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# Review

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



Chemosphere

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# 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.

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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 plastic 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 polybrominated 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.

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# 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%-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, 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, 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).

| Ta | ble | 1 |
|----|-----|---|
|    |     |   |

| List of the most commonly produce | l polymers and their associated p | plastic additives. Adap | oted from Hansen et al. ( | 2013). |
|-----------------------------------|-----------------------------------|-------------------------|---------------------------|--------|
|-----------------------------------|-----------------------------------|-------------------------|---------------------------|--------|

| Polymer | Consumption in the EU27 (in million tons) in 2015 <sup>a</sup> | Additive types                    | Amount in polymers<br>(% w/w) | Hazardous substances <sup>b</sup>       |
|---------|--|-----------------------------------|-------------------------------|---|
| PP      | 9  | Antioxidant                       | 0.05-3                        | Bisphenol A; Octylphenol; Nonylphenol   |
|         |  | Flame retardant (cable insulation | 12-18                         | Brominated flame retardant; Boric acid; |
|         |  | and electronic applications)      |                               | Tris(2-chloroethyl)phosphate            |
| HDPE    | 8  | Antioxidant                       | 0.05-3                        | Bisphenol A; Octylphenol; Nonylphenol   |
|         |  | Flame retardant (cable            | 12-18                         | Brominated flame retardant; Boric acid; |
|         |  | insulation application)           |                               | Tris(2-chloroethyl)phosphate            |
| LDPE    | 6  | Antioxidant                       | 0.05-3                        | Bisphenol A; Octylphenol; Nonylphenol   |
|         |  | Flame retardant (cable            | 12-18                         | Brominated flame retardant; Boric acid; |
|         |  | insulation application)           |                               | 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            |

<sup>a</sup> According to PlasticsEurope (2016); PP: Polypropylene; HDPE: High Density Polyethylene; LDPE: Low Density Polyethylene; PVC: Polyvinyl Chloride; PUR: Polyurethane. <sup>b</sup> 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.

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).

# 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(2,4,6tribromophenoxy)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

#### Table 3

Plastic additives and their associated octanol-water partition coefficients (Log K<sub>ow</sub>). 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 <sub>ow</sub> |
|-------------------------------|--------------|---------------------|
| 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           |

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

#### 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 |
| Nonylphenol                       | Antioxidant and plasticizer in some plastics   | Endocrine disruptors           |
| Bisphenol A (BPA)                 | Monomer in polycarbonate and epoxy resins.   | Endocrine disruptors           |
| Irganox®                          | Antioxidant in some plastics.  | Estrogen minne                 |

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:  $\alpha$ -,  $\beta$ -, and  $\gamma$ -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, Authorization 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 und Umwelt, 2006). As a result, PVC can contain 10%–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 und Umwelt, 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<sup>®</sup> series are commonly used in plastics and they include Octadecyl 3-(3,5-di*tert*-butyl-4-hydroxyphenyl)propionate (Irganox<sup>®</sup> 1076 – Pubchem: 16386), Pentaerythrityl-tetrakis-3-(3,5-di*tert*-butyl-4-hydroxyphenyl)propionate (Irganox<sup>®</sup> 1010 – Pubchem ID: 64819) and 2,4-di-*tert*-butylphenol (Irgafos<sup>®</sup> 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 4–6) (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 of polybrominated | diphenyl ether | (PBDE) in seawater | $r$ in ng $L^{-1}$ . |
|----------------------------------|----------------|--------------------|----------------------|
|----------------------------------|----------------|--------------------|----------------------|

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

#### Table 5

Concentrations of phthalates in seawater in  $\mu g L^{-1}$ .

