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Occurrence and effects of plastic additives on marine environments and organisms: A review

Hermabessiere Ludovic [1], Dehaut Alexandre [1], Paul-Pont Ika [2], Lacroix Camille [3], Jezequel Ronan [3],
Soudant Philippe [2], Duflos Guillaume [1, *]

[1] Anses, Lab Securite Aliments, Blvd Bassin Napoleon, F-62200 Boulogne Sur Mer, France.
[2] Inst Univ Europeen Mer, Lab Sci Environm Marin LEMAR, UBO CNRS IRD IFREMER UMR6539, Technopole Brest Iroise,Rue Dumont dUryille, F-29280 Plouzane, France.
[3] CEDRE, 715 Rue Alain Colas, F-29218 Brest 2, France.

* Corresponding author : Guillaume Duflos, email address : guillaume.duflos@anses.fr

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Abstract :

Plastics debris, especially microplastics, have been found worldwide in all marine compartments. Much research has been carried out on adsorbed pollutants on plastic pieces and hydrophobic organic compounds (HOC) associated with microplastics. However, only a few studies have focused on plastic additives. These chemicals are incorporated into plastics from which they can leach out as most of them are not chemically bound. As a consequence of plastic accumulation and fragmentation in oceans, plastic additives could represent an increasing ecotoxicological risk for marine organisms. The present work reviewed the main class of plastic additives identified in the literature, their occurrence in the marine environment, as well as their effects on and transfers to marine organisms. This work identified poly-brominated diphenyl ethers (PBDE), phthalates, nonylphenols (NP), bisphenol A (BPA) and antioxidants as the most common plastic additives found in marine environments. Moreover, transfer of these plastic additives to marine organisms has been demonstrated both in laboratory and field studies. Upcoming research focusing on the toxicity of microplastics should include these plastic additives as potential hazards for marine organisms, and a greater focus on the transport and fate of plastic additives is now required considering that these chemicals may easily leach out from plastics.

Highlights

► PBDEs, phthalathes, nonylphenol, BPA and antioxidants are common plastic additives. ► Evidence for transfer and uptake of plastic additives by marine organisms. ► Plastic additives have negative effects on marine organisms. ► New research on microplastics should include their additives as a potential hazard.

Keywords : Microplastics, Plastic additives, Bisphenol A, Phthalates, Brominated flame retardant

1. Introduction

Due to their numerous societal benefits, plastics hold an important place in human society (Andrady and Neal, 2009). Plastic, a man-made material, is inexpensive, strong, durable, lightweight and easy to produce (Thompson *et al.*, 2009). As a consequence, plastic production has been increasing since the 1950s, and notably rose from 225 million tons in 2004 to 322 million tons in 2015, representing a 43% increase over the last decade (PlasticsEurope, 2016). However, this estimate does not take into account the proportion of synthetic fibers which are widely used in the textile and fishery industries (Dris *et al.*, 2016) and there is an underestimation of 15% to 20% depending on the year (Industrievereinigung Chemiefaser, 2013). Low estimates predicted that floating marine plastic weigh between 70,000 and 270,000 tons (Cózar *et al.*, 2014; Eriksen *et al.*, 2014; Van Sebille *et al.*, 2015). Small pieces of plastics called microplastics (MP) account for a total of 51 trillion plastic debris (Van Sebille *et al.*, 2015).

Microplastics have been defined as plastics particles smaller than 5 mm (Arthur *et al.*, 2009). Growing attention has been accorded to microplastics during the last decade, since the publication by Thompson *et al.* (2004). Micro-sized plastic pieces originate from two distinct pathways: primary and secondary sources. Primary sources of MP correspond to (i) plastics that are directly manufactured at micrometric size, including plastic pellets (Barnes *et al.*, 2009; Cole *et al.*, 2011), (ii) MP from exfoliating cosmetics (Chang, 2015; Fendall and Sewell, 2009; Napper *et al.*, 2015; Zitko and Hanlon, 1991) and (iii) clothing fibers found in wastewater treatment plants (Browne *et al.*, 2011; Carr *et al.*, 2016). Secondary MP results from the breakdown of larger pieces due to mechanical abrasion and photochemical oxidation in the environment (Andrady, 2011; Bouwmeester *et al.*, 2015; Lambert and Wagner, 2016). MP can also degrade into smaller pieces called nanoplastics (Gigault *et al.*, 2016; Koelmans *et al.*, 2015; Lambert and Wagner, 2016).

Due to their small size, MP can be ingested by a wide range of marine organisms such as zooplankton, bivalves and worms (De Witte *et al.*, 2014; Devriese *et al.*, 2015; Graham and Thompson, 2009; Rochman *et al.*, 2015; Sussarellu *et al.*, 2016; Van Cauwenberghe and Janssen, 2014; Van Cauwenberghe *et al.*, 2015) and by organisms from higher trophic levels such as fish (Boerger *et al.*, 2010; Carpenter *et al.*, 1972; Dantas *et al.*, 2012; Foekema *et al.*, 2013; Lusher *et al.*, 2013; Neves *et al.*, 2015; Possatto *et al.*, 2011; Rochman *et al.*, 2015) and marine mammals (Eriksson and Burton, 2003; Lusher *et al.*, 2015). This ingestion of MPs can result in physical damage such as obstruction or internal abrasions (Wright *et al.*, 2013). In addition to these physical threats, MP can potentially transfer chemicals adsorbed on their surface (Mato *et al.*, 2001; Teuten *et al.*, 2007; Teuten *et al.*, 2009) or plastic additives. However, less attention has been paid to the transfer of plastic additives to marine organisms in comparison with hydrophobic organic compounds (HOC), despite the fact that many additives have been recognized as hazardous (Lithner *et al.*, 2011). Therefore, the transport and fate of plastic additives leaching out from plastic debris should definitely be carefully addressed in future field, laboratory and modelling works.

Plastics are made by polymerizing monomers and other substances (Lithner *et al.*, 2011) including plastic additives. Plastic additives are chemical compounds, like plasticizers, which provide required properties to a plastic polymer or are incorporated to facilitate the manufacturing process (OECD, 2004). Moreover, some plastic additives are used as monomers, for example bisphenol A is the monomer of polycarbonate (PC) but also a stabilizer in other polymers. Plastic additives are mainly used as plasticizers, flame retardants, stabilizers, antioxidants and pigments. Phthalates, BPA, nonylphenols, and brominated flame retardants (BFR) are the most common additives recovered from the environment (Bergé *et al.*, 2012; David *et al.*, 2009; de Boer *et al.*, 1998; de los Ríos *et al.*, 2012; Mackintosh *et al.*, 2004; Net *et al.*, 2015; Xie *et al.*, 2005; Xie *et al.*, 2007) and represent a hazard to the environment and organisms (Lithner *et al.*, 2011; Meeker *et al.*, 2009; Oehlmann *et al.*, 2009). These plastic additives are released into the marine environment by numerous pathways including industrial and municipal wastewater, atmospheric deposition, runoff and river transport resulting from application of sewage sludge in agriculture. In addition leaching of plastic additives from macro and microplastics is known to occur in the marine environment. Thus, the accumulation and degradation of plastic debris might represent another major input of these chemical compounds in oceans. As a consequence, more research is needed on the hazards of plastic additives associated with microplastics.

