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► To cite this version:

Ludovic Hermabessiere, Alexandre Dehaut, Ika Paul-Pont, Camille Lacroix, Ronan Jézéquel, et al.. Occurrence and effects of plastic additives on marine environments and organisms: a review. Chemosphere, 2017, 182, pp.781-793. 10.1016/j.chemosphere.2017.05.096 . anses-02556379

HAL Id: anses-02556379

<https://anses.hal.science/anses-02556379v1>

Submitted on 28 Apr 2020

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1 **Occurrence and effects of plastic additives on marine environments and**
2 **organisms: a review**

3 Ludovic HERMABESSIERE^a, Alexandre DEHAUT^a, Ika PAUL-PONT^b, Camille
4 LACROIX^c, Ronan JEZEQUEL^c, Philippe SOUDANT^b & Guillaume DUFLOS^{a*}

5 ^a Anses, Laboratoire de Sécurité des Aliments, Boulevard du Bassin Napoléon, 62200
6 Boulogne-sur-Mer, France

7 ^b Laboratoire des Sciences de l'Environnement Marin (LEMAR),
8 UMR6539/UBO/CNRS/IRD/IFREMER, Institut Universitaire Européen de la Mer,
9 Technopôle Brest-Iroise, rue Dumont d'Urville, 29280 Plouzané, France

10 ^c CEDRE, 715 rue Alain Colas, 29218 Brest Cedex 2, France

11 * Corresponding author:

12 guillaume.duflos@anses.fr

13 Phone: + 33 3 21 99 25 00

14 Published in *Chemosphere* and available online at <https://doi.org/10.1016/j.chemosphere.2017.05.096>

15 **Abstract**

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

32 **Keywords**

33 Microplastics, Plastic additives, Bisphenol A, Phthalates, Brominated Flame retardant

34 **1. Introduction**

35 Due to their numerous societal benefits, plastics hold an important place in human society
36 ([Andrady and Neal, 2009](#)). Plastic, a man-made material, is inexpensive, strong, durable,
37 lightweight and easy to produce ([Thompson *et al.*, 2009](#)). As a consequence, plastic production
38 has been increasing since the 1950s, and notably rose from 225 million tons in 2004 to 322
39 million tons in 2015, representing a 43% increase over the last decade ([PlasticsEurope, 2016](#)).
40 However, this estimate does not take into account the proportion of synthetic fibers which are
41 widely used in the textile and fishery industries ([Dris *et al.*, 2016](#)) and there is an
42 underestimation of 15% to 20% depending on the year ([Industrievereinigung Chemiefaser,
43 2013](#)). Low estimates predicted that floating marine plastic weigh between 70,000 and 270,000
44 tons ([Cózar *et al.*, 2014](#); [Eriksen *et al.*, 2014](#); [Van Sebille *et al.*, 2015](#)). Small pieces of plastics
45 called microplastics (MP) account for a total of 51 trillion plastic debris ([Van Sebille *et al.*,
46 2015](#)).

47 Microplastics have been defined as plastics particles smaller than 5 mm ([Arthur *et al.*,
48 2009](#)). Growing attention has been accorded to microplastics during the last decade, since the
49 publication by [Thompson *et al.* \(2004\)](#). Micro-sized plastic pieces originate from two distinct
50 pathways: primary and secondary sources. Primary sources of MP correspond to (i) plastics that
51 are directly manufactured at micrometric size, including plastic pellets ([Barnes *et al.*, 2009](#);
52 [Cole *et al.*, 2011](#)), (ii) MP from exfoliating cosmetics ([Chang, 2015](#); [Fendall and Sewell, 2009](#);
53 [Napper *et al.*, 2015](#); [Zitko and Hanlon, 1991](#)) and (iii) clothing fibers found in wastewater
54 treatment plants ([Browne *et al.*, 2011](#); [Carr *et al.*, 2016](#)). Secondary MP results from the
55 breakdown of larger pieces due to mechanical abrasion and photochemical oxidation in the
56 environment ([Andrady, 2011](#); [Bouwmeester *et al.*, 2015](#); [Lambert and Wagner, 2016](#)). MP can
57 also degrade into smaller pieces called nanoplastics ([Gigault *et al.*, 2016](#); [Koelmans *et al.*, 2015](#);
58 [Lambert and Wagner, 2016](#)).

59 Due to their small size, MP can be ingested by a wide range of marine organisms such as
60 zooplankton, bivalves and worms (De Witte *et al.*, 2014; Devriese *et al.*, 2015; Graham and
61 Thompson, 2009; Rochman *et al.*, 2015; Sussarellu *et al.*, 2016; Van Cauwenberghe and
62 Janssen, 2014; Van Cauwenberghe *et al.*, 2015) and by organisms from higher trophic levels
63 such as fish (Boerger *et al.*, 2010; Carpenter *et al.*, 1972; Dantas *et al.*, 2012; Foekema *et al.*,
64 2013; Lusher *et al.*, 2013; Neves *et al.*, 2015; Possatto *et al.*, 2011; Rochman *et al.*, 2015) and
65 marine mammals (Eriksson and Burton, 2003; Lusher *et al.*, 2015). This ingestion of MPs can
66 result in physical damage such as obstruction or internal abrasions (Wright *et al.*, 2013). In
67 addition to these physical threats, MP can potentially transfer chemicals adsorbed on their
68 surface (Mato *et al.*, 2001; Teuten *et al.*, 2007; Teuten *et al.*, 2009) or plastic additives.
69 However, less attention has been paid to the transfer of plastic additives to marine organisms in
70 comparison with hydrophobic organic compounds (HOC), despite the fact that many additives
71 have been recognized as hazardous (Lithner *et al.*, 2011). Therefore, the transport and fate of
72 plastic additives leaching out from plastic debris should definitely be carefully addressed in
73 future field, laboratory and modelling works.