| Location   | DMP  | DEP   | DnBP  | DiBP                | BBP  | DEHP  | DnOP                        | Reference  |
|--|--|---|---|---------------------|--|---|-----------------------------|--|
| Tees Bay, UK<br>North Sea, Germany<br>Surface waters, the<br>Netherlands       | $\begin{array}{c} <\!1\times10^{-3}\\ 0.2\times10^{-3}\\ 0.004{-}0.49 \end{array}$ | $\begin{array}{c} 0.025{-}0.5\\ 0.67\times10^{-2}\\ 0.07{-}2.3 \end{array}$ | $\begin{array}{l} 0.47 {-} 0.55 \\ 1.7 \times 10^{-3} \\ {<} 0.066 {-} 3.1 \end{array}$ | 0.66–1.1            | $\begin{array}{c} 0.05\times 10^{-3}\\ 0.001{-}1.8\end{array}$ | $\begin{array}{c} 0.98{-}2.2\\ 2.2\times10^{-3}\\ 0.9{-}5\end{array}$ | 0.002-0.078                 | Law et al. (1991)<br>Xie et al. (2005)<br>Vethaak<br>et al. (2005) |
| Bay of Biscay, Spain<br>Coastal seawater,<br>Mediterranean Sea,<br>Spain       | $(7.5 \pm 0.4) \times 10^{-3}$<br>0.003-0.14                                       | $(33 \pm 3) \times 10^{-3}$<br>0.024-0.48                                   | $(83 \pm 7) \ge 10^{-3}$  |                     | $(8 \pm 1) \ge 10^{-3}$<br>0.001-0.10                          | $(64 \pm 4) \times 10^{-3}$<br>0.03-0.62                              | $(3.6 \pm 0.4) \ge 10^{-3}$ | Prieto et al. (2007)<br>Sánchez-Avila<br>et al. (2012)             |
| Port sea, Mediterranean<br>Sea. Spain  | 0.004-0.012  | 0.024-0.87  |   |                     | 0.003-0.80   | 0.06-5.97   |                             | Sánchez-Avila<br>et al. (2012)                                     |
| River — sea interface,<br>Mediterranean Sea,<br>Spain                          | 0.005  | 0.07–0.16   |   |                     | 0.003-0.07   | 0.02-0.21   |                             | Sánchez-Avila<br>et al. (2012)                                     |
| Liguarian Sea,<br>Mediterranean Sea,<br>Italy <sup>a</sup>                     |  |   |   |                     |  | 18.38 ± 44.39   |                             | Fossi et al. (2012)  |
| Sardinian Sea,<br>Mediterranean Sea,<br>Italy <sup>a</sup>                     |  |   |   |                     |  | 23.42 ± 32.46   |                             | Fossi et al. (2012)  |
| Barkley Sound, Canada<br>Puget Sound, USA<br>Klang River estuary,<br>Australia |  |   | 0.18–3.0  |                     |  | 0.01-0.95<br>0.06-0.64<br>3.10-64.3                                   |                             | Keil et al. (2011)<br>Keil et al. (2011)<br>Tan (1995)             |
| Caspian Sea, Iran  | 0.49   | 0.52  |   |                     |  |   |                             | Hadjmohammadi<br>et al. (2011)                                     |
| Arctic   | $40 \times 10^{-6}$  | $138\times10^{-6}$  | $51 	imes 10^{-6}$  | $22 \times 10^{-6}$ | $8\times10^{-6}$   | $448 \times 10^{-6}$  |                             | Xie et al. (2007)  |

<sup>a</sup> Data in ng/g fresh weight.

#### Table 6

Ranges of concentrations of nonylphenol and BPA in seawater in  $\mu g \; L^{-1}\!.$ 

| Chemicals   | Location                         | Concentrations ( $\mu g L^{-1}$ )       | References              |
|-------------|----------------------------------|---|-------------------------|
| Nonylphenol | German Bight, North Sea, Germany | 0.006-0.033                             | Bester et al. (2001)    |
|             |                                  | $9 \times 10^{-5} - 0.0014$             | 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              | $0.2 	imes 10^{-5} - 9.3 	imes 10^{-5}$ | 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)   |

(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).

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).

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  $0.2 \times 10^{-5} \,\mu g \, L^{-1}$  in the Sea of Japan to 4.6  $\,\mu g \, L^{-1}$  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  $\mu$ g  $L^{-1}$  in coastal waters of Singapore (Table 6).

Overall, plastic additives have been detected worldwide in estuarine and marine waters at concentrations ranging from pg/L to  $\mu$ g/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<sub>ow</sub>, higher concentrations are expected in sediment and marine organisms.

#### Table 7

| Concentrations of polybrominated of | diphenyl ether (PB | BDE) in marine sediments | in $\mu g \ kg^{-1}$ | dry weight. |
|-------------------------------------|--------------------|--------------------------|----------------------|-------------|
|-------------------------------------|--------------------|--------------------------|----------------------|-------------|

| Location                         | $\Sigma$ PBDE (µg kg <sup>-1</sup> dry weight) | Range (µg kg <sup>-1</sup><br>dry weight) | BDE congeners detected   | Most abundant<br>congener | References              |
|----------------------------------|--|---|--|---------------------------|-------------------------|
| North Sea, the Netherlands       | 126.3  | 0.4–32                                    | BDE-28, -47, -66, -71, -75, -77,<br>-85, -99, -100, -119, -138, -153,<br>-190, -209                          | BDE-209                   | Klamer et al. (2005)    |
| Scheldt Estuary, the Netherlands | 2198   | 0.2-1650                                  | BDE-28, -47, -66, -85, -99, -100,<br>-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,<br>-71, -77, -85, -99, -100, -119, -126,<br>-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,<br>-71, -77, -85, -99, -100, -119, -126,<br>-138, -153, -154, -183, -209 | BDE-209                   | Moon et al. (2007a)     |