The aim of this review is to (i) list and describe the most predominant plastic additives used worldwide in the plastic industry, (ii) present an overview of the occurrence of plastics additives in the marine environment, and (iii) document the effects of plastic additives on marine organisms. Lastly, recommendations will be made in order to identify the polymer-additives pairs of major concern on which further research should focus.

2. Chemicals used as plastic additives

Multiple types and families of chemicals are mixed with polymers to produce plastics. The type of additive depends on the plastic polymer and the requirements of the final product (Table 1).

Table 1: List of the most commonly produced polymers and their associated plastic additives. Adapted from Hansen *et al.* (2013)

Polymer	Consumption in the EU27 (in million tons) in 2015[1]	Additive types	Amount in polymers (% w/w)	Hazardous substances[2]
PP	9	Antioxidant	0.05 – 3	Bisphenol A; Octylphenol; Nonylphenol
		Flame retardant (cable insulation and electronic applications)	12 – 18	Brominated flame retardant; Boric acid; Tris(2-chloroethyl)phosphate
HDPE	8	Antioxidant	0.05 - 3	Bisphenol A; Octylphenol; Nonylphenol
		Flame retardant (cable insulation application)	12 -18	Brominated flame retardant; Boric acid; Tris(2-chloroethyl)phosphate
LDPE	6	Antioxidant	0.05 – 3	Bisphenol A; Octylphenol; Nonylphenol
		Flame retardant (cable insulation application)	12 – 18	Brominated flame retardant; Boric acid; Tris(2-chloroethyl)phosphate
PVC	5	Plasticizer	10 – 70	Phthalate
		Stabilizer	0.5 – 3	Bisphenol A; Nonylphenol
PUR	3.5	Flame retardant	12 - 18	Brominated flame retardant; Boric acid; Tris(2-chloroethyl)phosphate

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[1] According to PlasticsEurope (2016); PP: Polypropylene; HDPE: High Density Polyethylene; LDPE: Low Density Polyethylene; PVC: Polyvinyl Chloride; PUR: Polyurethane.

[2] Hazardous substances refer to chemicals that pose a risk to the environment and to human health as defined by the REACH regulation in the European Union according to the European Chemical Agency (2017).

The following section describes the most common additive types used in the manufacturing processes (Table 2) that have been reported in macro- and microplastic debris collected in environmental surveys: brominated flame retardants, phthalates used as plasticizers, nonylphenols, bisphenol A and antioxidants.

Table 2: List of common plastic additives and their associated functions and potential effects

Additives	Function	Effects
Brominated Flame Retardants (BFR)	Reduce flammability in plastic. Also adsorbed on plastic from the surrounding environment.	Potential endocrine disruptors
Phthalates	Plasticizers to soften plastic mainly in polyvinyl chloride.	Endocrine disruptors
Nonylphenol	Antioxidant and plasticizer in some plastics	Endocrine disruptors
Bisphenol A (BPA)	Monomer in polycarbonate and epoxy resins. Antioxidant in some plastics.	Endocrine disruptors Estrogen mimic
Irganox®	Antioxidant in some plastics.	

The main plastic additives described are listed in Table 3 with their associated octanol-water partition coefficient (K_{ow}). K_{ow} has been used for predicting how a chemical will concentrate in marine organisms and an increase in log K_{ow} indicates an increase in the potential bioconcentration in organisms (Net *et al.*, 2015).

Table 3: Plastic additives and their associated octanol-water partition coefficients (Log K_{ow}). Data were extracted from the following reviews: Bergé *et al.* (2012), Espinosa *et al.* (2016), Net *et al.* (2015) and Oehlmann *et al.* (2008)

Full name	Abbreviation	Log K_{ow}
butyl benzyl phthalate	BBP	4.70
di(2-ethylexyl) phthalate	DEHP	7.73
diethyl phthalate	DEP	2.54
diisobutyl phthalate	DiBP	4.27
diisodecyl phthalate	DiDP	9.46
diisononyl phthalate	DiNP	8.60
dimethyl phthalate	DMP	1.61
di-*n*-butyl phthalate	DnBP	4.27
di-*n*-octyl phthalate	DnOP	7.73
hexabromocyclododecane	HBCD	5.07 – 5.47
polybrominated diphenyl ether	PBDE	5.52 – 11.22
tetrabromobisphenol A	TBBPA	4.5
bisphenol A	BPA	3.40
nonylphenol	NP	4.48 – 4.80

2.1. Brominated flame retardants

Brominated flame retardants (BFR) are a class of additives used in plastic products to reduce flammability. These BFR are used in a variety of consumer products ranging from electronic devices to insulation foams. BFRs include a wide range of chemicals, of which polybrominated diphenyl ethers (PBDE), hexabromocyclododecane (HBCD – Pubchem ID: 18529) and tetrabromobisphenol A (TBBPA – Pubchem ID: 6618) (Talsness *et al.*, 2009) represent the main BFRs used in the plastic industry. These 3 classes (PBDE, HBCD and TBBPA) are described in details below. Lately, attention has been given to other emerging BFRs such as 1,2-bis(2,4,6-tribromophenoxy)ethane (BTBPE – Pubchem ID: 37840), decabromodiphenylethane (DBDPE – Pubchem ID: 10985889) and hexabromobenzene (HBB – Pubchem ID: 6905) as these have been identified in many environmental compartments, organisms, food and humans (European Food Safety Authority, 2012). As they are not chemically bound to the polymer matrix, they can leach into the surrounding environment

(Engler, 2012; Meeker *et al.*, 2009) with an exception for TBBPA which is chemically bound to the polymer (Morris *et al.*, 2004).