74 Plastics are made by polymerizing monomers and other substances (Lithner *et al.*, 2011)
75 including plastic additives. Plastic additives are chemical compounds, like plasticizers, which
76 provide required properties to a plastic polymer or are incorporated to facilitate the
77 manufacturing process (OECD, 2004). Moreover, some plastic additives are used as monomers,
78 for example bisphenol A is the monomer of polycarbonate (PC) but also a stabilizer in other
79 polymers. Plastic additives are mainly used as plasticizers, flame retardants, stabilizers,
80 antioxidants and pigments. Phthalates, BPA, nonylphenols, and brominated flame retardants
81 (BFR) are the most common additives recovered from the environment (Bergé *et al.*, 2012;
82 David *et al.*, 2009; de Boer *et al.*, 1998; de los Ríos *et al.*, 2012; Mackintosh *et al.*, 2004; Net
83 *et al.*, 2015; Xie *et al.*, 2005; Xie *et al.*, 2007) and represent a hazard to the environment and

84 organisms (Lithner *et al.*, 2011; Meeker *et al.*, 2009; Oehlmann *et al.*, 2009). These plastic
85 additives are released into the marine environment by numerous pathways including industrial
86 and municipal wastewater, atmospheric deposition, runoff and river transport resulting from
87 application of sewage sludge in agriculture. In addition leaching of plastic additives from macro
88 and microplastics is known to occur in the marine environment. Thus, the accumulation and
89 degradation of plastic debris might represent another major input of these chemical compounds
90 in oceans. As a consequence, more research is needed on the hazards of plastic additives
91 associated with microplastics.

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

97 **2. Chemicals used as plastic additives**

98 Multiple types and families of chemicals are mixed with polymers to produce plastics. The
99 type of additive depends on the plastic polymer and the requirements of the final product (Table
100 1).

101

102 **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

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 \(2017\)](#).

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

108 **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.	

109
 110 The main plastic additives described are listed in Table 3 with their associated octanol-water
 111 partition coefficient (K_{ow}). K_{ow} has been used for predicting how a chemical will concentrate in
 112 marine organisms and an increase in $\log K_{ow}$ indicates an increase in the potential
 113 bioconcentration in organisms (Net *et al.*, 2015).

114

115 **Table 3: Plastic additives and their associated octanol-water partition coefficients (Log K_{ow}).** Data were extracted from
 116 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- <i>n</i> -butyl phthalate	DnBP	4.27
di- <i>n</i> -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

117 **2.1. Brominated flame retardants**

118 Brominated flame retardants (BFR) are a class of additives used in plastic products to reduce
 119 flammability. These BFR are used in a variety of consumer products ranging from electronic
 120 devices to insulation foams. BFRs include a wide range of chemicals, of which polybrominated
 121 diphenyl ethers (PBDE), hexabromocyclododecane (HBCD – Pubchem ID: 18529) and
 122 tetrabromobisphenol A (TBBPA – Pubchem ID: 6618) ([Talsness et al., 2009](#)) represent the
 123 main BFRs used in the plastic industry. These 3 classes (PBDE, HBCD and TBBPA) are
 124 described in details below. Lately, attention has been given to other emerging BFRs such as
 125 1,2-bis(2,4,6-tribromophenoxy)ethane (BTBPE – Pubchem ID: 37840),
 126 decabromodiphenylethane (DBDPE – Pubchem ID: 10985889) and hexabromobenzene (HBB
 127 – Pubchem ID: 6905) as these have been identified in many environmental compartments,
 128 organisms, food and humans ([European Food Safety Authority, 2012](#)). As they are not
 129 chemically bound to the polymer matrix, they can leach into the surrounding environment

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

132 PBDEs are hydrophobic substances that include numerous formulations used in plastics as
133 flame retardants. Indeed, there are three main commercial formulations called penta-, octa- and
134 deca-BDEs (Chua *et al.*, 2014). These additives are ubiquitous, toxic, persistent and
135 bioaccumulate in the environment and are of great concern for human health (Engler, 2012).
136 As a result, penta- and octa-BDEs have been banned by the European Union since 2004
137 (European Directive, 2003), while deca-BDE was banned only in 2009 from electronic and
138 electrical applications in the European Union (European Council Decision, 2009). These
139 formulations can no longer be used in mixtures or products with a concentration higher than
140 0.1% by mass. In addition, tetra- to hepta-BDEs are listed for elimination in the Annex A of the
141 Stockholm Convention on persistent organic pollutant (POP) (Stockholm Convention, 2016).
142 Moreover, in Canada the use of tetra- to deca- BDE has been restricted under the SOR/2008-
143 218 Regulations (Consolidated Regulation, 2008). Since 2006, penta- to octa- BDE have been
144 subjected to a 90 day notification before importation or production in the US. Finally, deca-
145 BDE importation and production have been entirely stopped (US Environmental Protection
146 Agency, 2006, 2012) since 2013.

