

## Invited Commentary

## Microplastics as a Vector of Hydrophobic Contaminants: Importance of Hydrophobic Additives

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### EDITOR'S NOTE:

This is 1 of 15 invited commentaries in the series "Current Understanding of Distribution, Effects, and Risks Posed by Microplastics in the Environment." These peer-reviewed commentaries reflect the views and knowledge from international experts and are intended to inform our current understanding of microplastics fate and effects in the environment.

### ABSTRACT

Despite a recent boom in research on the environmental fate, distribution, and harmful effects of chemical substances associated with marine plastic debris, no consensus has been reached on whether chemicals originating from microplastics cause serious environmental harm. For the risk assessment of chemical contaminants associated with microplastics, it would be useful to group organic chemicals into 2 categories: additives and nonadditives. Whereas plastic particles are not likely to be diffuse sources of chemicals that are not intentionally added to plastic products, continuous leaching of additives would result in higher concentrations, at least at a local scale. Unlike plasticizers and flame retardants, which have been relatively well investigated, antioxidants and photostabilizers have been rarely studied, even though many of them are highly hydrophobic and are not readily biodegradable. More research on the fate and effects of chemicals via microplastics should focus on those additives. *Integr Environ Assess Manag* 2017;13:494–499. © 2017 SETAC

**Keywords:** Microplastics Additives UV stabilizers Antioxidants Flame retardants

### INTRODUCTION

Recent evidence on the global distribution of microplastics has raised serious concerns about potential adverse ecological and human health effects (Moore 2008). One of the most important hypotheses on the adverse effects is that microplastics can carry harmful chemicals and facilitate their transport into organisms (Teuten et al. 2009). A strong sorption capacity of microplastics and nanoplastics for hydrophobic organic chemicals has been determined by measuring (ad)sorption or partition coefficients for many chemicals, such as persistent organic pollutants, pharmaceuticals, and a few plastic additives (Lee et al. 2014; Velzeboer et al. 2014; Wang et al. 2015; Wu et al. 2016).

Because of the high sorptive capacity of microplastics for hydrophobic organic chemicals, adverse effects caused by microplastics in combination with organic pollutants have been demonstrated (e.g., Ma et al. 2016). A few laboratory-scale studies also showed that plastic particles have the potential to accelerate the uptake of chemicals by organisms

that ingest plastic particles (Chua et al. 2014; Wardrop et al. 2016). However, the observed effects in test organisms would not be environmentally relevant because the dose of plastic particles in laboratory tests is extremely greater than in the field conditions of 100 to 1000 g/km<sup>2</sup>, even in the inner accumulation zones of the ocean gyres (Cózar et al. 2014). Modeling studies have revealed that for aquatic organisms, fractional intake via the ingestion of plastic particles would be much smaller than other direct and dietary uptake routes (Gouin et al. 2011; Bakir et al. 2016). Another aspect that needs to be considered is the direction of chemical flows due to the difference in chemical potential. Many anthropogenic chemicals that are not added in plastic products sorb to plastic particles because of very high partition coefficients between plastic and seawater (Lee et al. 2014; Velzeboer et al. 2014; Liu et al. 2016). However, the fugacity of these persistent chemicals in the plastic phase might be lower than those in water or in the organisms if the time to attain phase equilibrium is long (Lee et al. 2017). In this case, plastic particles should be regarded as a sink for those hydrophobic chemicals and might be used as widespread passive samplers for highly hydrophobic organic chemicals (Karapanagioti et al. 2011). However, the opposite is

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Published 25 April 2017 on [wileyonlinelibrary.com/journal/ieam](http://wileyonlinelibrary.com/journal/ieam).

expected for plastic additives because they are intentionally added to plastic products up to a few tens of percent (Al-Odaini et al. 2015; Jang et al. 2016).

It is of great interest whether organic chemicals from microplastics cause serious environmental risks. Recent findings are still not sufficient to reach a consensus on this issue. Until now, the majority of studies on the fate, distribution, and effects of hydrophobic organic pollutants associated with plastic particles have focused on plastic additives including flame retardants (Al-Odaini et al. 2015; Jang et al. 2016), plasticizers (Fries et al. 2013), and other nonadditives (e.g., persistent organic pollutants, polycyclic aromatic hydrocarbons, and pharmaceuticals and personal care products), which are not intentionally added to plastic matrix (Karapanagioti et al. 2011; Rochman et al. 2013; Lee et al. 2014; Velzeboer et al. 2014; Wang et al. 2015; Liu et al. 2016). Although studies have been conducted for brominated flame retardants, plasticizers such as di-ethylhexyl phthalate, and unbound monomers such as bisphenol A, there are a variety of chemicals that are intentionally added to plastic products to enhance physical and chemical properties. A few nontarget screening studies revealed that microplastics contain various chemical substances such as antioxidants and photostabilizers (Gauquie et al. 2015; Rani et al. 2015). Many of them are hydrophobic and might be released slowly from widespread plastic particles. Leaching of brominated flame retardants from marine Styrofoam buoys would be an example (Jang et al. 2016). In the present study, we evaluate the environmental fate of representative chemical additives in plastic products based on their properties and propose future research needs on hydrophobic organic chemicals associated with plastic particles in the environment.