PBDEs are hydrophobic substances that include numerous formulations used in plastics as flame retardants. Indeed, there are three main commercial formulations called penta-, octa- and deca-BDEs (Chua *et al.*, 2014). These additives are ubiquitous, toxic, persistent and bioaccumulate in the environment and are of great concern for human health (Engler, 2012). As a result, penta- and octa-BDEs have been banned by the European Union since 2004 (European Directive, 2003), while deca-BDE was banned only in 2009 from electronic and electrical applications in the European Union (European Council Decision, 2009). These formulations can no longer be used in mixtures or products with a concentration higher than 0.1% by mass. In addition, tetra- to hepta-BDEs are listed for elimination in the Annex A of the Stockholm Convention on persistent organic pollutant (POP) (Stockholm Convention, 2016). Moreover, in Canada the use of tetra- to deca- BDE has been restricted under the SOR/2008-218 Regulations (Consolidated Regulation, 2008). Since 2006, penta- to octa-BDE have been subjected to a 90 day notification before importation or production in the US. Finally, deca-BDE importation and production have been entirely stopped (US Environmental Protection Agency, 2006, 2012) since 2013.

HBCD has three dominant stereoisomers: α-, β-, and γ-HBCD (European Food Safety Authority, 2011a). These BFRs are listed as POPs in the Stockholm Convention (Stockholm Convention, 2016) and the three isomers are subject to a request for authorization in the European Union (European Council Regulation, 2006). HBCDs are found in expanded PS (EPS) and extruded PS (XPS) up to 4-7% by weight (Al-Odaini *et al.*, 2015). Its use has been subjected to authorization in the European Union since 2006 in the annex XIV of the Registration, Evaluation, Authorisation and Restriction of Chemicals (REACH) regulation (European Food Safety Authority, 2011a). Moreover, in 2013 HBCD was listed for elimination in the Annex A of the Stockholm Convention with specific exemption for use and production in EPS and XPS (Cruz *et al.*, 2015; Stockholm Convention, 2016). In the US, Environmental Protection Agency conducted a risk assessment for HBCD according to the 2010 "Toxic Substances Control Act" action plan (US Environmental Protection Agency, 2010b).

TBBPA is produced by brominating bisphenol A (European Food Safety Authority, 2011b). According to the European Food Safety Authority (2011b), TBBPA is the most commonly produced BFR in the world and represents 60% of the BFR market. This BFR is used in acrylonitrile butadiene styrene (ABS) and in other plastic such as high impact PS and phenolic resin (Cruz *et al.*, 2015). Until now, no legislation concerning TBBPA has been applied in the European Union (Vandermeersch *et al.*, 2015).

2.2. Phthalates

Phthalic acid esters (PAE) or phthalates are a family of plastic additives used as plasticizers, mainly in PVC production (Arbeitsgemeinschaft PVC und Umwelt e.V, 2006). As a result, PVC can contain 10% to 60% phthalates by weight (Net *et al.*, 2015). As phthalates are not chemically bound to the polymer matrix, they can easily leach into the environment during manufacturing, use and disposal (Net *et al.*, 2015). PAEs have been found in a wide range of environments (as reviewed by Net *et al.* (2015)) and this is of concern, since some phthalates have been defined as endocrine disruptors, even at low concentrations (Oehlmann *et al.*, 2009).

In 2015, 8.4 million tons of plasticizers were used around the world, and di(2-ethylexyl) phthalate (DEHP – Pubchem ID: 8343) was the most commonly used plasticizer, representing 37.1% of the global plasticizer market (ECPI, 2016). Europe accounted for 1.3 million tons of the global plasticizer market in 2015 (ECPI, 2016), but DEHP was not the most commonly used plasticizer in Europe, as suggested by its 20% decrease in consumption between 1999 and 2004. DEHP has gradually been replaced by diisononyl phthalate (DiNP – Pubchem ID: 590836), diisodecyl phthalate (DiDP – Pubchem ID: 33599) and di(2-Propyl Heptyl) phthalate (DPHP – Pubchem ID: 92344), which represented 57% of plasticizer consumption in Europe in 2015 (Arbeitsgemeinschaft PVC und Umwelt e.V, 2006; ECPI, 2016).

2.3. Bisphenol A

BPA (Pubchem ID: 6623) is the most representative chemical of the bisphenol group and is one of the most commonly produced chemicals worldwide, with over three million tons produced annually (Laing *et al.*, 2016). BPA is mainly used as a monomer for polycarbonate (PC) plastics (65% of volume used) and epoxy resins (30% of volume used), which are for instance the main component of the lining layer of aluminum cans (Crain *et al.*, 2007; ICIS, 2003). BPA can also be used as an antioxidant or as a plasticizer in other polymers (PP, PE and PVC) (Rani *et al.*, 2015). Leaching of BPA can occur (Sajiki and Yonekubo, 2003), leading to the release of this additive from food and drink packaging, which is considered as a source of exposure for human beings (Vandermeersch *et al.*, 2015). Despite its potential to leach from food packaging and the fact that it has been identified as a significant endocrine disruptor (Oehlmann *et al.*, 2009), BPA is still allowed in the European Union for use in food contact material (European Council Regulation, 2011). Other bisphenol analogs, such as bisphenol B, bisphenol F and bisphenol S are used in plastics and may represent a threat to the environment even though their toxicity is still unknown (Chen *et al.*, 2016).

2.4. Nonylphenols

Nonylphenols (NP) are intermediate products of the degradation of a widely used class of surfactants and antioxidants: nonylphenol ethoxylates (NPE) (Engler, 2012). NP and NPE are organic chemicals used for many applications such as paints, pesticides, detergents and personal care products (US Environmental Protection Agency, 2010a). They can also be used as antioxidants and plasticizers for the production of plastics (Rani *et al.*, 2015; US Environmental Protection Agency, 2010a). Furthermore, NP have been found to leach out from plastic bottles to their water content (Loyo-Rosales *et al.*, 2004). Moreover, effluents from wastewater treatment plants are the major source of NP and NPE in the environment (Soares *et al.*, 2008). NP are considered as endocrine disruptors and their use is prohibited in the European Union due to their effects on the environment and human health (Rani *et al.*, 2015).

2.5. Antioxidants

Antioxidants are used as additives in many synthetic polymers including polyolefins (mainly PE and PP) which represent 60% of global demand for antioxidant additives (Höfer, 2012). Antioxidants are used to prevent the ageing of plastics and to delay oxidation (Lau and Wong, 2000). However, as with other plastic additives, antioxidants can leach out of the plastic and can migrate to food from plastic packaging and pose a threat in terms of food safety (Lau and Wong, 2000). Antioxidants from the Irganox® series are commonly used in plastics and they include Octadecyl 3-(3,5-di-*tert*-butyl-4-hydroxyphenyl)propionate (Irganox® 1076 – Pubchem: 16386), Pentaerythrityl-tetrakis-3-(3,5-di-*tert*-butyl-4-hydroxyphenyl)propionate (Irganox® 1010 – Pubchem ID: 64819) and 2,4-di-*tert*-butylphenol (Irgafos® 168 – Pubchem ID: 91601) (Lau and Wong, 2000).

