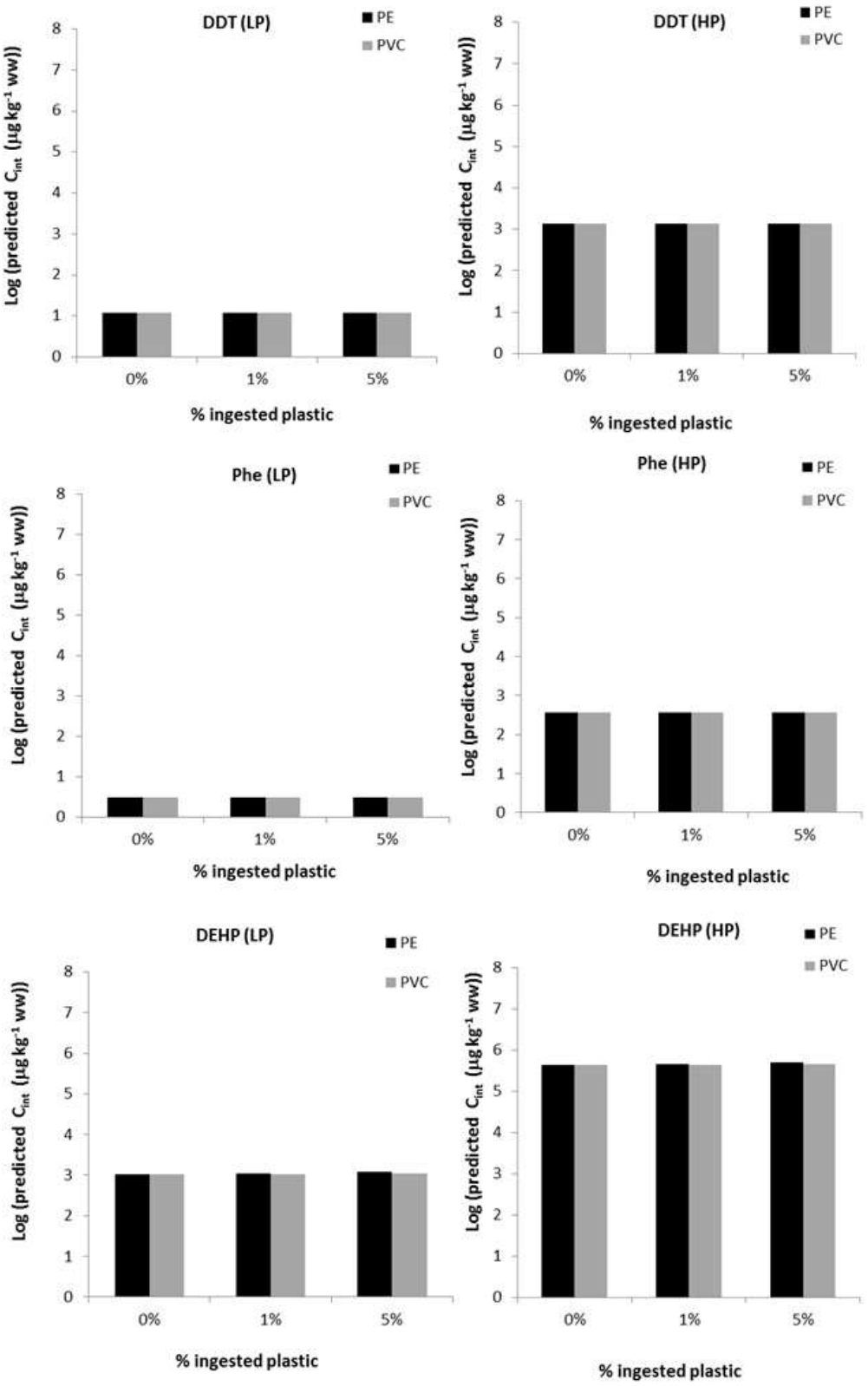

Fig. 1 Predicted internal concentration (Cint) in µg kg^{-1} ww of DDT (top), Phe (middle) and DEHP (bottom) in the tissues of a lugworm ingesting PE (black) or PVC (grey) particles in the low pollution (LP) and high pollution (HP) scenarios.

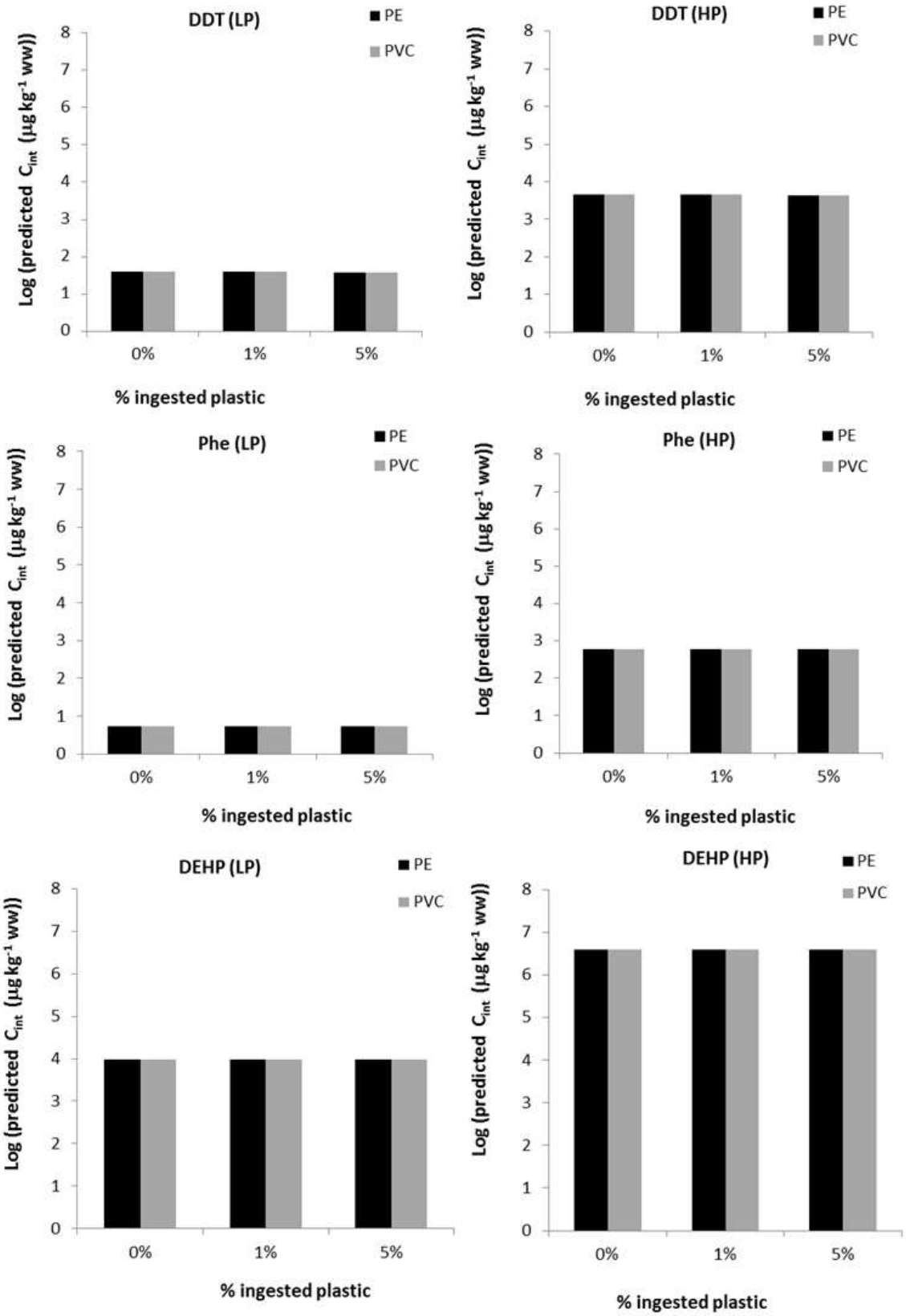

Fig. 2 Predicted internal concentration (Cint) in µg kg-1 ww of DDT (top), Phe (middle) and DEHP (bottom) in the tissues of a marine fish ingesting PE (black) or PVC (grey) particles in the low pollution (LP) and high pollution (HP) scenarios.