147 HBCD has three dominant stereoisomers: α -, β -, and γ -HBCD (European Food Safety
148 Authority, 2011a). These BFRs are listed as POPs in the Stockholm Convention (Stockholm
149 Convention, 2016) and the three isomers are subject to a request for authorization in the
150 European Union (European Council Regulation, 2006). HBCDs are found in expanded PS
151 (EPS) and extruded PS (XPS) up to 4-7% by weight (Al-Odaini *et al.*, 2015). Its use has been
152 subjected to authorization in the European Union since 2006 in the annex XIV of the
153 Registration, Evaluation, Authorisation and Restriction of Chemicals (REACH) regulation
154 (European Food Safety Authority, 2011a). Moreover, in 2013 HBCD was listed for elimination

155 in the Annex A of the Stockholm Convention with specific exemption for use and production
156 in EPS and XPS (Cruz *et al.*, 2015; Stockholm Convention, 2016). In the US, Environmental
157 Protection Agency conducted a risk assessment for HBCD according to the 2010 “Toxic
158 Substances Control Act” action plan (US Environmental Protection Agency, 2010b).

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

165 **2.2. Phthalates**

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

173 In 2015, 8.4 million tons of plasticizers were used around the world, and di(2-ethylexyl)
174 phthalate (DEHP – Pubchem ID: 8343) was the most commonly used plasticizer, representing
175 37.1% of the global plasticizer market (ECPI, 2016). Europe accounted for 1.3 million tons of
176 the global plasticizer market in 2015 (ECPI, 2016), but DEHP was not the most commonly used
177 plasticizer in Europe, as suggested by its 20% decrease in consumption between 1999 and 2004.
178 DEHP has gradually been replaced by diisononyl phthalate (DiNP – Pubchem ID: 590836),

179 diisodecyl phthalate (DiDP – Pubchem ID: 33599) and di(2-Propyl Heptyl) phthalate (DPHP –
180 Pubchem ID: 92344), which represented 57% of plasticizer consumption in Europe in 2015
181 ([Arbeitsgemeinschaft PVC und Umwelt e.V, 2006](#); [ECPI, 2016](#)).

182 **2.3. Bisphenol A**

183 BPA (Pubchem ID: 6623) is the most representative chemical of the bisphenol group and is
184 one of the most commonly produced chemicals worldwide, with over three million tons
185 produced annually ([Laing et al., 2016](#)). BPA is mainly used as a monomer for polycarbonate
186 (PC) plastics (65% of volume used) and epoxy resins (30% of volume used), which are for
187 instance the main component of the lining layer of aluminum cans ([Crain et al., 2007](#); [ICIS,
188 2003](#)). BPA can also be used as an antioxidant or as a plasticizer in other polymers (PP, PE and
189 PVC) ([Rani et al., 2015](#)). Leaching of BPA can occur ([Sajiki and Yonekubo, 2003](#)), leading to
190 the release of this additive from food and drink packaging, which is considered as a source of
191 exposure for human beings ([Vandermeersch et al., 2015](#)). Despite its potential to leach from
192 food packaging and the fact that it has been identified as a significant endocrine disruptor
193 ([Oehlmann et al., 2009](#)), BPA is still allowed in the European Union for use in food contact
194 material ([European Council Regulation, 2011](#)). Other bisphenol analogs, such as bisphenol B,
195 bisphenol F and bisphenol S are used in plastics and may represent a threat to the environment
196 even though their toxicity is still unknown ([Chen et al., 2016](#)).

197 **2.4. Nonylphenols**

198 Nonylphenols (NP) are intermediate products of the degradation of a widely used class of
199 surfactants and antioxidants: nonylphenol ethoxylates (NPE) ([Engler, 2012](#)). NP and NPE are
200 organic chemicals used for many applications such as paints, pesticides, detergents and personal
201 care products (US Environmental Protection Agency, 2010a). They can also be used as
202 antioxidants and plasticizers for the production of plastics ([Rani et al., 2015](#); US Environmental

203 Protection Agency, 2010a). Furthermore, NP have been found to leach out from plastic bottles
204 to their water content (Loyo-Rosales *et al.*, 2004). Moreover, effluents from wastewater
205 treatment plants are the major source of NP and NPE in the environment (Soares *et al.*, 2008).
206 NP are considered as endocrine disruptors and their use is prohibited in the European Union
207 due to their effects on the environment and human health (Rani *et al.*, 2015).

208 **2.5. Antioxidants**

209 Antioxidants are used as additives in many synthetic polymers including polyolefins
210 (mainly PE and PP) which represent 60% of global demand for antioxidant additives (Höfer,
211 2012). Antioxidants are used to prevent the ageing of plastics and to delay oxidation (Lau and
212 Wong, 2000). However, as with other plastic additives, antioxidants can leach out of the plastic
213 and can migrate to food from plastic packaging and pose a threat in terms of food safety (Lau
214 and Wong, 2000). Antioxidants from the Irganox® series are commonly used in plastics and
215 they include Octadecyl 3-(3,5-di-*tert*-butyl-4-hydroxyphenyl)propionate (Irganox® 1076 –
216 Pubchem: 16386), Pentaerythrityl-tetrakis-3-(3,5-di-*tert*-butyl-4-hydroxyphenyl)propionate
217 (Irganox® 1010 – Pubchem ID: 64819) and 2,4-di-*tert*-butylphenol (Irgafos® 168 – Pubchem
218 ID: 91601) (Lau and Wong, 2000).