## ROLES OF PLASTIC PARTICLES ON DYNAMIC TRANSPORT PROCESSES OF ORGANIC CHEMICALS

Fate and transport of organic chemicals in the environment are described by both chemical equilibrium and kinetics. Major processes that determine the fate and transport of organic chemicals and the role of plastic particles are described in Figure 1. Sorption equilibrium between plastics and water is evaluated by equilibrium partition or sorption coefficients. Diffusion in the plastic phase is often the rate-limiting process governing the overall rate of the transfer either from water to plastic or from plastic to water. Thus, the phase equilibrium between plastic and water may not be attained under environmental conditions. The fugacity gradient for additives may be opposite from that for nonadditives under the environmental conditions as illustrated in Figure 1.

Where biomagnification occurs, the fugacity of hydrophobic organic chemicals in organisms at higher trophic levels will be greater than that in water. For nonadditives, plastics are not likely to play important roles because fugacity in plastic particles cannot be greater than that in water. For additives, comparison of the rate of chemical uptake via plastic particles with the rate of uptake in bioconcentration and/or

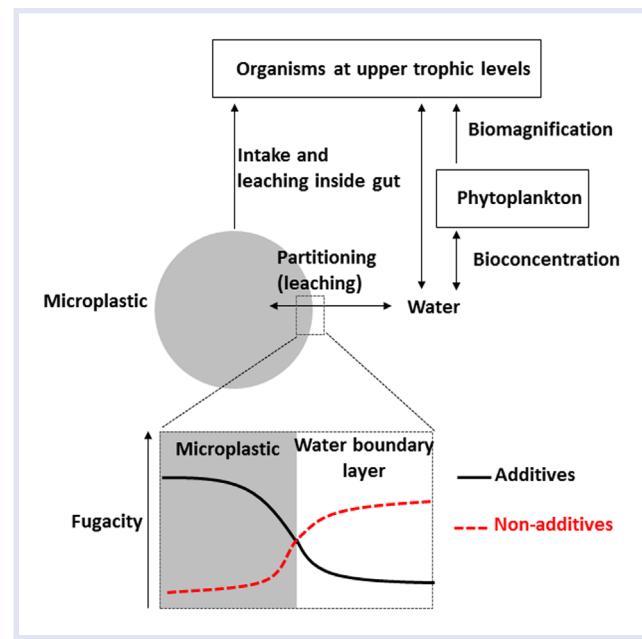


Figure 1. Dynamic processes determining the fate and transport of hydrophobic organic chemicals from plastic particles.

biomagnification processes would be useful. A few laboratory studies have revealed that organisms may gain hydrophobic additives such as brominated flame retardants by ingesting plastic particles (e.g., Chua et al. 2014; Wardrop et al. 2016), whereas modeling studies suggested that the additional uptake via the ingestion of plastic particles would be minimal (Gouin et al. 2011; Bakir et al. 2016). Further studies are required to allocate contributions of competing processes with multimedia monitoring of hydrophobic additives.

## ORGANIC ADDITIVES IN PLASTIC PRODUCTS

Many different types of plastic additives exist, including fillers, pigments, plasticizers, stabilizers, flame retardants, antistatic and conductive additives, food contact, and medical additives, among others (Murphy 2001). Table 1 shows examples of chemical substances, with their physical and chemical properties, that are widely used as plastic additives. The domain of physical and chemical properties is very wide. As shown, many organic substances are not expected to be readily biodegradable using a battery of BIOWIN™ models (USEPA 2012). The range of hydrophobicity based on  $\log K_{ow}$  is also very wide. Experimental values are available only for relatively well-studied plasticizers and flame retardants (World Health Organization 1996; Tittlemier et al. 2002; Braekkeveld et al. 2003; Hunziker et al. 2004; Kuramochi et al. 2007, 2014; Arnot et al. 2009). Many additives containing long alkyl chains are predicted to have extremely high  $\log K_{ow}$  values and very low water solubilities estimated by the EPI Suite™ program (USEPA 2012). These superhydrophobic additives are not likely to bioaccumulate because of their large molecular size and very limited water solubility, which leads to an extremely long bioconcentration half-life (Kwon et al. 2016). However, many additives such as benzotriazole UV stabilizers and phenolic antioxidants are