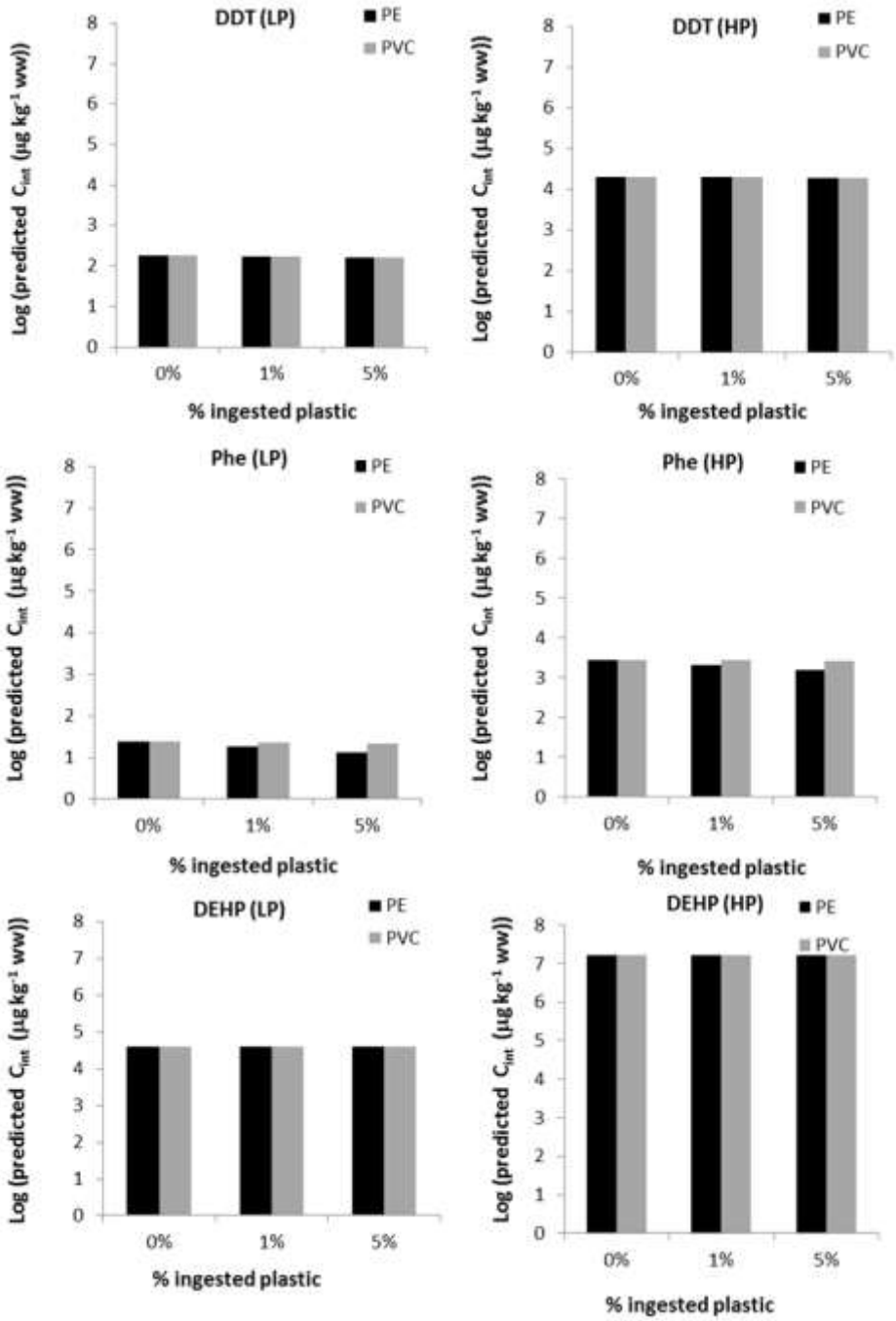

Fig. 3 Predicted concentration (Cint) in µg kg-1 ww of DDT (top), Phe (middle) and DEHP (bottom) in the tissues of a seabird ingesting PE (black) or PVC (grey) particles in the low pollution (LP) and high pollution (HP) scenarios.

Relative importance of microplastics as a pathway for the transfer of hydrophobic organic chemicals to marine life

Supporting information

Adil Bakir [ab], *Isabel A. O'Connor* [c], *Steven J. Rowland* [b], *A. Jan Hendricks* [c] *and Richard C. Thompson* [a]

[a] *Marine Biology and Ecology Research Centre (MBERC) School of Marine Science and Engineering, University of Plymouth, Drake Circus, Plymouth, Devon, PL4 8AA, United Kingdom*

[b] *Petroleum and Environmental Geochemistry Group (PEGG) School of g, University of Plymouth, Drake Circus, Plymouth, Devon, PL4 8AA, United Kingdom*

[c] *Radbound University Nijmegen, Institute for water and Wetland research, Department of Environmental Science, P.O. Box 9010, NL-6500 GL, Nijmegen, The Netherlands*

Table S1. Reported DDT, phenanthrene (Phe) and DEHP concentrations for low and high polluted sites and used in this study.

POP	Log Kow	Reference	HOC concentration (μg/L)		Reference	
			Low	High		
DDT	6.36	Walker (2008)	0.0002	0.023	(Wurl and Obbard, 2005)	
Phe	4.5	MacKay et al. (1993)	0.00324	0.377	(Law et al., 1997)	(Pérez-Carrera et al., 2007)
DEHP	7.5	Neff (2002)	0.0053	2.2	(Xie et al., 2005)	(Matthiessen et al., 1993)

Table S2. (A) Reported amounts of plastic debris collected from a range of marine organisms and (B) reported gut retention time for sediments/anthropogenic particles

(A)

Organism	Plastic stomach content (g)	Sampling year	References
Planktivorous fish	0.00157	2008	(Boerger et al., 2010)
Harbour seals	24.4	2001-2002 2009-2010	(Bravo Rebolledo et al., 2012)
Cape Petrel	0.0201	1984-1987	(van Franeker and Bell, 1988)
Southern fulmar	0.0106	1984-1987	(van Franeker and Bell, 1988)
Seabirds	0.30	1987	(Blight and Burger, 1997)
Northern fulmar	0.280	2003-2007	(van Franeker et al., 2011b)
Northern fulmars	0.385	2009-2010	(Avery-Gomm et al., 2012)

(B)

Organism	Particle type	Gut retention time (hours)	References
Arenicola marina	sediment	2	(Bock and Miller, 1999) (Chen and Mayer, 1999)
Fish	food	4-158	(Fänge and Grove, 1979)
Seabird	food	11	(Hilton et al., 2000)