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

223 3. Plastic additives in the environment

224 3.1. Marine waters

225 Marine waters are affected by anthropogenic pressures as this natural compartment is the
 226 final receptacle of all discharge waters. Consequently, chemical pollutants including plastics
 227 additives have been detected in worldwide marine waters (Tables 4, 5 and 6) (Bergé *et al.*,
 228 2013; Net *et al.*, 2015). Of all BFR, PBDE are the most commonly studied molecules in marine
 229 environments. PBDE have been widely found and multiple congeners have been monitored
 230 (Table 4). Concentrations varied from a few ng L⁻¹ to more than 10 ng L⁻¹ and congeners varied
 231 among the studied sites (Table 4).

232 **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 <i>et al.</i> (2012)
Surface microlayer, China Sea, Hong- Kong	0.33	0.004 – 0.056	BDE-28, -47, - 99, -100, -156, - 183	BDE-156	Wurl <i>et al.</i> (2006)
Subsurface water, China Sea, Hong- Kong	0.1	0.002 – 0.082	BDE-28, -47, - 99, -100, -183	BDE-47	Wurl <i>et al.</i> (2006)

233
 234 Many studies on the contamination of the marine environment by phthalates showed
 235 concentrations ranging from a few pg L⁻¹ to around 10 µg L⁻¹, with DEHP being the most
 236 concentrated phthalate found in marine waters (Table 5).

237 **Table 5: Concentrations of phthalates in seawater in $\mu\text{g L}^{-1}$**

Location	DMP	DEP	DnBP	DiBP	BBP	DEHP	DnOP	Reference
Tees Bay, UK	$< 1 \times 10^{-3}$	0.025 – 0.5	0.47 – 0.55	0.66 – 1.1		0.98 – 2.2		Law et al. (1991)
North Sea, Germany	0.2×10^{-3}	0.67×10^{-2}	1.7×10^{-3}		0.05×10^{-3}	2.2×10^{-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	$(7.5 \pm 0.4) \times 10^{-3}$	$(33 \pm 3) \times 10^{-3}$	$(83 \pm 7) \times 10^{-3}$		$(8 \pm 1) \times 10^{-3}$	$(64 \pm 4) \times 10^{-3}$	$(3.6 \pm 0.4) \times 10^{-3}$	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,	0.005	0.07 – 0.16			0.003 – 0.07	0.02 – 0.21		Sánchez-Avila et al. (2012)

239 Nonylphenol was detected in marine waters of Europe, Asia and North America ([Bergé et](#)
 240 [al., 2012](#); [David et al., 2009](#)) (Table 6) and concentrations ranged from $0.2 \times 10^{-5} \mu\text{g L}^{-1}$ in the
 241 Sea of Japan to $4.6 \mu\text{g L}^{-1}$ in the Mediterranean Sea (Table 6). BPA, as for other additives, has
 242 been globally quantified in marine waters all around the world and concentrations ranged from
 243 ng L^{-1} in China to $\mu\text{g L}^{-1}$ in coastal waters of Singapore (Table 6).

244 **Table 6: Ranges of concentrations of nonylphenol and BPA in seawater in $\mu\text{g L}^{-1}$**

Chemicals	Location	Concentrations ($\mu\text{g L}^{-1}$)	References
Nonylphenol	German Bight, North Sea,	0.006 – 0.033	Bester et al. (2001)
	Germany	9×10^{-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	0.2×10^{-5} – 9.3×10^{-5}	Kannan et al. (1998)
	BPA	Surface waters, the Netherlands	0.009 - 1
Venetian Lagoon, Italy		<0.012 – 0.33	Belfroid et al. (2002)
Jiaozhou Bay, China		<0.001 – 0.145	Pojana et al. (2007)
Estuaries, Japan		0.001 – 0.092	Fu et al. (2007)
		0.036 – 0.058	Kawahata et al. (2004)

Coastal waters, Singapore

0.01 – 2.47

Basheer *et al.*
(2004)

245

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

251 3.2. Sediment

252 As for marine waters, sediments are also affected by anthropogenic discharges and chemicals
253 including plastic additives. Regarding BFRs, multiple BDE congeners have been found in
254 marine sediments with BDE-209 being the major PBDE quantified in most studies at
255 concentrations ranging from ng/kg to mg/kg (Table 7). In the Netherlands, HBCD were also
256 found in sediments from the North Sea and Scheldt Estuary, respectively, at levels of 0.76 to
257 $6.9 \mu\text{g kg}^{-1}$ dry weight (dw) and 30 to $71 \mu\text{g kg}^{-1}$ dw (Klamer *et al.*, 2005; Verslycke *et al.*,
258 2005).

259

260 **Table 7: Concentrations of polybrominated diphenyl ether (PBDE) in marine sediments in $\mu\text{g kg}^{-1}$ dry weight**