#### Table 8

Ranges of concentrations of nonylphenol and BPA in marine sediments in  $\mu g kg^{-1} dry$  weight. nd: not detected.

| Chemicals   | Location                      | Concentrations ( $\mu g \ kg^{-1} \ dry \ weight$ ) | Reference              |
|-------------|-------------------------------|---|------------------------|
| Nonylphenol | 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)    |
| BPA         | 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) |

### 3.2. 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  $0.76-6.9 \ \mu g \ kg^{-1} \ dry \ weight (dw) \ and \ 30-71 \ \mu g \ kg^{-1} \ dw \ (Klamer$ et al., 2005; Verslycke et al., 2005).

TBBPA was also found in the Scheldt Estuary at levels below 0.1  $\mu$ g kg<sup>-1</sup> 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 3390  $\mu$ g kg<sup>-1</sup>. Phthalates in marine sediments from the Gulf of Mexico were detected on average at 7.6 and 6.6  $\mu$ g kg<sup>-1</sup> dw for di-*n*butyl phthalate (DnBP - Pubchem ID: 3026) and DEHP respectively (Giam et al., 1978). In Singapore Bay, phthalates reached 890 to 2790  $\mu$ g kg<sup>-1</sup> dw for DEHP (Chee et al., 1996). For nonylphenol (Bergé et al., 2012; David et al., 2009), concentrations ranged from less than 1  $\mu g\,kg^{-1}\,dw$  in estuaries in the Netherlands to more than 20,000  $\mu$ g kg<sup>-1</sup> 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  $\mu$ g kg<sup>-1</sup> dw in Japan and China to hundreds of  $\mu$ g kg<sup>-1</sup> 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.

# 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  $10^{-1}$ – $10^{6}$  ng g<sup>-1</sup>) (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 ( $\Sigma_{13}$ BDE ranged from 6.6 to 440 µg kg<sup>-1</sup> lipid weight) (Bellas et al., 2014; Johansson et al., 2006; Ramu et al., 2007), fish ( $\Sigma_7$ BDE ranged from 30.6 to 281 µg kg<sup>-1</sup> lipid weight) (Peng et al., 2007) and mammals (around 100  $\mu$ g kg<sup>-1</sup> 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 ( $\Sigma_{10}$ BDE: 2095–3562 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  $\mu$ g g<sup>-1</sup> 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, vellowtail (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 vellowtail 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, 2014, 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).

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  $1 \times 1$  cm pieces of each plastic in 20 mL of filtered seawater for 24 h at 28 °C (Li et al., 2016). Survival was significantly lowered at the highest leachate concentration (0.10 and 0.50  $m^2$  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, Beigarn 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., 2009, 2012), 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<sup>-1</sup> 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<sub>50</sub> ranging from 2 to 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

#### 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,<br>HDPE, PTFE, ABS, PP, MDPE | 24 and 48 h       | 70–100 g $L^{-1}$                      | Mortality  | Lithner et al. (2009)         |
| Daphnia magna           | PP, HDPE, PVC, ABS, Epoxy resin                                | 24 and 48 h       | Up to 250 g $L^{-1}$                   | Mortality  | Lithner et al. (2012)         |
| Nitroca sinipes         | PP, PVC, PS, PET, PUR, LDPE, HDPE,                             | 96 h              | 100 g L <sup>-1</sup>                  | Mortality  | Bejgarn et al. (2015)         |
| Amnhihalanus amnhitrite | PFT HDPF PVC IDPF PP PS PC                                     | 24 48 72 and 96 h | $0.1 - 0.5 \text{ m}^2 \text{ J}^{-1}$ | Settlement | Li et al. (2016)              |
| Perna perna             | Virgin (PP) and beached pellets                                | 48 h              | 25% of pellets (v/v)                   | Embryo     | Gandara e Silva et al. (2016) |
| Pseudochromis fridmani  | PE (two different origins)                                     | 48 h              | -                                      | 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: Poly-urethane; PVC: Polyvinyl Chloride.

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., 2009, 2012). 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  $\pm$  5.5 and 2.5  $\pm$  0.2  $\mu g$   $L^{-1}$  , and 60% and 11% of the fish died after 48 h 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 nontarget 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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