Table S3. Reported DDTs concentrations for a range of marine organisms

Marine organism		Sampling location	Sampling time	DDT concentration ($\mu g\ kg^{-1}$ wet weight)		References
Classification	Species			p,p'-DDE	p,p'-DDT	
Polychaeta	*Dendronereis spp.*	Malay Pinsula	1985	69-71	n.s	(Everaarts et al., 1991)
	Arenicola marina	Dutch Wadden Sea	1979	60-160	n.s	(Duinker et al., 1983)
	Diopatra ornate *Pista alata*	Coastal ocean, Southern California	1995	13517*	< 2.3	(Zeng and Tran, 2002)
	Paraprionospio pinnata		1996	14798*	< 9.2	
Mediterranean mussel	*Mitylus galloprovencialis*	Italian coasts	2002	0.19-1.49	nd-0.24	(Perugini et al., 2004)
Norway lobster	*Nephrops norvegicus*			0.10-0.36	nd-0.18	
Red mullet	*Mullus barbatus*			0.31-2.23	nd-0.58	
European flying squid	*Sepia officinalis*			nd-0.39	nd-0.11	
Common cuttle-fish	*Totarodes sagitattus*			nd	nd	
European anchovy	*Engraulis encrasicholus*			nd-0.30	nd-0.66	
European pilchard	*Sardina pilchardus*			0.33-2.58	nd-0.36	
Atlantic mackerel	*Scomber scombrus*			0.01-2.76	nd-0.51	
Bartail flathead	*Platycephalus indicus*	South China	2004	19.5	8.40	(Cheung et al., 2007)
Snubnose poampano	*Trachinotus blochii*			244	133	
Goldspotted rabbitfish	*Siganus punctatus*			10.7	n.d.	
Tongue sole	*Cynoglossus robustus*			12.6	n.d	
Northern Fulmar	*Procellariidae*	Northern Baffin Bay	1998	3093 (fat) 149 (liver)	360 (fat) 10.1 (liver)	(Buckman et al., 2004)
Kelp gull **eggs**	*Larus dominicanus*	Maiquillahue Bay	1998-1999	151 (∑ DDTs)		(Cifuentes et al., 2003)
Pink-footed Shearwater **eggs**	*Puffinus creatopus*	Juan Fernandez Islands		163.1 (∑ DDTs)		
Black-footed albatross		North Pacific	1992-1993		35.5	(Auman et al., 1997)
Laysan albatross					11.5	
Black-footed albatross			1997-1998	13000-73000	1200-4400	(Guruge et al., 2001)
Laysan albatross				190-1400	3200-9500	

nd: not detected

Table S4. Reported phenanthrene (Phe) concentrations for a range of marine organisms

Marine organism		Sampling location	Sampling time	Phe concentration ($\mu g\ kg^{-1}$ wet weight)	References
Classification	Species				
Marine mollusc	*Mytilus galloprovinclialis*	Western Mediterranean sea	2004-2006	4.44 (dry wt)	(Galgani et al., 2011)
Marine fish	Doma	Mumbai transharbour, Maharashtra	2006-2008	0.61	(Dhananjayan and Muralidharan, 2012)
	Mandeli			0.99	
	Brown spotted grouper	Arabian Gulf	1997	1.3	(Al-Hassan et al., 2003)
	Yellow finned black Sea bream			0.86	
	River Shad			1.07	
	Silvery Grunt			0.94	
	Silvery Pomfret			1.31	
	Scomberomorus commerson	Western coast of Alexandria	2005	0.608	(Said, 2007)
	Sphyraena sphyraena			338.76	

Table S5. Reported DEHP concentrations for a range of marine organisms

Marine organism		Sampling location	Sampling time	DEHP concentration (μg kg^{-1} wet weight)	Reference
Classification	Species				
Polychaete	*Neanthes virens*	Portland, Maine, USA	1980	380-490	(Ray et al., 1983b)
clams	ns			110-170	
Fish	Various fish species (liver)	Tees Bay, United Kingdom	ns	43-85.9	(Waldock, 1983)
	Various fish species (muscle)			13-51.3	

n.s. not specified

Bioaccumulation models

Approach A:

The concentration of the pollutant in each species *i* (C_i in μg kg^{-1}) was calculated with the assumption that an individual ingesting and egesting sufficient plastic over its life time to reach equilibrium between lipid and plastic debris, Eq.S1:

$$C_i = \frac{C_p}{K_{pw}} \cdot K_{ow} \cdot p_{lip} \quad \textbf{(Eq. S1)}$$

where C_p = concentration of the pollutant in plastic (μg kg^{-1})

K_{pw} = plastic water partition coefficient

K_{ow} = octanol water partition coefficient

p_{lip} = lipid fraction of bird (kg kg^{-1})

Approach B:

The concentration of the pollutant in species *i* (C_i in μg kg^{-1}), that species *i* would achieve after a lifelong plastic ingestion was calculated with the assumption that all pollutants sorbed onto plastic were transferred to the organisms using Eq.S2:

$$C_i = k_{p,in} \cdot a \cdot C_p \quad \textbf{(Eq. S2)}$$

where $k_{p,in}$ = plastic ingestion rate (kg kg^{-1} d^{-1})

a = lifespan of species *i* (d^{-1})

Allometric regressions suggested a lifespan of 18 years and 398 days for the seabird and the fish, respectively (Hendriks, 2007; Lindstedt and Calder III, 1981), which was in agreement with reported lifespans for species of similar weights (Beukema and De Vlas, 1979; Botkin and Miller, 1974; Hennicke et al., 2012). For the lifespan of the lugworm, the reported value of 6 years was used (Beukema and De Vlas, 1979).

Plastic ingestion rate was calculated as Eq.S3:

$$k_{p,in} = \gamma_p \cdot q_T \cdot w^{-\kappa} \quad \textbf{(Eq. S3)}$$

where γ_p = Plastic ingestion coefficient (kg^K d^{-1})

q_T = Temperature correction factor (kg kg^{-1})

w = Species weight (kg)

κ = rate coefficient (-)

It should be noted that Eq.S1 gives a theoretical limit to Eq.S2, as no additional pollutants will be absorbed from the plastic after equilibrium between the lipid fraction of the species and the plastic is reached. Approach B should therefore help to indicate whether lifelong plastic ingestion could be sufficient to reach equilibrium between the plastic and the species.

Approach C:

In approach C, we applied the OMEGA model to calculate internal concentration of pollutants in species of different trophic levels such as a seabird, a fish and a lugworm. The

model is described in detail in Hendriks et al. (2001)(Hendriks et al., 2001b). Here, we only describe briefly the relevant processes used for this study.