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

261

262 TBBPA was also found in the Scheldt Estuary at levels below $0.1 \mu\text{g kg}^{-1} \text{ dw}$ (Verslycke *et al.*,
263 2005). In their study, Klamer *et al.* (2005) also reported the presence of phthalates in North Sea
264 sediments, namely dimethyl phthalate (DMP – Pubchem ID: 8554), diethyl phthalate (DEP –
265 Pubchem ID: 6781), DBP, BBP, DEHP and DOP with DEHP being the most concentrated
266 phthalate with 170 to $3,390 \mu\text{g kg}^{-1}$. Phthalates in marine sediments from the Gulf of Mexico
267 were detected on average at 7.6 and $6.6 \mu\text{g kg}^{-1} \text{ dw}$ for di-*n*-butyl phthalate (DnBP – Pubchem
268 ID: 3026) and DEHP respectively (Giam *et al.*, 1978). In Singapore Bay, phthalates reached
269 890 to $2,790 \mu\text{g kg}^{-1} \text{ dw}$ for DEHP (Chee *et al.*, 1996). For nonylphenol (Bergé *et al.*, 2012;
270 David *et al.*, 2009), concentrations ranged from less than $1 \mu\text{g kg}^{-1} \text{ dw}$ in estuaries in the
271 Netherlands to more than $20,000 \mu\text{g kg}^{-1} \text{ dw}$ in the sediments of Tokyo Bay (Table 8). Like
272 BFRs, NP and phthalates, BPA has also been found worldwide in sediments (Table 8). Indeed,
273 BPA concentrations ranged from a few $\mu\text{g kg}^{-1} \text{ dw}$ in Japan and China to hundreds of $\mu\text{g kg}^{-1}$
274 dw in the Venetian Lagoon (Table 8).

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

280

281 **Table 8: Ranges of concentrations of nonylphenol and BPA in marine sediments in $\mu\text{g kg}^{-1}$ dry weight. nd: not detected.**

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

282 **3.3. Microplastics**

283 To date, only a few studies have focused on the detection of plastic additives from MP
284 collected in marine environments (Faure *et al.*, 2015; Fries *et al.*, 2013; Hirai *et al.*, 2011; Mato
285 *et al.*, 2001; Rani *et al.*, 2015; Rochman *et al.*, 2014). Mato *et al.* (2001) detected nonylphenols
286 in PP pellets deployed in Tokyo Bay and suggested that these compounds came from plastic
287 additives found in the PP pellets themselves. In another study, Hirai *et al.* (2011) measured high
288 concentrations of PBDEs, BPA and nonylphenols in PE and PP fragments collected on remote
289 or urban beaches and in the open ocean. It was stated that they originated from plastic additives
290 used for the manufacture of PP and PE. A wide range of plastic additives were also identified
291 using Pyrolysis-GC/MS with thermal desorption in MP collected from sediment of Norderney
292 Island (Fries *et al.*, 2013). MP particles were identified as PE, PP, PS and polyamide-6 (PA-6).

293 They were associated with DEHP, DnBP, diisobutyl phthalate (DiBP – Pubchem ID: 6782),
294 and 2,4-di-*tert*-butylphenol (2,4-DTBP – Pubchem ID: 7311), used here as antioxidant
295 additives for PE and PP, DnBP, DiBP, DEP and DMP for PS, and DEHP, and DiBP and DEP
296 for PA-6 (Fries *et al.*, 2013). Moreover, Rani *et al.* (2015) detected multiple plastic additives in
297 plastic marine debris found on a beach in Geoje, South Korea. Indeed, the authors found BPA
298 and phthalates in PP and PE plastic marine debris as well as antioxidants including Irganox 76
299 and 2,4-DTBP in PP and PE plastic marine debris. In a study dealing with plastic debris in the
300 Atlantic Ocean, BPA, PBDEs and 4-nonylphenol were detected in plastic samples found at sea
301 and the authors suggested that this chemical came mainly from plastic additives (Rochman *et*
302 *al.*, 2014). Moreover, some plastic additives were detected at concentrations up to 6 orders of
303 magnitude higher than the concentrations measured in the surrounding water (Rochman *et al.*,
304 2014). In a more recent study, Faure *et al.* (2015) quantified MP pollution in Swiss lakes and
305 detected MP associated with plastic additives including PBDEs, NPs, BPA and phthalates at
306 concentrations comparable to those reported in marine studies (from 10^{-1} to 10^6 ng g⁻¹) (Faure
307 *et al.*, 2015).

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

314 **4. Transfer and toxicity of plastics additives for marine organisms**

315 **4.1. Contamination of marine organisms by plastic additives**

316 PBDE have been detected in tissues of numerous marine organisms such as bivalves
317 ($\Sigma_{13}\text{BDE}$ ranged from 6.6 to 440 $\mu\text{g kg}^{-1}$ lipid weight) (Bellas *et al.*, 2014; Johansson *et al.*,
318 2006; Ramu *et al.*, 2007), fish ($\Sigma_7\text{BDE}$ ranged from 30.6 to 281 $\mu\text{g kg}^{-1}$ lipid weight) (Peng *et*
319 *al.*, 2007) and mammals (around 100 $\mu\text{g kg}^{-1}$ wet weight (ww) in sperm whale (*Physeter*
320 *microcephalus*) blubber) (de Boer *et al.*, 1998), suggesting that transfer from seawater, food or
321 plastics to organisms occurs. In their work on the contamination of the Scheldt Estuary,
322 Verslycke *et al.* (2005) found PBDE in sediment and in mysid shrimp (*Neomysis integer*) living
323 in this estuary ($\Sigma_{10}\text{BDE}$: 2095 to 3562 ng g^{-1} lipid weight), and they highlighted that
324 bioaccumulation was highest for BDE-47, -99 and -100 and lowest for BDE-209 because (i)
325 highest brominated accumulate slower than lowest brominated congeners in the marine
326 environment and (ii) they can be debrominated photolytically or biologically (Verslycke *et al.*,
327 2005). Phthalates (DMP, DEP, DiBP, DnBP, BBP, DEHP, DnOP, DnNP) were found in a wide
328 range of organisms, including 18 different species ranging from primary producers (plankton
329 and macroalgae) to picked dogfish (*Squalus acanthias*), but no biomagnification of the studied
330 phthalates was observed through the food web (Mackintosh *et al.*, 2004). Recently, Cheng *et*
331 *al.*, (2013) also detected phthalates (DMP, DEP, dipropyl phthalate (DPRP – PubChem ID:
332 8559), DiBP, DnBP, 2-Methoxyethyl phthalate (DMEP – PubChem ID: 8344), DHP, BBP,
333 DEHP, DOHP, DnOP, DNP+DiDP) in fish at concentrations ranging from 0.2 to 1.23 $\mu\text{g g}^{-1}$
334 ww (Cheng *et al.*, 2013). NP has been detected in many organisms commonly consumed as
335 seafood products including oysters (*Crassostrea gigas*) (Cheng *et al.*, 2006), mussels (*Perna*
336 *perna*) (Isobe *et al.*, 2007) and fishes (Ferrara *et al.*, 2008). For instance, Basheer *et al.* (2004)
337 found NP and BPA in multiple fresh seafood products, including prawns (*Penaeus monodon*),