Owing to the variety of plastic additives (BFR, phthalates, BPA, NP and antioxidants) used for plastic products conception and their detection in macro- and microplastic debris collected in environmental surveys, their occurrence in environmental matrices (water, sediment, biota) is expected and may pose major environmental concern as described below.

3. Plastic additives in the environment

3.1. Marine waters

Marine waters are affected by anthropogenic pressures as this natural compartment is the final receptacle of all discharge waters. Consequently, chemical pollutants including plastics additives have been detected in worldwide marine waters (Tables 4, 5 and 6) (Bergé *et al.*, 2013; Net *et al.*, 2015). Of all BFR, PBDE are the most commonly studied molecules in marine environments. PBDE have been widely found and multiple congeners have been monitored (Table 4). Concentrations varied from a few ng L^{-1} to more than 10 ng L^{-1} and congeners varied among the studied sites (Table 4).

Table 4: Concentrations of polybrominated diphenyl ether (PBDE) in seawater in ng L^{-1}

Location	ΣPBDE (ng L^{-1})	Range (ng L^{-1})	BDE congeners detected	Dominant congener	References
Port sea, Mediterranean Sea, Spain	23.2	4.2 – 19	BDE-28, -47	BDE-28	Sánchez-Avila *et al.* (2012)
Surface microlayer, China Sea, Hong-Kong	0.33	0.004 – 0.056	BDE-28, -47, -99, -100, -156, -183	BDE-156	Wurl *et al.* (2006)
Subsurface water, China Sea, Hong-Kong	0.1	0.002 – 0.082	BDE-28, -47, -99, -100, -183	BDE-47	Wurl *et al.* (2006)

Many studies on the contamination of the marine environment by phthalates showed concentrations ranging from a few pg L^{-1} to around 10 µg L^{-1}, with DEHP being the most concentrated phthalate found in marine waters (Table 5).

Table 5: Concentrations of phthalates in seawater in µg L^{-1}

Location	DMP	DEP	DnBP	DiBP	BBP	DEHP	DnOP	Reference
Tees Bay, UK	< 1 x 10^{-3}	0.025 – 0.5	0.47 – 0.55	0.66 – 1.1		0.98 – 2.2		Law *et al.* (1991)
North Sea, Germany	0.2 x 10^{-3}	0.67 x 10^{-2}	1.7 x 10^{-3}		0.05 x 10^{-3}	2.2 x 10^{-3}		Xie *et al.* (2005)
Surface waters, the Netherlands	0.004 – 0.49	0.07 – 2.3	<0.066 – 3.1		0.001 – 1.8	0.9 - 5	0.002 – 0.078	Vethaak *et al.* (2005)
Bay of Biscay, Spain	(7.5 ± 0.4) x 10^{-3}	(33 ± 3) x 10^{-3}	(83 ± 7) x 10^{-3}		(8 ± 1) x 10^{-3}	(64 ± 4) x 10^{-3}	(3.6 ± 0.4) x 10^{-3}	Prieto *et al.* (2007)
Coastal seawater, Mediterranean Sea, Spain	0.003 – 0.14	0.024 – 0.48			0.001 – 0.10	0.03 – 0.62		Sánchez-Avila *et al.* (2012)
Port sea, Mediterranean Sea, Spain	0.004 – 0.012	0.024 – 0.87			0.003 – 0.80	0.06 – 5.97		Sánchez-Avila *et al.* (2012)
River – sea interface, Mediterranean	0.005	0.07 – 0.16			0.003 – 0.07	0.02 – 0.21		Sánchez-Avila *et al.* (2012)

Sea, Spain							
Liguarian Sea, Mediterranean Sea, Italy [1]						18.38 ± 44.39	Fossi *et al.* (2012)
Sardinian Sea, Mediterranean Sea, Italy [1]						23.42 ± 32.46	Fossi *et al.* (2012)
Barkley Sound, Canada			0.18 – 3.0			0.01 – 0.95	Keil *et al.* (2011)
Puget Sound, USA						0.06 – 0.64	Keil *et al.* (2011)
Klang River estuary, Australia						3.10 – 64.3	Tan (1995)
Caspian Sea, Iran	0.49	0.52					Hadjmohammadi *et al.* (2011)
Arctic	40 x 10^{-6}	138 x 10^{-6}	51 x 10^{-6}	22 x 10^{-6}	8 x 10^{-6}	448 x 10^{-6}	Xie *et al.* (2007)

Nonylphenol was detected in marine waters of Europe, Asia and North America (Bergé *et al.*, 2012; David *et al.*, 2009) (Table 6) and concentrations ranged from 0.2 x 10^{-5} µg L^{-1} in the Sea of Japan to 4.6 µg L^{-1} in the Mediterranean Sea (Table 6). BPA, as for other additives, has been globally quantified in marine waters all around the world and concentrations ranged from ng L^{-1} in China to µg L^{-1} in coastal waters of Singapore (Table 6).

Table 6: Ranges of concentrations of nonylphenol and BPA in seawater in µg L^{-1}

Chemicals	Location	Concentrations (µg L^{-1})	References
Nonylphenol	German Bight, North Sea, Germany	0.006 – 0.033	Bester *et al.* (2001)
		9 x 10^{-5} – 0.0014	Xie *et al.* (2006)
	Estuaries, the Netherlands	0.031 – 0.934	Jonkers *et al.* (2003)
	Estuaries, UK	0.1 – 2.6	Blackburn *et al.* (1999)
	Mediterranean Sea, Spain	0.3 – 4.1	Petrovic *et al.* (2002)
	Venetian Lagoon, Italy	0.004 – 0.211	Pojana *et al.* (2007)
	Jamaica Bay, US	0.077 – 0.416	Ferguson *et al.* (2001)
	Masan Bay, South Korea	0.0097 – 0.928	Li *et al.* (2008)
	Sea of Japan, Japan	0.2 x 10^{-5} – 9.3 x 10^{-5}	Kannan *et al.* (1998)
BPA	Surface waters, the Netherlands	0.009 - 1	Vethaak *et al.* (2005)
		<0.012 – 0.33	Belfroid *et al.* (2002)
	Venetian Lagoon, Italy	<0.001 – 0.145	Pojana *et al.* (2007)
	Jiaozhou Bay, China	0.001 – 0.092	Fu *et al.* (2007)
	Estuaries, Japan	0.036 – 0.058	Kawahata *et al.* (2004)
	Coastal waters, Singapore	0.01 – 2.47	Basheer *et al.* (2004)

Overall, plastic additives have been detected worldwide in estuarine and marine waters at concentrations ranging from pg/L to µg/L with PBDE and DEHP being the most commonly reported congeners among BFR and phthalates, respectively. In addition to BPA and NP are also frequently detected in seawater. As most of the plastic additives exhibit high K_{ow}, higher concentrations are expected in sediment and marine organisms.