The internal concentration of species *i* (C_i) at steady state equals the ratio of the sum of uptake divided by the sum of elimination. In this study, we considered uptake from water ($k_{w,X,in} \cdot C_w$), from food ($k_{f,X,in} \cdot C_{food}$) and from plastic ($k_{p,X,in} \cdot C_p$) as well as the elimination with water ($k_{w,X,out}$), food ($k_{f,X,out}$), plastic ($k_{p,X,out}$) and biomass dilution from growth or reproduction ($k_{b,X,out}$) as shown in Eq.S4:

$$C_i = \frac{k_{w,X,in} \cdot C_w + k_{f,X,in} \cdot C_{food} + k_{p,X,in} \cdot C_p}{\Sigma k_{w,X,out} + k_{f,X,out} + k_{p,X,out} + k_{b,X,out}} \quad \textbf{(Eq.S4)}$$

In this study, we neglected the uptake from air as well as elimination by metabolism. The uptake and excretion via water was calculated using Eqs.S5 and S6 (see Table S6 for the definition of the symbols):

$$k_{w,X,in} = \frac{w^{-k}}{\rho_{H_2O,w} + \frac{\rho_{CH_2,i}}{K_{ow}} + \frac{1}{\gamma_w}} \quad \textbf{(Eq. S5)}$$

$$k_{w,X,out} = \frac{1}{p_{lip,i} \cdot (K_{ow} - 1) + 1} \cdot \frac{w^{-k}}{\rho_{H_2O,w} + \frac{\rho_{CH_2,i}}{K_{ow}} + \frac{1}{\gamma_w}} \quad \textbf{(Eq. S6)}$$

The uptake from food and excretion with faeces were modeled using Eqs. S7 and S8 (see Table S6 for definition of the symbols):

$$k_{f,X,in} = \frac{1}{1 - p_f} \cdot \frac{1}{p_{lip,i-1} \cdot (K_{ow} - 1) + 1} \cdot \frac{w^{-k}}{\rho_{H_2O,f} + \frac{\rho_{CH_2,i}}{K_{ow} \cdot q_T} + \frac{1}{p_{lip,i-1} \cdot K_{ow} \cdot (1 - p_f) \cdot \gamma_f \cdot q_T}} \quad \textbf{(Eq. S7)}$$

$$k_{f,X,out} = \frac{1}{p_{lip,i} \cdot (K_{ow} - 1) + 1} \cdot \frac{w^{-k}}{\rho_{H_2O,f} + \frac{\rho_{CH_2,i}}{K_{ow} \cdot q_T} + \frac{1}{p_{lip,i-1} \cdot K_{ow} \cdot (1 - p_f) \cdot \gamma_f \cdot q_T}} \quad \textbf{(Eq. S8)}$$

The elimination by biomass dilution was modeled as followed (see Table S6 for definition of the symbols):

$$k_{b,X,out} = \gamma_b \cdot q_T \cdot w^{-k} \quad \textbf{(Eq. S9)}$$

Plastic was treated like undigested food, such that uptake and excretion of the pollutant with plastic was estimated using Eq. S10 and Eq. S11:

$$k_{p,X,in} = \frac{1}{1-p_p} \cdot \frac{1}{K_{pw}} \cdot \frac{w^{-k}}{\rho_{H_2O,f} + \frac{\rho_{CH_2,i}}{K_{ow} \cdot q_T} + \frac{1}{K_{pw} \cdot (1-p_p) \cdot \gamma_p \cdot q_T}} \quad \textbf{(Eq. S10)}$$

$$k_{p,X,out} = \frac{1}{p_{lip,i} \cdot (K_{ow}-1)+1} \cdot \frac{w^{-k}}{\rho_{H_2O,f} + \frac{\rho_{CH_2,i}}{K_{ow} \cdot q_T} + \frac{1}{K_{pw} \cdot (1-p_p) \cdot \gamma_p \cdot q_T}} \quad \textbf{(Eq. S11)}$$

The pollutant concentration in food was estimated using a standard food chain bioaccumulation model. It was assumed, that the seabird (trophic level 4) feeds on the marine fish species (trophic level 3), which in turn feeds on zooplankton feeding on phytoplankton. The potential transfer of plastic debris within the food chain was neglected. Lugworms are detritivores that feed on organic carbon contained in soil (typical marine sediment with 1% of organic carbon). The pollutant concentration in wet organic matter in sediment representing C_{food} was calculated using Eq. S12:

$$C_{food} = K_{wom} \cdot C_w \quad \textbf{(Eq. S12)}$$

And the wet organic matter- water partition coefficient was calculated assuming that wet organic matter contains 90% water, and 50% of the dry organic matter consists of organic carbon (Eq. S13):

$$K_{wom} = 0.1 \cdot 0.5 \cdot K_{oc} \quad \textbf{(Eq. S13)}$$

92 Table S6. Overview of processes, rates and parameters used in the model

Symbol	Description	Unit	Typical value	Reference
a	Lifespan	d	Table S7	
C_w	Concentration in water	µg L^{-1}	variable	
C_{food}	Concentration in food	µg kg^{-1}	variable	
C_p	Concentration in plastic	µg kg^{-1}	variable	
C_i	Concentration in organism	µg kg^{-1}	Eq. S1	Hendriks et al., 2001
γ_w	water absorption-excretion coefficient			
	water breathing	$kg^K\ d^{-1}$	200	Hendriks et al., 2001
	air breathing	$kg^K\ d^{-1}$	0.2	Hendriks et a.l, 2001
γ_f	Food ingestion coefficient	$kg^K\ d^{-1}$	0.005	Hendriks et al., 2001
γ_b	Biomass (re)production coefficient	$kg^K\ d^{-1}$	0.0006	Hendriks et al., 2001
γ_p	Plastic ingestion coefficient	$kg^K\ d^{-1}$	0.01 . γ_f 0.05 · γ_f 0.50 . γ_f	This study
$k_{w,X,in}$	Rate constant for pollutant absorption from water	L $kg^{-1}\ d^{-1}$	Eq. S5	Hendriks et al., 2001
$k_{f,X,in}$	Rate constant for pollutant absorption from food	kg $kg^{-1}\ d^{-1}$	Eq. S7	Hendriks et al., 2001
$k_{p,X,in}$	Rate constant for pollutant absorption from plastic	kg $kg^{-1}\ d^{-1}$	Eq. S10	Hendriks et al., 2001
$k_{w,X,out}$	Rate constant for pollutant excretion with water	d^{-1}	Eq. S6	Hendriks et al., 2001
$k_{f,X,out}$	Rate constant for pollutant excretion with egestion	d^{-1}	Eq. S8	Hendriks et al., 2001
$k_{p,X,out}$	Rate constant for pollutant excretion with water		Eq. S11	Hendriks et al., 2001
$k_{b,X,out}$	Rate constant for biomass dilution by growth or reproduction	d^{-1}	Eq. S9	Hendriks et al., 2001
K_{oc}	Organic carbon water partition coefficient	[-]	variable	KocWin, Episuite (EPA, 2013)
K_{ow}	Octanol-water partition coefficient	[-]	variable	
K_{pw}	Plastic-water partition coefficient	[-]	Variable	Bakir et al., 2014
	Log Kpw Phe-PVC		3.36	
	Log Kpw Phe-PE		4.71	
	Log Kpw DDT-PVC		5.02	
	Log Kpw DDT-PE		4.99	
	Log Kpw DEHP-PVC		4.08	
	Log Kpw DEHP-PE		4.99	
$K_{sw\ om}$	Wet organic matter water partition coefficient	[-]	Eq. S13	
κ	Rate exponent	[-]	0.25	Hendriks et al., 2001
p_{oc}	Fraction of organic carbon in soil	kg kg^{-1}	0.01	Kile et al., 1995
p_f	Fraction of food assimilated			
	Herbivore	kg kg^{-1}	0.4	Hendriks et al., 2001
	carnivore	kg kg^{-1}	0.8	Hendriks et al., 2001
p_p	Fraction of plastic assimilated	kg kg^{-1}	0	Assumption in this study
$P_{lip,i}$	Fraction of neutral lipid in organism (i) or in food (i-1)	kg kg^{-1}	Table S7	
q_T	Temperature correction factor			
	Cold-blood	kg kg^{-1}	1	Hendriks et al., 2001
	Warm-blooded	kg kg^{-1}	10	Hendriks et al., 2001
$\rho_{CH2,i}$	Lipid layer resistance			
	Plants	d kg^{-1}	$4.6 \cdot 10^3$	Hendriks et al., 2001
	Animals	d kg^{-1}	68	Hendriks et al., 2001
$\rho_{H20,w}$	Water layer resistance from/to water	d kg^{-K}	$2.8 \cdot 10^{-3}$	Hendriks et al., 2001
$\rho_{H20,f}$	Water layer resistance from/to food	d kg^{-K}	$1.1 \cdot 10^{-5}$	Hendriks et al., 2001
X	Substance	[-]		
w_i	Species weight	kg	Table S7	

93 N.B. K_{oc} was calculated using KocWin v2.00 in Episuite. Estimation Programs Interface Suite™ for Microsoft® Windows, v 4.1 (EPA, 2013).