338 crabs (*Portunus pelagicus*), blood cockles (*Anadara granosa*), white clams (*Meretrix meretrix*),
339 squid (*Loligo* sp.) and fish (*Decapterus russelli*), from a supermarket in Singapore.

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

343 **4.2. Plastics additive transfer to marine organisms**

344 **4.2.1. Evidence from laboratory experiments**

345 To investigate the potential leaching of additives from MP in environments characterized
346 by different conditions (pH, temperature, salinity, etc), several laboratory studies have been
347 conducted over the last years. First, the influence of gut surfactant was tested on the desorption
348 of adsorbed chemicals, including perfluorooctanoic acid (PFOA – Pubchem ID: 9554) and
349 DEHP, from PVC and PE in a study undertaken by [Bakir *et al.* \(2014\)](#). Desorption was higher
350 in gut surfactant at 38°C (*i.e.* warm blooded animals) than in gut surfactant at 18°C (*i.e.* cold
351 blooded animals) and in seawater at 18°C for DEHP. PFOA exhibited low affinity for PVC or
352 PE regardless of the surfactant ([Bakir *et al.*, 2014](#)). The same authors suggested that the passage
353 of plastic through the gut could enhance desorption of pollutants and act as a transfer route for
354 accumulation of these pollutants. However, in a more recent study, [Bakir *et al.* \(2016\)](#)
355 demonstrated, using a one-compartment model, that MP do not provide an additional pathway
356 for the transfer of adsorbed chemicals, including DEHP and PFOA, from seawater to marine
357 organisms even if MP transits through the gut. Some laboratory studies have used MP in the
358 presence of additives to determine if these chemicals can transfer to organisms. For instance,
359 [Chua *et al.* \(2014\)](#) exposed the marine amphipod *Allorchestes compressa* to PBDE in the
360 presence or absence of microbeads with PBDEs adsorbed on microbeads. Both microbead
361 ingestion and PBDE transfer *via* the microbeads were demonstrated at the end of the exposure.

362 However, concentrations of PBDEs were lower in amphipods exposed to PBDE adsorbed on
363 microbeads than in amphipods exposed to PBDEs without microbeads (Chua *et al.*, 2014)
364 suggesting that transfer of PBDE adsorbed on MP can occur but at a lesser extent than the
365 transfer via water. Similarly, Wardrop *et al.* (2016) exposed rainbow fish (*Melanotaenia*
366 *fluviatilis*) to microbeads spiked with PBDEs (BDE-28, -47, -100, -99, -153, -154, -183 and -
367 209) and compared them to control fish and fish exposed to microbeads alone. Here, PBDEs
368 were analyzed in fish tissues excluding the stomach, liver, gall bladder and gonads to exclude
369 spiked microbeads from the PBDEs analyses. During exposure, fish exposed to microbeads
370 spiked with PBDEs showed higher concentrations than the two controls, and lower brominated
371 congeners were better transferred in fish tissues than higher brominated congeners. On the other
372 hand control fish and fish exposed to PBDE-free microbeads showed the same low levels of
373 PBDEs concentration in their tissues suggesting that MP do not reduce contaminant body
374 burden as it was previously hypothesized (Koelmans *et al.*, 2013a, b; Koelmans *et al.*, 2016).
375 More realistic experiments have been performed using plastics incubated or collected in natural
376 environments. For instance Rochman *et al.* (2013) used low-density polyethylene (LDPE)
377 pellets deployed in seawater for two months and showed that the LDPE pellets adsorbed
378 chemicals from the surrounding environment. Exposure of Japanese medaka (*Oryzias latipes*)
379 to these pellets resulted in the accumulation of significant amounts of PBDEs and was
380 associated with liver toxicity and pathology including glycogen depletion and cell necrosis for
381 example (Rochman *et al.*, 2013). Bioaccumulation of PBDEs was also demonstrated in a
382 terrestrial invertebrate, the house cricket (*Acheta domesticus*), as a result of PUR foam ingestion
383 (Gaylor *et al.*, 2012). Another laboratory study demonstrated that the transfer of nonylphenol,
384 triclosan and PBDE-47 can occur *via* MP ingestion in the lugworm (*Arenicola marina*) with
385 possible effects on lugworm behavior (Browne *et al.*, 2013).