3.2. Sediment

As for marine waters, sediments are also affected by anthropogenic discharges and chemicals including plastic additives. Regarding BFRs, multiple BDE congeners have been found in marine sediments with BDE-209 being the major PBDE quantified in most studies at concentrations ranging from ng/kg to mg/kg (Table 7). In the Netherlands, HBCD were also found in sediments from the North Sea and Scheldt Estuary, respectively, at levels of 0.76 to 6.9 µg kg^{-1} dry weight (dw) and 30 to 71 µg kg^{-1} dw (Klamer *et al.*, 2005; Verslycke *et al.*, 2005).

Table 7: Concentrations of polybrominated diphenyl ether (PBDE) in marine sediments in μg kg^{-1} dry weight

Location	ΣPBDE (μg kg^{-1} dry weight)	Range (μg kg^{-1} dry weight)	BDE congeners detected	Most abundant congener	References
North Sea, the Netherlands	126.3	0.4 - 32	BDE-28, -47, -66, -71, -75, -77, -85, -99, -100, -119, -138, -153, -190, -209	BDE-209	Klamer *et al.* (2005)
Scheldt Estuary, the Netherlands	2198	0.2 – 1650	BDE-28, -47, -66, -85, -99, -100, -138, -153, -154, -209	BDE-209	Verslycke *et al.* (2005)
Coastal waters, South Korea	27.8	0.0037 – 27.4	BDE-3, -7, -15, -28, -47, -49, -66, -71, -77, -85, -99, -100, -119, -126, -138, -153, -154, -183, -209	BDE-209	Moon *et al.* (2007b)
Industrialized bays, South Korea	357.8	0.0012 - 283	BDE-3, -7, -15, -28, -47, -49, -66, -71, -77, -85, -99, -100, -119, -126, -138, -153, -154, -183, -209	BDE-209	Moon *et al.* (2007a)

TBBPA was also found in the Scheldt Estuary at levels below 0.1 µg kg^{-1} dw (Verslycke *et al.*, 2005). In their study, Klamer *et al.* (2005) also reported the presence of phthalates in North Sea sediments, namely dimethyl phthalate (DMP – Pubchem ID: 8554), diethyl phthalate (DEP – Pubchem ID: 6781), DBP, BBP, DEHP and DOP with DEHP being the most concentrated phthalate with 170 to 3,390 µg kg^{-1}. Phthalates in marine sediments from the Gulf of Mexico were detected on average at 7.6 and 6.6 µg kg^{-1} dw for di-*n*-butyl phthalate (DnBP – Pubchem ID: 3026) and DEHP respectively (Giam *et al.*, 1978). In Singapore Bay, phthalates reached 890 to 2,790 µg kg^{-1} dw for DEHP (Chee *et al.*, 1996). For nonylphenol (Bergé *et al.*, 2012; David *et al.*, 2009), concentrations ranged from less than 1 µg kg^{-1} dw in estuaries in the Netherlands to more than 20,000 µg kg^{-1} dw in the sediments of Tokyo Bay (Table 8). Like BFRs, NP and phthalates, BPA has also been found worldwide in sediments (Table 8). Indeed, BPA concentrations ranged from a few µg kg^{-1} dw in Japan and China to hundreds of µg kg^{-1} dw in the Venetian Lagoon (Table 8).

Whether the plastic additives detected in marine sediments come from diffuse sources (wastewater, atmospheric deposition, sewage sludge, etc.) or leachate from plastic debris is unclear even though an increasing amount of evidence (Al-Odaini *et al.*, 2015) suggests that microplastic and plastic debris in general likely constitute sources of plastic additives in the marine environment.

Table 8: Ranges of concentrations of nonylphenol and BPA in marine sediments in µg kg $^{-1}$ dry weight. nd: not detected.

Localization	Nonylphenol range (µg kg $^{-1}$ dry weight)	BPA range (µg kg $^{-1}$ dry weight)	Reference
North Sea, Germany	13 – 55		Bester *et al.* (2001)
Estuaries, the Netherlands	0.9 – 1080		Jonkers *et al.* (2003)
Mediterranean Sea, Spain	18 – 590		Petrovic *et al.* (2002)
Venetian Lagoon, Italy	47 – 192		Pojana *et al.* (2007)
Jamaica Bay, US	7 – 13,700		Ferguson *et al.* (2001)
Southern California Bight, US	130 – 3200		Schlenk *et al.* (2005)
Masan Bay, South Korea	92 – 557		Li *et al.* (2008)
Tokyo Bay, Japan	142 – 20,700		Kurihara *et al.* (2007)
	120 - 640		Isobe *et al.* (2001)
The Netherlands		<1.1 - 43	Vethaak *et al.* (2005)
Venetian Lagoon, Italy		<2.0 – 118	Pojana *et al.* (2007)
Jiaozhou Bay, China		0.7 – 17	Fu *et al.* (2007)
Estuaries, Japan		nd – 2.7	Kawahata *et al.* (2004)

3.3. Microplastics

To date, only a few studies have focused on the detection of plastic additives from MP collected in marine environments (Faure *et al.*, 2015; Fries *et al.*, 2013; Hirai *et al.*, 2011; Mato *et al.*, 2001; Rani *et al.*, 2015; Rochman *et al.*, 2014). Mato *et al.* (2001) detected nonylphenols in PP pellets deployed in Tokyo Bay and suggested that these compounds came from plastic additives found in the PP pellets themselves. In another study, Hirai *et al.* (2011) measured high concentrations of PBDEs, BPA and nonylphenols in PE and PP fragments collected on remote or urban beaches and in the open ocean. It was stated that they originated from plastic additives used for the manufacture of PP and PE. A wide range of plastic additives were also identified using Pyrolysis-GC/MS with thermal desorption in MP collected from sediment of Norderney Island (Fries *et al.*, 2013). MP particles were identified as PE, PP, PS and polyamide-6 (PA-6). They were associated with DEHP, DnBP, diisobutyl phthalate (DiBP – Pubchem ID: 6782), and 2,4-di-*tert*-butylphenol (2,4-DTBP – Pubchem ID: 7311), used here as antioxidant additives for PE and PP, DnBP, DiBP, DEP and DMP for PS, and DEHP, and DiBP and DEP for PA-6 (Fries et al., 2013). Moreover, Rani *et al.* (2015) detected multiple plastic additives in plastic marine debris found on a beach in Geoje, South Korea. Indeed, the authors found BPA and phthalates in PP and PE plastic marine debris as well as antioxidants including Irganox 76 and 2,4-DTBP in PP and PE plastic marine debris. In a study dealing with plastic debris in the Atlantic Ocean, BPA, PBDEs and 4-nonylphenol were detected in plastic samples found at sea and the authors suggested that this chemical came mainly from plastic additives (Rochman *et al.*, 2014). Moreover, some plastic additives were detected at concentrations up to 6 orders of magnitude higher than the concentrations measured in the surrounding water (Rochman *et al.*, 2014). In a more recent study, Faure *et al.* (2015) quantified MP pollution in Swiss lakes and detected MP associated with plastic additives including PBDEs, NPs, BPA and phthalates at concentrations comparable to those reported in marine studies (from 10^{-1} to 10^{6} ng g^{-1}) (Faure *et al.*, 2015).