94

Table S7. Characteristics of the species used in the model. Reported are their trophic level (TL), mass (w, in kg), lifespan (a, in days) and their lipid content ($p_{lip,i}$). Also listed are their food source, and whether the species was a target species (TS) in the model or a species in the food chain (FC).

Species	**Seabird**	**Marine fish**	**Zooplankton**	**Phytoplankton**	**Lugworm**
TL	4	3	2	1	2
w	1	0.0075	10^{-6}	10^{-12}	0.004
a	6424[a]	398[b]	-	-	2190[c]
P_{lip}	0.1[d]	0.05[d]	0.03[d]	0.01[d]	0.03[d]
Food	TL 3 (fish)	TL 2 (Zooplankton)	TL 1 (phytoplankton)	none	Organic carbon in soil
Role	TS	TS and FC	FC	FC	TS

[a] (Lindstedt and Calder III, 1981; Hennicke et al., 2012; Botkin and Miller, 1974)

[b] (Hendricks, 2007; Catul et al., 2011)

[c] e.g. (Beukema and De Vlas, 1979)

[d] sum of neutral and polar lipid (Hendricks et al., 2005)

Model output:

Table S8. Estimated concentration of DDT, phenanthrene (Phe) and DEHP in the lugworm *Arenicola marina* following ingestion of contaminated PE and PVC for scenarios A, B and C.

			PE			
			[DDT] in μg kg^{-1} ww			
Lugworm	low pollution			high pollution		
Approaches	1%	5%	50%	1%	5%	50%
A	13.75	13.75	13.75	1581	1581	1581
B	10.97	42.19	422	1261	4852	48516
C (incl. plastic)	11.77	11.92	12.80	1354	1371	1472
C (excl. plastic)	11.71	11.71	11.71	1347	1347	1347
			[Phe] in μg kg^{-1} ww			
A	3.07	3.07	3.07	358	358	358
B	94.51	363	3635	10997	42295	422948
C (incl. plastic)	3.09	3.09	3.08	360	360	359
C (excl. plastic)	3.09	3.09	3.09	360	360	360
			[DEHP] in μg kg^{-1} ww			
A	5028	5028	5028	2087103	2087103	2087103
B	295	1136	11365	122652	471739	4717388
C (incl. plastic)	1090	1187	2070	452441	492708	859370
C (excl. plastic)	1055	1055	1055	437801	437801	437801
			PVC			
			[DDT] in μg kg^{-1} ww			
Approaches	1%	5%	50%	1%	5%	50%
A	13.75	13.75	13.75	1581	1581	1581
B	11.86	45.62	456	1364	5247	52468
C (incl. plastic)	11.77	11.94	12.84	1354	1373	1476
C (excl. plastic)	11.71	11.71	11.71	1347	1347	1347
			[Phe] in μg kg^{-1} ww			
A	3.07	3.07	3.07	358	358	358
B	4.19	16.12	161	488	1875	18754
C (incl. plastic)	3.09	3.09	3.09	360	360	360
C (excl. plastic)	3.09	3.09	3.09	360	360	360
			[DEHP] in μg kg^{-1} ww			
A	5028	5028	5028	2087103	2087103	2087103
B	35.75	138	1375	14840	57077	570767
C (incl. plastic)	1059	1071	1214	439586	444647	503768
C (excl. plastic)	1055	1055	1055	437801	437801	437801

Table S9. Estimated concentration of DDT, phenanthrene (Phe) and DEHP in a marine fish following ingestion of contaminated PE and PVC for scenarios A, B and C. Highlighted in red are examples of the neutral transfer of plastic co-contaminants following ingestion.

			PE			
			[DDT] in µg kg^{-1} ww			
Fish	low pollution			high pollution		
Approaches	1%	5%	50%	1%	5%	50%
A	22.91	22.91	22.91	2634	2634	2634
B	1.31	6.55	66	151	753	7535
C (incl. plastic)	39.23	38.14	31.63	4512	4386	3638
C (excl. plastic)	39.53	39.53	39.53	4546	4546	4546
			[Phe] in µg kg^{-1} ww			
A	5.12	5.12	5.12	596	596	596
B	11.29	56	565	1314	6569	65686
C (incl. plastic)	5.26	5.25	5.20	613	611	605
C (excl. plastic)	5.27	5.27	5.27	613	613	613
			[DEHP] in µg kg^{-1} ww			
A	8380	8380	8380	3478505	3478505	3478505
B	35	176	1765	14653	73264	732639
C (incl. plastic)	9488	9477	9370	3938468	3934025	3889502
C (excl. plastic)	9491	9491	9491	3939593	3939593	3939593
			PVC			
			[DDT] in µg kg^{-1} ww			
Approaches	1%	5%	50%	1%	5%	50%
A	22.91	22.91	22.91	2634	2634	2634
B	1.42	7.09	71	163	815	8149
C (incl. plastic)	39.21	38.04	31.31	4509	4374	3601
C (excl. plastic)	39.53	39.53	39.53	4546	4546	4546
			[Phe] in µg kg^{-1} ww			
A	5.12	5.12	5.12	596	596	596
B	0.50	2.50	25	58	291	2913
C (incl. plastic)	5.27	5.27	5.26	613	613	612
C (excl. plastic)	5.27	5.27	5.27	613	613	613
			[DEHP] in µg kg^{-1} ww			
A	8380	8380	8380	3478505	3478505	3478505
B	4.27	21	214	1773	8864	88644
C (incl. plastic)	9491	9489	9475	3939456	3938912	3932874
C (excl. plastic)	9491	9491	9491	3939593	3939593	3939593

Table S10. Estimated concentration of DDT, phenanthrene (Phe) and DEHP in a seabird following ingestion of contaminated PE and PVC for scenarios A, B and C. Highlighted in red are examples of the neutral transfer of plastic co-contaminants following ingestion.