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

388 **4.2.2. Evidence from field studies**

389 Levels of accumulated plastic additives in the environment or organisms have often been
390 considered as a proxy indicator of plastic exposure in the marine environment as a consequence
391 of the release of additives from plastic debris. For instance, a study on *Puffinus tenuirostris*
392 showed that this bird ingested plastics at sea and that these plastics transferred flame retardant
393 additives (PBDE) including BDE-209, which is specific to plastic (Tanaka *et al.*, 2013). In
394 another study, the authors demonstrated that the transfer of PBDE may occur mainly by plastic
395 ingestion through exposure by prey ingestion (Tanaka *et al.*, 2015). In another study, Rochman
396 *et al.* (2014) examined the possible relationship between plastic densities at sea and levels of
397 chemicals in fish inhabiting those areas. The results showed that PBDEs, especially the highest
398 brominated congeners (BDE-209), may be an indicator of plastic pollution as previously
399 suggested (Tanaka *et al.*, 2013). In the North Pacific Gyre, yellowtail (*Seriola lalandi*) were
400 sampled to evaluate levels of plastic in the stomach and concentrations of pollutants and
401 additives in their tissues (Gassel *et al.*, 2013). Ten percent of the yellowtail had ingested
402 plastics, and PBDE and nonylphenol were concomitantly found in the fish tissues (Gassel *et*
403 *al.*, 2013). Gassel *et al.* (2013) suggested that contamination of fish by nonylphenol and PBDE-
404 209 could originate from the ingested plastic as mentioned above (Hirai *et al.*, 2011; Rochman
405 *et al.*, 2014; Tanaka *et al.*, 2013; Teuten *et al.*, 2009). Other chemicals are also used as proxies
406 for MP contamination such as DEHP (Fossi *et al.*, 2012; Fossi *et al.*, 2014; Fossi *et al.*, 2016).
407 More recently, a study showed a higher accumulation of HBCDs in mussels (*Mytilus*
408 *galloprovincialis*) inhabiting styrofoam debris (EPS) in comparison with mussels living on
409 other plastic debris or rocks (Jang *et al.*, 2016). The authors also suggested that the isomeric

410 profiles of detected HBCDs support the transfer of this flame retardant from the styrofoam
411 debris to mussels through ingestion of EPS particles.

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

415 **4.3. Toxicity of plastic additives demonstrated by leaching experiments**

416 Evidence for plastic toxicity has been rising in the last years. While direct toxicity can occur
417 due to the physical impacts of plastic ingestion (for a review, see [Wright *et al.* \(2013\)](#)), indirect
418 toxicity may be observed in relation to the release of hazardous chemicals from plastics. As
419 most plastic additives are not chemically but physically bound to the plastic, they can be
420 released into the environment and become available to organisms. Recent studies demonstrated,
421 using leaching experiments, that various plastics are toxic to a wide range of organisms (Table
422 9).

423

424 **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
<i>Daphnia magna</i>	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)
<i>Daphnia magna</i>	PP, HDPE, PVC, ABS, Epoxy resin	24 and 48 hours	Up to 250 g L ⁻¹	Mortality	Lithner et al. (2012)
<i>Nitroca sinipes</i>	PP, PVC, PS, PET, PUR, LDPE, HDPE, ABS, PLA, Unknown	96 hours	100 g L ⁻¹	Mortality	Bejgarn et al. (2015)
<i>Amphibalanus amphitrite</i>	PET, HDPE, PVC, LDPE, PP, PS, PC	24, 48, 72 and 96 hours	0.1 – 0.5 m ² L ⁻¹	Settlement	Li et al. (2016)
<i>Perna perna</i>	Virgin (PP) and beached pellets	48 hours	25% of pellets (v/v)	Embryo development	Gandara e Silva et al. (2016)
<i>Pseudochromis fridmani</i>	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

426 [Li et al. \(2016\)](#) used the seven categories of recyclable plastics (High Density PE (HDPE),
427 LDPE, PP, PVC, Polycarbonate (PC), PET and PS) to quantify the impact of their leachate on
428 the survival and settlement of barnacle *Amphibalanus amphitrite* larvae. Leachates were
429 prepared by placing 1 x 1 cm pieces of each plastic in 20 mL of filtered seawater for 24h at
430 28°C ([Li et al., 2016](#)). Survival was significantly lowered at the highest leachate concentration
431 (0.10 and 0.50 m² of plastic material L⁻¹) for all plastics and PVC was the most toxic plastic for
432 *A. Amphitrite* larvae. Additionally, settlement was also inhibited with all plastics leachates ([Li](#)
433 [et al., 2016](#)). Similarly, [Bejgarn et al. \(2015\)](#) exposed the copepod *Nitocra sinipes* to the
434 leachate of weathered or non-weathered plastics. Here, leaching experiments were performed
435 with leachates prepared with 10 g of each plastic placed in 100 mL of brackish seawater from
436 the Baltic rotating at 6-21 rpm for 72h in the dark ([Bejgarn et al., 2015](#)). Of the twenty-one
437 plastics tested, eight (DVD-case (PP), biodegradable bag, costume- (PVC), flyswatter
438 packaging (PVC), computer housing (unknown), garden hose (PVC), car dashboard (unknown)
439 and phone cover (PUR)) demonstrated toxicity (mortality after 96h) to *N. sinipes*, and after
440 weathering, toxicity either increased or decreased depending on the plastics ([Bejgarn et al.,](#)
441 [2015](#)). Two leaching studies were carried out on the copepod *Daphnia magna* ([Lithner et al.,](#)
442 [2012; Lithner et al., 2009](#)), a common organism used in ecotoxicological studies. [Lithner et al.](#)
443 [\(2009\)](#) prepared their leachates by placing plastic pieces in deionized water to obtain a liquid
444 to solid ratio of 10 L kg⁻¹ which was horizontally shaken at 60 rpm for 24h (16h of fluorescent
445 light and 8h of darkness) at 20°C. Out of the thirty-two plastics tested only nine, including five
446 composed of PVC, showed acute toxicity (immobility after 24 and 48h ; EC₅₀ ranging from 5
447 to 80 g plastic material L⁻¹) to *D. magna* and it has been suggested that the toxicity of PVC was
448 due to the phthalate content ([Lithner et al., 2009](#)). In a second study, [Lithner et al. \(2012\)](#) used
449 PP, PE, PVC, acrylonitrile-butadiene-styrene (ABS) and epoxy resin, and they prepared their
450 leachates by adding 250 g of plastic in 1 L of deionized water shaken at 90 rpm at 50°C for 3