These six studies demonstrated that plastic additives, some of which are known to be potential endocrine disruptors, are quantifiable in MPs found in sediments or in marine waters, suggesting that leaching of additives occurs in the environment. This is clearly of great concern as microplastics exhibit a high propensity to enter all trophic levels due to their small size and ubiquity in marine environments, and given the fact that leaching may also occur in the digestive conditions of organisms upon MP ingestion.

4. Transfer and toxicity of plastics additives for marine organisms

4.1. Contamination of marine organisms by plastic additives

PBDE have been detected in tissues of numerous marine organisms such as bivalves (Σ_{13}BDE ranged from 6.6 to 440 µg kg^{-1} lipid weight) (Bellas *et al.*, 2014; Johansson *et al.*, 2006; Ramu *et al.*, 2007), fish (Σ_7BDE ranged from 30.6 to 281 µg kg^{-1} lipid weight) (Peng *et al.*, 2007) and mammals (around 100 µg kg^{-1} wet weight (ww) in sperm whale (*Physeter microcephalus*) blubber) (de Boer *et al.*, 1998), suggesting that transfer from seawater, food or plastics to organisms occurs. In their work on the contamination of the Scheldt Estuary, Verslycke *et al.* (2005) found PBDE in sediment and in mysid shrimp (*Neomysis integer*) living in this estuary (Σ_{10}BDE: 2095 to 3562 ng g^{-1} lipid weight), and they highlighted that bioaccumulation was highest for BDE-47, -99 and -100 and lowest for BDE-209 because (i) highest brominated accumulate slower than lowest brominated congeners in the marine environment and (ii) they can be debrominated photolytically or biologically (Verslycke *et al.*, 2005). Phthalates (DMP, DEP, DiBP, DnBP, BBP, DEHP, DnOP, DnNP) were found in a wide range of organisms, including 18 different species ranging from primary producers (plankton and macroalgae) to picked dogfish (*Squalus acanthias*), but no biomagnification of the studied phthalates was observed through the food web (Mackintosh *et al.*, 2004). Recently, Cheng *et al.*, (2013) also detected phthalates (DMP, DEP, dipropyl phthalate (DPRP – PubChem ID: 8559), DiBP, DnBP, 2-Methoxyethyl phthalate (DMEP – PubChem ID: 8344), DHP, BBP, DEHP, DOHP, DnOP, DNP+DiDP) in fish at concentrations ranging from 0.2 to 1.23 µg g^{-1} ww (Cheng *et al.*, 2013). NP has been detected in many organisms commonly consumed as seafood products including oysters (*Crassostrea gigas*) (Cheng *et al.*, 2006), mussels (*Perna perna*) (Isobe *et al.*, 2007) and fishes (Ferrara *et al.*, 2008). For instance, Basheer *et al.* (2004) found NP and BPA in multiple fresh seafood products, including prawns (*Penaeus monodon*), crabs (*Portunus pelagicus*), blood cockles (*Anadara* *granosa*), white clams (*Meretrix meretrix*), squid (*Loligo* sp.) and fish (*Decapterus russelli*), from a supermarket in Singapore.

Overall, these results suggest that contamination of marine organisms by plastic additives may occur *via* natural pathways (*i.e.* waterborne or foodborne exposure) or *via* ingestion of plastic debris including MP.

4.2. Plastics additive transfer to marine organisms

4.2.1. Evidence from laboratory experiments

To investigate the potential leaching of additives from MP in environments characterized by different conditions (pH, temperature, salinity, etc), several laboratory studies have been conducted over the last years. First, the influence of gut surfactant was tested on the desorption of adsorbed chemicals, including perfluorooctanoic acid (PFOA – Pubchem ID: 9554) and DEHP, from PVC and PE in a study undertaken by Bakir *et al.* (2014). Desorption was higher in gut surfactant at 38°C (*i.e.* warm blooded animals) than in gut surfactant at 18°C (*i.e.* cold blooded animals) and in seawater at 18°C for DEHP. PFOA exhibited low affinity for PVC or PE regardless of the surfactant (Bakir *et al.*, 2014). The same authors suggested that the passage of plastic through the gut could enhance desorption of pollutants and act as a transfer route for accumulation of these pollutants. However, in a more recent study, Bakir *et al.* (2016) demonstrated, using a one-compartment model, that MP do not provide an additional pathway for the transfer of adsorbed chemicals, including DEHP and PFOA, from seawater to marine organisms even if MP transits through the gut. Some laboratory studies have used MP in the presence of additives to determine if these chemicals can transfer to organisms. For instance, Chua *et al.* (2014) exposed the marine amphipod *Allorchestes compressa* to PBDE in the presence or absence of microbeads with PBDEs adsorbed on microbeads. Both microbead ingestion and PBDE transfer *via* the microbeads were demonstrated at the end of the exposure. However, concentrations of PBDEs were lower in amphipods exposed to PBDE adsorbed on microbeads than in amphipods exposed to PBDEs without microbeads (Chua *et al.*, 2014) suggesting that transfer of PBDE adsorbed on MP can occur but at a lesser extent than the transfer via water. Similarly, Wardrop *et al.* (2016) exposed rainbow fish (*Melanotaenia fluviatilis*) to microbeads spiked with PBDEs (BDE-28, -47,-100, -99, -153, -154, -183 and -209) and compared them to control fish and fish exposed to microbeads alone. Here, PBDEs were analyzed in fish tissues excluding the stomach, liver, gall bladder and gonads to exclude spiked microbeads from the PBDEs analyses. During exposure, fish exposed to microbeads spiked with PBDEs showed higher concentrations than the two controls, and lower brominated congeners were better transferred in fish tissues than higher brominated congeners. On the other hand control fish and fish exposed to PBDE-free microbeads showed the same low levels of PBDEs concentration in their tissues suggesting that MP do not reduce contaminant body burden as it was previously hypothesized (Koelmans *et al.*, 2013a, b; Koelmans *et al.*, 2016). More realistic experiments have been performed using plastics incubated or collected in natural environments. For instance Rochman *et al.* (2013) used low-density polyethylene (LDPE) pellets deployed in seawater for two months and showed that the LDPE pellets adsorbed chemicals from the surrounding environment. Exposure of Japanese medaka (*Oryzias latipes*) to these pellets resulted in the accumulation of significant amounts of PBDEs and was associated with liver toxicity and pathology including glycogen depletion and cell necrosis for example (Rochman *et al.*, 2013). Bioaccumulation of PBDEs was also demonstrated in a terrestrial invertebrate, the house cricket (*Acheta domesticus*), as a result of PUR foam ingestion (Gaylor *et al.*, 2012). Another laboratory study demonstrated that the transfer of nonylphenol, triclosan and PBDE-47 can occur *via* MP ingestion in the lugworm (*Arenicola marina*) with possible effects on lugworm behavior (Browne *et al.*, 2013).