PE						
	[DDT] in μg kg^{-1} ww					
Seabird	low pollution			high pollution		
Approaches	1%	5%	50%	1%	5%	50%
A	45.82	45.82	45.82	5269	5269	5269
B	62.24	311.22	3112	7158	35790	357900
C (incl. plastic)	175.61	166.48	114.14	20195	19145	13126
C (excl. plastic)	178.12	178.12	178.12	20484	20484	20484
	[Phe] in μg kg^{-1} ww					
A	10.25	10.25	10.25	1192	1192	1192
B	536.29	2681	26814	62401	312007	3120067
C (incl. plastic)	18.07	13.21	10.71	2103	1537	1246
C (excl. plastic)	23.81	23.81	23.81	2770	2770	2770
	[DEHP] in μg kg^{-1} ww					
A	16760	16760	16760	6957011	6957011	6957011
B	1677	8384	83836	695998	3479990	34799900
C (incl. plastic)	39301	39166	37781	16313675	16257409	15682649
C (excl. plastic)	39335	39335	39335	16327888	16327888	16327888
PVC						
	[DDT] in μg kg^{-1} ww					
Approaches	1%	5%	50%	1%	5%	50%
A	45.82	45.82	45.82	5269	5269	5269
B	67.31	336.57	3366	7741	38705	387055
C (incl. plastic)	175.41	165.62	111.64	20172	19046	12838
C (excl. plastic)	178.12	178.12	178.12	20484	20484	20484
	[Phe] in μg kg^{-1} ww					
A	10.25	10.25	10.25	1192	1192	1192
B	23.78	118.90	1189	2767	13835	138348
C (incl. plastic)	23.38	21.91	15.44	2721	2549	1796
C (excl. plastic)	23.81	23.81	23.81	2770	2770	2770
	[DEHP] in μg kg^{-1} ww					
A	16760	16760	16760	6957011	6957011	6957011
B	202.87	1014	10144	84210	421051	4210514
C (incl. plastic)	39331	39315	39130	16326165	16319283	16242797
C (excl. plastic)	39335	39335	39335	16327888	16327888	16327888

References

Al-Hassan, J.M., Afzal, M., Rao, C.V.N., Fayad, S., 2003. Polycyclic Aromatic Hydrocarbons (PAHs) and Aliphatic Hydrocarbons (AHs) in Edible Fish from the Arabian Gulf. Bulletin of Environmental Contamination and Toxicology 70, 0205-0212.

Auman, H.J., Ludwig, J.P., Summer, C.L., Verbrugge, D.A., Froese, K.L., Colborn, T., Giesy, J.P., 1997. PCBS, DDE, DDT, and TCDD-EQ in two species of albatross on Sand Island, Midway Atoll, North Pacific Ocean. Environmental Toxicology and Chemistry 16, 498-504.

Avery-Gomm, S., O'Hara, P.D., Kleine, L., Bowes, V., Wilson, L.K., Barry, K.L., 2012. Northern fulmars as biological monitors of trends of plastic pollution in the eastern North Pacific. Marine Pollution Bulletin 64, 1776-1781.

Bakir, A., Rowland, S.J., Thompson, R.C., 2014. Enhanced desorption of persistent organic pollutants from microplastics under simulated physiological conditions. Environmental Pollution 185, 16-23.

Besseling, E., Wegner, A., Foekema, E.M., van den Heuvel-Greve, M.J., Koelmans, A.A., 2012. Effects of Microplastic on Fitness and PCB Bioaccumulation by the Lugworm Arenicola marina (L.). Environmental Science & Technology 47, 593-600.

Beukema, J., De Vlas, J., 1979. Population parameters of the lugworm, Arenicola marina, living on tidal flats in the Dutch Wadden Sea. Netherlands Journal of Sea Research 13, 331-353.

Blight, L.K., Burger, A.E., 1997. Occurrence of plastic particles in seabirds from the eastern North Pacific. Marine Pollution Bulletin 34, 323-325.

Bock, M.J., Miller, D.C., 1999. Particle selectivity, gut volume, and the response to a step change in diet for deposit-feeding polychaetes. Limnology and oceanography 44, 1132-1138.

Boerger, C.M., Lattin, G.L., Moore, S.L., Moore, C.J., 2010. Plastic ingestion by planktivorous fishes in the North Pacific Central Gyre. Marine Pollution Bulletin 60, 2275-2278.

Botkin, D.B., Miller, R.S., 1974. Mortality rates and survival of birds. American Naturalist, 181-192.

Bravo Rebolledo, E.L., Van Franeker, J.A., Jansen, O.E., Brasseur, S.M.J.M., 2012. Plastic ingestion by harbour seals (Phoca vitulina) in The Netherlands. Marine Pollution Bulletin.

Browne, M.A., Dissanyake, A., Galloway, T.S., Lowe, D., Thompson, R.C., 2008. Ingested microplastic translocates to the circulatory system of the mussel, Mytilus edulis (L.). Environ.Sci.Technol. 42, 5026-5031.

Browne, M.A., Galloway, T.S., Thompson, R.C., 2010. Spatial patterns of plastic debris along estuarine shorelines. Environmental Science & Technology 44, 3404-3409.

Browne, M.A., Niven, S.J., Galloway, T.S., Rowland, S.J., Thompson, R.C., 2013. Microplastic moves pollutants and additives to worms, reducing functions linked to health and biodiversity. Current Biology 23, 2388-2392.

Buckman, A.H., Norstrom, R.J., Hobson, K.A., Karnovsky, N.J., Duffe, J., Fisk, A.T., 2004. Organochlorine contaminants in seven species of Arctic seabirds from northern Baffin Bay. Environmental Pollution 128, 327-338.

Carson, H.S., Colbert, S.L., Kaylor, M.J., McDermid, K.J., 2011. Small plastic debris changes water movement and heat transfer through beach sediments. Marine Pollution Bulletin 62, 1708-1713.

Chang, B.V., Liao, C.S., Yuan, S.Y., 2005. Anaerobic degradation of diethyl phthalate, di-n-butyl phthalate, and di-(2-ethylhexyl) phthalate from river sediment in Taiwan. Chemosphere 58, 1601-1607.

Chen, Z., Mayer, L., 1999. Sedimentary metal bioavailability determined by the digestive constraints of marine deposit feeders: gut retention time and dissolved amino acids. Marine Ecology-Progress Series 176, 139.