451 days. As previously demonstrated, PVC caused acute toxicity (immobility after 24 and 48h ;
452 EC₅₀ ranging from 2-235 g plastic material L⁻¹) probably in relation to its phthalate leachates,
453 however the acute toxicity observed with the epoxy resin was not attributed to a specific
454 chemical compound (Lithner *et al.*, 2012). A more recent study evaluated the toxicity of virgin
455 and beached pellets on the embryo development of brown mussels (*Perna perna*) (Gandara e
456 Silva *et al.*, 2016). Here, the authors exposed the brown mussel embryo to 2 mL of virgin (PP)
457 or beached (42% PE and 58% unknown composition) pellets in 8 mL of seawater, and toxicity
458 was assessed by determining the percentage of dead or abnormal embryos (Gandara e Silva *et*
459 *al.*, 2016). The leaching experiment led to 23.5% and 100% dead or abnormal embryos for
460 virgin and beached pellets, respectively. It has been suggested that the difference in toxicity
461 was mainly due to the difference in the chemical load of the pellets used (Gandara e Silva *et*
462 *al.*, 2016). Beached pellets were exposed to *in situ* contamination leading to adsorption of
463 pollutants and to additives already found inside the polymeric matrix. These leaching
464 experiments showed that plastic leachates and especially PVC leachates (*i.e.* phthalates) can
465 lead to adverse effects on organisms. However, the toxicity highlighted in these five
466 experiments was not attributed to specific chemical compounds (Bejgarn *et al.*, 2015; Gandara
467 e Silva *et al.*, 2016; Li *et al.*, 2016; Lithner *et al.*, 2012; Lithner *et al.*, 2009). As suggested by
468 Li *et al.* (2016), chemical identification should be undertaken during leaching experiments with
469 a focus on plastic additives in order to identify the compound or its degradation products
470 responsible for the observed toxicity. For instance, a more recent leaching study focused on the
471 effects of nonylphenol on the coral reef fish *Pseudochromis fridmani* by exposing single fish to
472 the leachate of plastic bags made of two PE (PE1 and PE2) from different manufacturers for
473 48h (Hamlin *et al.*, 2015). Nonylphenol leached in the water at higher concentrations for PE2
474 than for PE1; with respectively 41.0 ± 5.5 and 2.5 ± 0.2 $\mu\text{g L}^{-1}$, and 60% and 11% of the fish
475 died after 48 hours of exposure to leachates from PE2 and PE1, respectively. However, Hamlin

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

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

487 **4.4. Relative importance of HOC in comparison with plastic additives: case** 488 **of modelling studies**

489 The high affinity of plastic polymers for HOC has been demonstrated in numerous
490 laboratory experiments (Bakir *et al.*, 2014; Teuten *et al.*, 2007), and an increasing number of
491 studies have focused on the role of MP as a vector for HOC in marine organisms (Besseling *et*
492 *al.*, 2013; Rochman *et al.*, 2013). However, recent studies have suggested that given (i) baseline
493 contamination levels of seawater and marine organisms, and (ii) the low abundance of plastic
494 particles relative to other suspended particles found in oceans (such as organic matter, plankton,
495 detritus etc), exposure to HOC *via* plastic may be negligible compared to natural pathways
496 (Bakir *et al.*, 2016; Beckingham and Ghosh, 2017; Koelmans *et al.*, 2013a, b; Koelmans *et al.*,
497 2016; Paul-Pont *et al.*, 2016). Moreover, Koelmans *et al.* (2016) suggested that MPs ingestion
498 by marine biota does not increase their exposure to HOCs and could have a cleaning effect
499 while concerns have arisen regarding risk due to plastic additives.

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

507 **5. Conclusion**

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

524 **Acknowledgments**

525 Ludovic Hermabessiere is grateful to the Hauts-de-France Region and ANSES (French Agency
526 for Food, Environmental and Occupational Health & Safety) for the financial support of his
527 PhD. This work was funded by ANSES through a “*Convention de Recherche et*
528 *Développement*” (2015-CRD-11). This paper was also supported by the CESA of the French
529 National Research Agency (ANR) (ANR-15-CE34-0006-02), as part of the NANOPLASTICS
530 project. Authors also thank James Marchant for his helpful revision of English and his
531 comments on the manuscript.

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