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

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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 Modelling 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.

1. 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 (<5 mm) 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

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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, 2014, 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., 2013, 2014). 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