184 Cheung, K.C., Leung, H.M., Kong, K.Y., Wong, M.H., 2007. Residual levels of DDTs and
185 PAHs in freshwater and marine fish from Hong Kong markets and their health risk
186 assessment. Chemosphere 66, 460-468.
187 Cifuentes, J.M., Becker, P.H., Sommer, U., Pacheco, P., Schlatter, R., 2003. Seabird eggs
188 as bioindicators of chemical contamination in Chile. Environmental Pollution 126, 123-137.
189 Denuncio, P., Bastida, R., Dassis, M., Giardino, G., Gerpe, M., Rodríguez, D., 2011. Plastic
190 ingestion in Franciscana dolphins, Pontoporia blainvillei (Gervais and d'Orbigny, 1844), from
191 Argentina. Marine Pollution Bulletin 62, 1836-1841.
192 Dhananjayan, V., Muralidharan, S., 2012. Polycyclic Aromatic Hydrocarbons in Various
193 Species of Fishes from Mumbai Harbour, India, and Their Dietary Intake Concentration to
194 Human. International Journal of Oceanography 2012.
195 Di Gangi, J., 1999. Phthalates in PVC medical products from 12 countries. Washington, DC:
196 Greenpeace.
197 Duinker, J.C., Hillebrand, M.T.J., Boon, J.P., 1983. Organochlorines in benthic invertebrates
198 and sediments from the Dutch Wadden Sea; identification of individual PCB components.
199 Netherlands Journal of Sea Research 17, 19-38.
200 Endo, S., Yuyama, M., Takada, H., 2013. Desorption kinetics of hydrophobic organic
201 contaminants from marine plastic pellets. Marine Pollution Bulletin 74, 125-131.
202 Engler, R.E., 2012. The Complex Interaction between Marine Debris and Toxic Chemicals in
203 the Ocean. Environmental Science & Technology 46, 12302-12315.
204 EPA, U., 2013. Exposure Assessment Tools and Models.
205 Everaarts, J.M., Bano, N., Swennen, C., Hillebrand, M.T.J., 1991. Cyclic chlorinated
206 hydrocarbons in benthic invertebrates from three coastal areas in Thailand and Malaysia.
207 Journal of the Science Society of Thailand 17, 31-49.
208 Fänge, R., Grove, D., 1979. 4 Digestion, in: W.S. Hoar, D.J.R., Brett, J.R. (Eds.), Fish
209 Physiology. Academic Press, pp. 161-260.
210 Foekema, E.M., De Gruijter, C., Mergia, M.T., van Franeker, J.A., Murk, A.J., Koelmans,
211 A.A., 2013. Plastic in North Sea Fish. Environmental Science & Technology 47, 8818-8824.
212 Galgani, F., Martínez-Gómez, C., Giovanardi, F., Romanelli, G., Caixach, J., Cento, A.,
213 Scarpato, A., BenBrahim, S., Messaoudi, S., Deudero, S., Boulahdid, M., Benedicto, J.,
214 Andral, B., 2011. Assessment of polycyclic aromatic hydrocarbon concentrations in mussels
215 (Mytilus galloprovincialis) from the Western basin of the Mediterranean Sea. Environmental
216 Monitoring and Assessment 172, 301-317.
217 GESAMP, 2015. Sources, fate and effects of microplastics in the marine environment: a
218 global assessment, in: Kershaw, P.J. (Ed.), p. 96 p.
219 Goldstein, M.C., Rosenberg, M., Cheng, L., 2012. Increased oceanic microplastic debris
220 enhances oviposition in an endemic pelagic insect. Biology Letters.
221 Gouin, T., Roche, N., Lohmann, R., Hodges, G., 2011. A thermodynamic approach for
222 assessing the environmental exposure of chemicals absorbed to microplastic. Environmental
223 Science & Technology 45, 1466-1472.
224 Graham, E.R., Thompson, J.T., 2009. Deposit- and suspension-feeding sea cucumbers
225 (Echinodermata) ingest plastic fragments. Journal of Experimental Marine Biology and
226 Ecology 368, 22-29.
227 Guruge, K.S., Watanabe, M., Tanaka, H., Tanabe, S., 2001. Accumulation status of
228 persistent organochlorines in albatrosses from the North Pacific and the Southern Ocean.
229 Environmental Pollution 114, 389-398.
230 Hendriks, A.J., 2007. The power of size: A meta-analysis reveals consistency of allometric
231 regressions. Ecological Modelling 205, 196-208.
232 Hendriks, A.J., Traas, T.P., Huijbregts, M.A.J., 2005. Critical body residues linked to octanol
233 - Water partitioning, organism composition, and LC50 QSARs: Meta-analysis and model.
234 Environmental Science and Technology 39, 3226-3236.
235 Hendriks, A.J., van der Linde, A., Cornelissen, G., Sijm, D.T., 2001a. The power of size. 1.
236 Rate constants and equilibrium ratios for accumulation of organic substances related to
237 octanol ‑ water partition ratio and species weight. Environmental Toxicology and Chemistry
238 20, 1399-1420.

239 Hendriks, A.J., Van der Linde, A., Cornelissen, G., Sijm, D.T.H.M., 2001b. The power of size.
240 1. Rate constants and equilibrium ratios for accumulation of organic substances related to
241 octanol-water partition ratio and species weight. Environmental Toxicology and Chemistry 20,
242 1399-1420.
243 Hennicke, J.C., King, B., Drynan, D., Hardy, L.J., Stokes, A., Taylor, S., 2012. New life-span
244 records of the brow booby Sula Leucogaster. Marine Ornithology 40, 125-126.
245 Herzke, D., Anker-Nilssen, T., Nøst, T.H., Götsch, A., Christensen-Dalsgaard, S., Langset,
246 M., Fangel, K., Koelmans, A.A., 2015. Negligible Impact of Ingested Microplastics on Tissue
247 Concentrations of Persistent Organic Pollutants in Northern Fulmars off Coastal Norway.
248 Environmental Science & Technology.
249 Hilton, G.M., Furness, R.W., Houston, D.C., 2000. A comparative study of digestion in North
250 Atlantic seabirds. Journal of Avian Biology 31, 36-46.
251 Huang, P.-C., Tien, C.-J., Sun, Y.-M., Hsieh, C.-Y., Lee, C.-C., 2008. Occurrence of
252 phthalates in sediment and biota: Relationship to aquatic factors and the biota-sediment
253 accumulation factor. Chemosphere 73, 539-544.
254 Institute, S.E.R., 2009. Screening of new contaminants in samples from the Norwegian
255 Arctic. Silver, Platinum, Sucralose, Bisphenol A, Tetrabrombisphenol A, Siloxanes, Phtalates
256 (DEHP) and Phosphororganic flame retardants.
257 Kavlock, R., Boekelheide, K., Chapin, R., Cunningham, M., Faustman, E., Foster, P., Golub,
258 M., Henderson, R., Hinberg, I., Little, R., 2002. NTP center for the evaluation of risks to
259 human reproduction: phthalates expert panel report on the reproductive and developmental
260 toxicity of di (2-ethylhexyl) phthalate. Reproductive toxicology 16, 529-653.
261 Koelmans, A.A., Bakir, A., Burton, G.A., Janssen, C.R., 2016. Microplastic as a vector for
262 chemicals in the aquatic environment: critical review and model-supported reinterpretation of
263 empirical studies. Environmental Science & Technology 50, 3315-3326.
264 Koelmans, A.A., Besseling, E., Foekema, E.M., 2014. Leaching of plastic additives to marine
265 organisms. Environmental Pollution 187, 49-54.
266 Koelmans, A.A., Besseling, E., Wegner, A., Foekema, E.M., 2013. Plastic as a carrier of
267 POPs to aquatic organisms: a model analysis. Environmental Science & Technology 47,
268 7812-7820.
269 Kühn, S., van Franeker, J.A., 2012. Plastic ingestion by the northern fulmar (< i> Fulmarus
270 glacialis</i>) in Iceland. Marine Pollution Bulletin 64, 1252-1254.
271 Law, R.J., Dawes, V.J., Woodhead, R.J., Matthiessen, P., 1997. Polycyclic aromatic
272 hydrocarbons (PAH) in seawater around England and Wales. Marine Pollution Bulletin 34,
273 306-322.
274 Lindstedt, S., Calder III, W., 1981. Body size, physiological time, and longevity of
275 homeothermic animals. Quarterly Review of Biology, 1-16.
276 Lusher, A.L., McHugh, M., Thompson, R.C., 2013. Occurrence of microplastics in the
277 gastrointestinal tract of pelagic and demersal fish from the English Channel. Marine Pollution
278 Bulletin 67, 94-99.
279 MacKay, D., Mackay, D., Shiu, W.Y., Ma, K.C., 1993. Illustrated Handbook of Physical-
280 chemical Properties and Environmental Fate for Organic Chemicals: Volatile organic
281 chemicals. Lewis Publishers.
282 Mallory, M.L., 2008. Marine plastic debris in northern fulmars from the Canadian high Arctic.
283 Marine Pollution Bulletin 56, 1501-1504.
284 Mallory, M.L., Roberston, G.J., Moenting, A., 2006. Marine plastic debris in northern fulmars
285 from Davis Strait, Nunavut, Canada. Marine Pollution Bulletin 52, 813-815.
286 Matthiessen, P., Thain, J.E., Law, R.J., Fileman, T.W., 1993. Attempts to assess the
287 environmental hazard posed by complex mixtures of organic chemicals in UK estuaries.
288 Marine Pollution Bulletin 26, 90-95.
289 Murray, F., Cowie, P.R., 2011. Plastic contamination in the decapod crustacean Nephrops
290 norvegicus (Linnaeus, 1758). Marine Pollution Bulletin 62, 1207-1217.
291 Napper, I.E., Bakir, A., Rowland, S.J., Thompson, R.C., 2015. Characterisation, quantity and
292 sorptive properties of microplastics extracted from cosmetics. Marine Pollution Bulletin 99,
293 178-185.