Table 1. Representative plastic additives and their chemical properties

Additive type	Example substances	Molecular weight	Log $K_{ow}^a$	Vapor pressure (Pa) <sup>a</sup>	Water solubility (mg/L) <sup>a</sup>	Ready biodegradability <sup>a</sup>
Antioxidants	Phenols	BHT (2,6-di-t-butyl-4-methylphenol) IRGANOX 1010	220.36 1177.67	5.03 19.60	0.24 $9.8 \times 10^{-29}$	5.7 $5.2 \times 10^{-3}$
	Amines	AMINOX (acetone diphenylamine) 2,2,4-trimethyl-1,2-dihydroquinoline	378.52 173.26	7.20 3.30	$2.5 \times 10^{-8}$ 2.1	$2.9 \times 10^{-3}$
Photostabilizers	BNX 2000 (N,N-dibenzylhydroxylamine)	213.28	2.86	$2.8 \times 10^{-6}$	130	No
	Phosphites	PEP-8 [3,9-bis(octadecyloxy)-2,4,8,10-tetraoxa-3,9-diphosphaspiro[5.5]undecane] TPP (triphenyl phosphite)	733.06 310.29	15.05 6.62	$1.1 \times 10^{-14}$ 0.010	$2.9 \times 10^{-12}$
HALS	135A (isodecyl diphenyl phosphite)	374.46	8.52	$7.0 \times 10^{-6}$	$2.3 \times 10^{-4}$	No
	Benzotriazoles	UV326	315.8	5.55	$2.1 \times 10^{-7}$	0.68
Flame retardants	UV327	358	6.91	$3.6 \times 10^{-8}$	0.026	No
	UV328	351.5	7.25	$2.6 \times 10^{-8}$	0.015	No
Bromodiphenyl ethers	AO-20 [tris(3,5-di-tert-butyl-4-hydroxybenzyl) isocyanurate] AO-30 [1,1,3-tris(2-methyl-4-hydroxy-5-tert-butylphenyl)butane] AO-40 [4,4'-butylidenebis(6-tert-butyl-m-cresol)]	784.10 544.82 382.59	15.18 12.70 9.09	$6.2 \times 10^{-26}$ $2.1 \times 10^{-13}$ $7.0 \times 10^{-9}$	$4.0 \times 10^{-12}$ $2.0 \times 10^{-8}$ $2.6 \times 10^{-4}$	No
	Hexabromocyclohexane	$\alpha$ -HBCD $\beta$ -HBCD $\gamma$ -HBCD	641.7 641.7 641.7	5.38 <sup>b</sup> 5.47 <sup>b</sup> 5.80 <sup>b</sup>	$2.3 \times 10^{-6}$ $2.3 \times 10^{-6}$ $2.3 \times 10^{-6}$	$4.9 \times 10^{-5c}$ $1.5 \times 10^{-5c}$ $2.1 \times 10^{-6}$ (20°C) <sup>c</sup>
BDE 183 BDE 153 BDE 99	BDE 183	722.479	8.27 <sup>d</sup>	$4.7 \times 10^{-7e}$	$1.5 \times 10^{-9e}$	No
	BDE 153	643.584	8.05 <sup>f</sup>	$6.0 \times 10^{-7g}$	$8.7 \times 10^{-10e}$	No
	BDE 99	564.687	7.39 <sup>f</sup>	$2.2 \times 10^{-5g}$	$9.4 \times 10^{-6e}$	No

Plasticizers	Phthalates					
	Di-2-ethylhexyl phthalate (DEHP) <sup>a</sup>	390.57	4.88 <sup>h</sup>	$5.6 \times 10^{-12}$ <sup>h</sup>	0.023–0.34 <sup>h</sup>	Yes
	Di-isobutyl phthalate (DINP) <sup>a</sup>	418.62	9.37	$3.1 \times 10^{-5}$	$2.3 \times 10^{-5}$	No
	Di-isodecyl phthalate (DIDP) <sup>a</sup>	446.68	10.36	$3.1 \times 10^{-5}$	$2.2 \times 10^{-6}$	No
Adipates						
	Di-2-ethylhexyl adipate	370.58	8.12	$4.3 \times 10^{-4}$	$5.4 \times 10^{-4}$	Yes
	Di-isobutyl adipate	398.63	9.24	$4.4 \times 10^{-4}$	$4.0 \times 10^{-5}$	Yes

HALS = hindered amine light stabilizer; HBCD = hexabromocyclododecane.  
<sup>a</sup>Estimated values using EPI Suite program (US Environmental Protection Agency 2012) unless specified.

<sup>b</sup>Anot et al. 2009.

<sup>c</sup>Hanzlik et al. 2004.

<sup>d</sup>Braekeveldt et al. 2003.

<sup>e</sup>Titlemier et al. 2002.

<sup>f</sup>Kuramochi et al. 2007.