Overall, these laboratory experiments demonstrated transfer of plastic additives upon MP ingestion, sometimes in association with toxicity or behavior change.

4.2.2. Evidence from field studies

Levels of accumulated plastic additives in the environment or organisms have often been considered as a proxy indicator of plastic exposure in the marine environment as a consequence of the release of additives from plastic debris. For instance, a study on *Puffinus tenuirostris* showed that this bird ingested plastics at sea and that these plastics transferred flame retardant additives (PBDE) including BDE-209, which is specific to plastic (Tanaka *et al.*, 2013). In another study, the authors demonstrated that the transfer of PBDE may occur mainly by plastic ingestion through exposure by prey ingestion (Tanaka *et al.*, 2015). In another study, Rochman *et al.* (2014) examined the possible relationship between plastic densities at sea and levels of chemicals in fish inhabiting those areas. The results showed that PBDEs, especially the highest brominated congeners (BDE-209), may be an indicator of plastic pollution as previously suggested (Tanaka *et al.*, 2013). In the North Pacific Gyre, yellowtail (*Seriola lalandi*) were sampled to evaluate levels of plastic in the stomach and concentrations of pollutants and additives in their tissues (Gassel *et al.*, 2013). Ten percent of the yellowtail had ingested plastics, and PBDE and nonylphenol were concomitantly found in the fish tissues (Gassel *et al.*, 2013). Gassel *et al.* (2013) suggested that contamination of fish by nonylphenol and PBDE-209 could originate from the ingested plastic as mentioned above (Hirai *et al.*, 2011; Rochman *et al.*, 2014; Tanaka *et al.*, 2013; Teuten *et al.*, 2009). Other chemicals are also used as proxies for MP contamination such as DEHP (Fossi *et al.*, 2012; Fossi *et al.*, 2014; Fossi *et al.*, 2016). More recently, a study showed a higher accumulation of HBCDs in mussels (*Mytilus galloprovincialis*) inhabiting styrofoam debris (EPS) in comparison with mussels living on other plastic debris or rocks (Jang *et al.*, 2016). The authors also suggested that the isomeric profiles of detected HBCDs support the transfer of this flame retardant from the styrofoam debris to mussels through ingestion of EPS particles.

Field surveys showed that MP ingestion may constitute another route of transfer of plastic additives in marine organisms, leading to the use of plastic additives tissue content (mainly BDE-209 and DEHP) as a proxy for plastic exposure or ingestion.

4.3. Toxicity of plastic additives demonstrated by leaching experiments

Evidence for plastic toxicity has been rising in the last years. While direct toxicity can occur due to the physical impacts of plastic ingestion (for a review, see Wright *et al.* (2013)), indirect toxicity may be observed in relation to the release of hazardous chemicals from plastics. As most plastic additives are not chemically but physically bound to the plastic, they can be released into the environment and become available to organisms. Recent studies demonstrated, using leaching experiments, that various plastics are toxic to a wide range of organisms (Table 9).

427 Table 9: list of aquatic species, plastic polymer types, exposure times and endpoints used in various leaching experiments

Species	Plastic type	Exposure time	Exposure level	Endpoints	Reference
Daphnia magna	PC, PVC, PU, PE, LDPE, PMMA, PET, HDPE, PTFE, ABS, PP, MDPE	24 and 48 hours	70 – 100 g L^{-1}	Mortality	Lithner *et al.* (2009)
Daphnia magna	PP, HDPE, PVC, ABS, Epoxy resin	24 and 48 hours	Up to 250 g L^{-1}	Mortality	Lithner *et al.* (2012)
Nitroca sinipes	PP, PVC, PS, PET, PUR, LDPE, HDPE, ABS, PLA, Unknown	96 hours	100 g L^{-1}	Mortality	Bejgarn *et al.* (2015)
Amphibalanus amphitrite	PET, HDPE, PVC, LDPE, PP, PS, PC	24, 48, 72 and 96 hours	0.1 – 0.5 m² L^{-1}	Settlement	Li *et al.* (2016)
Perna perna	Virgin (PP) and beached pellets	48 hours	25% of pellets (v/v)	Embryo development	Gandara e Silva *et al.* (2016)
Pseudochromis fridmani	PE (two different origins)	48 hours	-	Mortality	Hamlin *et al.* (2015)

ABS: Acrylonitrile butadiene styrene; PC: Polycarbonate; PE: Polyethylene; LDPE: Low-Density Polyethylene; MDPE: Medium-Density Polyethylene; HDPE: High-Density Polyethylene; PET: Polyethylene terephthalate; PLA: Poly Lactic Acid; PMMA: Polymethyl Methacrylate; PP: Polypropylene; PTFE: Polytetrafluoroethylene; PU: Polyurethane; PVC: Polyvinyl Chloride

Li *et al.* (2016) used the seven categories of recyclable plastics (High Density PE (HDPE), LDPE, PP, PVC, Polycarbonate (PC), PET and PS) to quantify the impact of their leachate on the survival and settlement of barnacle *Amphibalanus amphitrite* larvae. Leachates were prepared by placing 1 x 1 cm pieces of each plastic in 20 mL of filtered seawater for 24h at 28°C (Li *et al.*, 2016). Survival was significantly lowered at the highest leachate concentration (0.10 and 0.50 m² of plastic material L^{-1}) for all plastics and PVC was the most toxic plastic for *A. Amphitrite* larvae. Additionally, settlement was also inhibited with all plastics leachates (Li *et al.*, 2016). Similarly, Bejgarn *et al.* (2015) exposed the copepod *Nitocra sinipes* to the leachate of weathered or non-weathered plastics. Here, leaching experiments were performed with leachates prepared with 10 g of each plastic placed in 100 mL of brackish seawater from the Baltic rotating at 6-21 rpm for 72h in the dark (Bejgarn *et al.*, 2015). Of the twenty-one plastics tested, eight (DVD-case (PP), biodegradable bag, costume- (PVC), flyswatter packaging (PVC), computer housing (unknown), garden hose (PVC), car dashboard (unknown) and phone cover (PUR)) demonstrated toxicity (mortality after 96h) to *N. sinipes*, and after weathering, toxicity either increased or decreased depending on the plastics (Bejgarn *et al.*, 2015). Two leaching studies were carried out on the copepod *Daphnia magna* (Lithner *et al.*, 2012; Lithner *et al.*, 2009), a common organism used in ecotoxicological studies. Lithner *et al.* (2009) prepared their leachates by placing plastic pieces in deionized water to obtain a liquid to solid ratio of 10 L kg^{-1} which was horizontally shaken at 60 rpm for 24h (16h of fluorescent light and 8h of darkness) at 20°C. Out of the thirty-two plastics tested only nine, including five composed of PVC, showed acute toxicity (immobility after 24 and 48h ; EC_{50} ranging from 5 to 80 g plastic material L^{-1}) to *D. magna* and it has been suggested that the toxicity of PVC was due to the phthalate content (Lithner *et al.*, 2009). In a second study, Lithner *et al.* (2012) used PP, PE, PVC, acrylonitrile-butadiene-styrene (ABS) and epoxy resin, and they prepared their leachates by adding 250 g of plastic in