Neff, J.M., 2002. Bioaccumulation in Marine Organisms: Effect of Contaminants from Oil Well Produced Water. Elsevier.
O'Connor, I.A., Huijbregts, M.A., Ragas, A.M., Hendriks, A.J., 2013a. Predicting the oral uptake efficiency of chemicals in mammals: Combining the hydrophilic and lipophilic range. Toxicology and applied pharmacology 266, 150-156.
O'Connor, I.A., Huijbregts, M.A.J., Ragas, A.M.J., Hendriks, A.J., 2013b. Predicting the oral uptake efficiency of chemicals in mammals: Combining the hydrophilic and lipophilic range. Toxicology and Applied Pharmacology 266, 150-156.
Organization, W.H., 1992. International programme on chemical safety "diethylhexyl phthalate", Environmental Health Criteria 131, Geneva.
Pérez-Carrera, E., León, V.M.L., Parra, A.G., González-Mazo, E., 2007. Simultaneous determination of pesticides, polycyclic aromatic hydrocarbons and polychlorinated biphenyls in seawater and interstitial marine water samples, using stir bar sorptive extraction–thermal desorption–gas chromatography–mass spectrometry. Journal of Chromatography A 1170, 82-90.
Persson, P.-E., Penttinen, H., Nuorteva, P., 1978. DEHP in the vicinity of an industrial area in Finland. Environmental Pollution (1970) 16, 163-166.
Perugini, M., Cavaliere, M., Giammarino, A., Mazzone, P., Olivieri, V., Amorena, M., 2004. Levels of polychlorinated biphenyls and organochlorine pesticides in some edible marine organisms from the Central Adriatic Sea. Chemosphere 57, 391-400.
Ray, L.E., Murray, H., Giam, C., 1983a. Organic pollutants in marine samples from Portland, Maine. Chemosphere 12, 1031-1038.
Ray, L.E., Murray, H.E., Giam, C.S., 1983b. Organic pollutants in marine samples from Portland, Maine. Chemosphere 12, 1031-1038.
Rochman, C.M., Hoh, E., Kurobe, T., Teh, S.J., 2013. Ingested plastic transfers hazardous chemicals to fish and induces hepatic stress. Scientific reports 3.
Ryan, P.G., 1990. The effects of ingested plastic and other marine debris on seabirds. NOAA Technical Memorandum.
Ryan, P.G., Connell, A.D., Gardner, B.D., 1988. Plastic ingestion and PCBs in seabirds: Is there a relationship? Marine Pollution Bulletin 19, 174-176.
Said, T.O., 2007. Determination of persistent organic pollutants in sediment and fish of the western coast of Alexandria, Egypt. Chemistry and Ecology 23, 289-302.
Tanaka, K., Takada, H., Yamashita, R., Mizukawa, K., Fukuwaka, M.-a., Watanuki, Y., 2013. Accumulation of plastic-derived chemicals in tissues of seabirds ingesting marine plastics. Marine Pollution Bulletin.
Tanaka, K., Takada, H., Yamashita, R., Mizukawa, K., Fukuwaka, M.-a., Watanuki, Y., 2015. Facilitated Leaching of Additive-Derived PBDEs from Plastic by Seabirds' Stomach Oil and Accumulation in Tissues. Environmental Science & Technology 49, 11799-11807.
Teuten, E.L., Rowland, S.J., Galloway, T.S., Thompson, R.C., 2007. Potential for Plastics to Transport Hydrophobic Contaminants. Environmental Science & Technology 41, 7759-7764.
Thompson, R.C., Olsen, Y., Mitchell, R.P., Davis, A., Rowland, S.J., John, A.W., McGonigle, D., Russell, A.E., 2004. Lost at sea: where is all the plastic? Science 304, 838-838.
van Franeker, J.A., 1985. Plastic ingestion in the North Atlantic fulmar. Marine Pollution Bulletin 16, 367-369.
van Franeker, J.A., Bell, P.J., 1988. Plastic ingestion by petrels breeding in Antarctica. Marine Pollution Bulletin 19, 672-674.
van Franeker, J.A., Blaize, C., Danielsen, J., Fairclough, K., Gollan, J., Guse, N., Hansen, P.-L., Heubeck, M., Jensen, J.-K., Le Guillou, G., 2011a. Monitoring plastic ingestion by the northern fulmar< i> Fulmarus glacialis</i> in the North Sea. Environmental Pollution 159, 2609-2615.
van Franeker, J.A., Blaize, C., Danielsen, J., Fairclough, K., Gollan, J., Guse, N., Hansen, P.-L., Heubeck, M., Jensen, J.-K., Le Guillou, G., Olsen, B., Olsen, K.-O., Pedersen, J., Stienen, E.W.M., Turner, D.M., 2011b. Monitoring plastic ingestion by the northern fulmar Fulmarus glacialis in the North Sea. Environmental Pollution 159, 2609-2615.

van Sebille, E., Wilcox, C., Lebreton, L., Maximenko, N., Hardesty, B.D., van Franeker, J., Eriksen, M., Siegel, D., Galgani, F., Lavender, L.K., 2015. A global inventory of small floating plastic debris. Environmental Research Letters 10, 124006.
Waldock, M., 1983. Determination of phthalate esters in samples from the marine environment using gas chromatography mass spectrometry. Chemistry in Ecology 1, 261-277.
Walker, C., Hopkin, S., Sibly, R., Peakall, D., 2001. Toxicity testing. Principles of ecotoxicology. 2nd edition. Taylor & Francis, 93-118.
Walker, C.H., 2008. Organic Pollutants: An Ecotoxicological Perspective, Second Edition. Taylor & Francis.
Ward, E.J., Shumway, S.E., 2004. Separating the grain from the chaff: particle selection in suspension- and deposit-feeding bivalves. Journal of Experimental Marine Biology and Ecology 300, 83-130.
Wright, S.L., Rowe, D., Thompson, R.C., Galloway, T.S., 2013. Microplastic ingestion decreases energy reserves in marine worms. Current Biology 23, R1031-R1033.
Wurl, O., Obbard, J.P., 2005. Chlorinated pesticides and PCBs in the sea-surface microlayer and seawater samples of Singapore. Marine Pollution Bulletin 50, 1233-1243.
Xie, Z., Ebinghaus, R., Temme, C., Caba, A., Ruck, W., 2005. Atmospheric concentrations and air–sea exchanges of phthalates in the North Sea (German Bight). Atmospheric Environment 39, 3209-3219.
Yuan, S.Y., Liu, C., Liao, C.S., Chang, B.V., 2002. Occurrence and microbial degradation of phthalate esters in Taiwan river sediments. Chemosphere 49, 1295-1299.
Zeng, E.Y., Tran, K., 2002. Distribution of chlorinated hydrocarbons in overlying water, sediment, polychaete, and hornyhead turbot (Pleuronichthys verticalis) in the Coastal ocean, Southern California, USA. Environmental Toxicology and Chemistry 21, 1600-1608.