<sup>g</sup>Kuramochi et al. 2014.

<sup>h</sup>World Health Organization 1996.

expected to have  $\log K_{ow}$  values between 5 and 8 and have potential for bioaccumulation. Although experimental studies are lacking on their environmental partitioning, metabolic transformation, and biodegradability, they might accumulate through the marine food chain based on estimated high partition coefficients and low biodegradability. Field observations of phenolic antioxidants and benzotriazole UV stabilizers in sediments, suspended particles, and fish partly support this (Wang et al. 2003; Lu et al. 2016; Wick et al. 2016), although it is not certain that plastic debris is a significant source of them in the environment. Thus, future research should be directed toward those additives that might have persistence, bioaccumulation potentials, and toxicity. Because additives such as flame retardants have other emission sources, the estimation of quantitative contribution of microplastics on the global and local emission of plastic additives is required. Although a few studies have evaluated toxic potentials of additives such as UV stabilizers (Kawamura et al. 2003; Morohoshi et al. 2005), ecotoxic effects of plastic additives are largely unstudied.

## LEACHING OF ADDITIVES

As shown in Figure 1, leaching of plastic additives can be explained by diffusion in the plastic phase, instantaneous partitioning at the interface of plastic and water, and mass transfer within the surrounding boundary layer. Depending on chemical properties, the overall leaching of additives is determined by internal diffusion in plastic or by the aqueous boundary layer diffusion (Endo et al. 2013; Narváez Valderrama et al. 2016). As hydrophobicity or the partition coefficient between plastic and water increases, the loss of additives from plastic particles is slower and is more likely to be dominated by the aqueous boundary layer diffusion. Thus, partition coefficient between plastic and water ( $K_{pw}$ ) should be the most important parameter for the evaluation of additive leaching. Nevertheless, partition coefficients of many additives have not been reported. For highly hydrophobic additives, direct determination of  $K_{pw}$  is not straightforward. Recent methods using 3-phase systems (e.g., Lee et al. 2014) or a cosolvent system (e.g., Smedes et al. 2009) would be required. When plastic particles are ingested, desorption under the digestive environment may be accelerated (Bakir et al. 2014; Tanaka et al. 2015). This could be explained by the decrease in the aqueous boundary layer around the plastic particle or by the facilitated transfer by surfactants, proteins, or lipids.

Another aspect that needs to be considered is the physical and chemical states of additives in plastics. Relatively small molecules such as di(2-ethylhexyl)phthalate and brominated flame retardants are thought to exist as freely diffusible forms in plastic products. However, other additives have rarely been studied. Additives are mixed with pristine plastic pellets at a wide range of temperature during the formulation, and some agents are used during curing and cross-linking (Murphy 2001). Physical and chemical conditions during formulation may limit the freely diffusible fraction of additives, and thus leaching to the environment. Oligomeric and polymeric additives and agents used for curing and cross-linking are not thought to

migrate readily from plastic products during their use. However, they may leach as plastics undergo weathering in the environment. Thus, it is required to know the freely diffusible fraction of additives and to know the changes in the leachable fraction as plastic particles degrade in the environment.

## POTENTIAL HARMFUL EFFECTS OF LEACHING AND PERSPECTIVES FOR FUTURE RESEARCH

Because the amount of various additives in plastic may be very high (Murphy 2001), plastic particles in the environment can be regarded as passive dosing media for these additives. Depending on the properties of additives, they can be released during the lifetime of plastic particles in the environment. The leaching rates of various additives in environmental conditions, as well as organisms' gut conditions, need to be investigated. Assuming the level of plastic particle density in the open ocean (Cózar et al. 2014), the resulting concentration of additives in water or other environmental media is likely to be extremely low. However, whether high plastic debris accumulation zones in the semienclosed and enclosed embayment are influenced by the leaching additives needs to be evaluated. For instance, hexabromocyclododecane in Styrofoam buoys massively used in oyster culture farms contributed to enrichment of hexabromocyclododecane in surrounding sediment (Al-Odaini et al. 2015). Furthermore, plastic particles provide surfaces for growth of attached species so-called plastisphere (Zettler et al. 2013; McCormick et al. 2014) that might be exposed to very high concentrations of additives. Plastic debris of densities that are greater than water has rarely been studied. This type of debris tends to sink in the sediment and may have a longer lifetime than floating particles exposed to UV light and oxygen. Higher persistence and chronic release of additives from sunken plastics would cause ecological concerns, especially for benthic organisms.

**Acknowledgment**—This work was partly supported by the research project entitled "Environmental Risk Assessment of Microplastics in the Marine Environment" from the Ministry of Oceans and Fisheries, Korea and by National Research Foundation of Korea grant 2015R1A2A04003958.

**Data availability**—All data are publicly available through the USEPA's EPI Suite™.

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