1 L of deionized water shaken at 90 rpm at 50°C for 3 days. As previously demonstrated, PVC caused acute toxicity (immobility after 24 and 48h ; EC_{50} ranging from 2-235 g plastic material L^{-1}) probably in relation to its phthalate leachates, however the acute toxicity observed with the epoxy resin was not attributed to a specific chemical compound (Lithner *et al.*, 2012). A more recent study evaluated the toxicity of virgin and beached pellets on the embryo development of brown mussels (*Perna perna*) (Gandara e Silva *et al.*, 2016). Here, the authors exposed the brown mussel embryo to 2 mL of virgin (PP) or beached (42% PE and 58% unknown composition) pellets in 8 mL of seawater, and toxicity was assessed by determining the percentage of dead or abnormal embryos (Gandara e Silva *et al.*, 2016). The leaching experiment led to 23.5% and 100% dead or abnormal embryos for virgin and beached pellets, respectively. It has been suggested that the difference in toxicity was mainly due to the difference in the chemical load of the pellets used (Gandara e Silva *et al.*, 2016). Beached pellets were exposed to *in situ* contamination leading to adsorption of pollutants and to additives already found inside the polymeric matrix. These leaching experiments showed that plastic leachates and especially PVC leachates (*i.e.* phthalates) can lead to adverse effects on organisms. However, the toxicity highlighted in these five experiments was not attributed to specific chemical compounds (Bejgarn *et al.*, 2015; Gandara e Silva *et al.*, 2016; Li *et al.*, 2016; Lithner *et al.*, 2012; Lithner *et al.*, 2009). As suggested by Li *et al.* (2016), chemical identification should be undertaken during leaching experiments with a focus on plastic additives in order to identify the compound or its degradation products responsible for the observed toxicity. For instance, a more recent leaching study focused on the effects of nonylphenol on the coral reef fish *Pseudochromis fridmani* by exposing single fish to the leachate of plastic bags made of two PE (PE1 and PE2) from different manufacturers for 48h (Hamlin *et al.*, 2015). Nonylphenol leached in the water at higher concentrations for PE2 than for PE1; with respectively 41.0 ± 5.5 and 2.5 ± 0.2 µg L^{-1}, and 60% and 11% of the fish died

after 48 hours of exposure to leachates from PE2 and PE1, respectively. However, Hamlin *et al.* (2015) only focused their work on nonylphenol and did not study PE1 and PE2 compositions in terms of other additive contents. This study demonstrated that exposing fish to two identical plastic polymers (PE) may result in drastically different outcomes, as the plastic additives incorporated in each plastics are dependent on the plastic manufacturer and most of the time, their exact compositions remain unknown (Hamlin *et al.*, 2015). Studies are required to explore potential differences between plastics from different manufacturers and toxicity related to the diversity of chemicals used in the plastic industry.

Exposure experiments based on leaching processes conducted on a wide range of polymers and target organisms confirmed toxicity of plastics additives, which highlights the need for non-target screening analysis covering a broad range of chemicals in order to better identify the main compound(s) responsible for the toxicity.

4.4. Relative importance of HOC in comparison with plastic additives: case of modelling studies

The high affinity of plastic polymers for HOC has been demonstrated in numerous laboratory experiments (Bakir *et al.*, 2014; Teuten *et al.*, 2007), and an increasing number of studies have focused on the role of MP as a vector for HOC in marine organisms (Besseling *et al.*, 2013; Rochman *et al.*, 2013). However, recent studies have suggested that given (i) baseline contamination levels of seawater and marine organisms, and (ii) the low abundance of plastic particles relative to other suspended particles found in oceans (such as organic matter, plankton, detritus etc), exposure to HOC *via* plastic may be negligible compared to natural pathways (Bakir *et al.*, 2016; Beckingham and Ghosh, 2017; Koelmans *et al.*, 2013a, b; Koelmans *et al.*, 2016; Paul-Pont *et al.*, 2016). Moreover, Koelmans *et al.* (2016) suggested that MPs ingestion by marine biota does not increase their exposure to HOCs and could have a cleaning effect while concerns have arisen regarding risk due to plastic additives.

So far most modelling studies have focused their work on adsorbed HOC (Bakir *et al.*, 2016; Koelmans *et al.*, 2016). However, no model is yet available on the transport and fate of plastic additives leaching from plastic debris although (i) plastic additives can be added in very high concentration depending on the application; and (ii) transfer of plastic additives to marine organism upon plastic ingestion has been demonstrated both in laboratory experiments and in field studies. It highlights the need to include these chemicals in future modelling work in order to better clarify their potential for transfer.

5. Conclusion

Plastic additives associated with MP have received less attention than HOC adsorbed on MPs and the present review highlighted the need for upcoming studies to better characterize plastic additives associated with microplastics found at sea as well as their potential release in environmental matrices. As PE and PP are the main plastic debris found at sea, these two polymers should be investigated alongside with PVC due to its particularly high concentration in hazardous additives. Non-target screening analysis is required to identify the broad range of plastic additives leaching from these polymers and to better identify the main compound(s) responsible for toxicity. In addition, special attention should definitely be paid to hazardous plastic additives known to be major endocrine disruptor, namely bisphenol A and phthalates. Experimental and modelling studies are required to better characterize (i) the transfer of plastic additives upon MP ingestion relative to waterborne and foodborne exposure, and (ii) the effects of plastic additives on marine organisms. Such experiments should be realized using standardized “laboratory-made” MP, in which plastic additives are well characterized and introduced in controlled amounts reflecting industrial processes. The impacts of ageing plastic under realistic conditions on the transfer of plastic additives also need to be evaluated to investigate more environmentally relevant scenarios.

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