Residual additives in marine microplastics and their risk assessment – A critical review

Bibiana Jara, Felipe Tucca, Benjamín Srain, Laurence Méjanelle, Mario Aranda, Camila Fernández, Silvio Pantoja-Gutiérrez

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Review

Residual additives in marine microplastics and their risk assessment – A critical review

Patrik Fauser [a,*], Katrin Vorkamp [a], Jakob Strand [b]

[a] Aarhus University, Department of Environmental Science, Frederiksborgvej 399, 4000 Roskilde, Denmark
[b] Aarhus University, Department of Ecoscience, Frederiksborgvej 399, 4000 Roskilde, Denmark

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ABSTRACT

This review summarizes the current state of knowledge regarding the risk assessment of plastic-associated residual additives, i.e. residual monomers, degradation products and additives, in the marine environment, also considering effects of weathering and bioavailability. Experimental studies have found a number of organic and metal additive compounds in leachates from plastics, and the analysis of weathered plastic particles, such as polyethylene, polypropylene and polystyrene particles sampled on beaches and shorelines, has identified residual additives, such as flame retardants, plasticizers, UV stabilizers and antioxidants. While the transfer of e.g. PBDEs to organisms upon ingestion has been demonstrated, studies on uptake and bioaccumulation of plastic-associated chemicals are inconclusive. Studies on hazard and risk assessments are few, and focus on monomers and/or a limited number of high concentration additives, such as phthalates and flame retardants. The risk assessment results vary between low, moderate and high risks of specific additives, and are not necessarily consistent for the same compound. Given the large number of chemicals potentially introduced into the marine environment with plastic particles and the challenges associated with the correct quantification of exposure concentrations and toxicity thresholds, the question arises whether new risk assessment concepts may be needed.

1. Introduction

Plastics in the marine environment can contain one or more chemicals that are either deliberately added (additives), formed during manufacture of the plastic, or absorbed from the ambient water, or all of the above (Koelmans et al., 2021). Chemical additives are added to plastics to modify their properties for a desired material performance, and include plasticizers, flame retardants, (photo-)stabilizers, antioxidants, and pigments, among others (Hansen et al., 2013). The literature often addresses the exposure of organisms to plastic-associated chemicals as the microplastic (MP) vector effect, highlighting the carrier function of the MP particle (e.g. Ziccardi et al., 2016; Caruso, 2019; Koelmans et al., 2021). Besides additives, plastic particles can contain residual monomers and unintentional by-products of reactions during manufacture (Lewandowski et al., 2005). Many of these chemicals have been recognized as hazardous, potentially affecting the health of humans and wildlife through e.g. developmental and reproductive toxicity, and, for residual monomers in particular, carcinogenicity and mutagenicity (Lithner et al., 2011; EC, 2012; Oehlmann et al., 2009; Meeker et al., 2009; Gallo et al., 2018; Campanale et al., 2020). Some plastics contain chemicals that are not intentionally added or formed during manufacture, but are inherent in the used materials or formed during use, such as cellulose acetate in cigarette butts that contain heavy metals from tobacco (Pelit et al., 2013) and organic compounds from the burning of tobacco (Slaughter et al., 2011). We summarize all these chemicals under the term “residual additives”.

This review distinguishes between applied plastic, meaning plastic that is in use in materials or products, and weathered plastic, meaning plastic fragments or particles that are released to the environment and undergo degradation. Studies on the abundance of residual additives in weathered plastic particles are generally limited (Fred-Ahmadu et al., 2020). Marine organisms can be exposed to residual additives via two pathways: i) contact and intake of plastic particles from the environment, in different states of weathering (direct exposure), ii) leaching of plastic additives to the environment, and subsequent contact with the organism (indirect exposure). Plastic additives have been shown to be quantifiable in MP found in sediments and marine waters, and because these additives are not ubiquitous in the marine environment, plastic particles could be a primary source, at least at a local scale (Kwon et al., 2017; Hermabessiere et al., 2017). This review does not consider the

* Corresponding author.
E-mail address: paf@envs.au.dk (P. Fauser).

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sorption of chemicals from the surrounding marine environment, e.g. sorption of hydrophobic organic chemicals (HOCs), including persistent organic pollutants (POPs) (Ziccardi et al., 2016; Caruso, 2019). However, the discussion of leaching and bioavailability includes findings from studies on sorption and desorption processes of HOCs.

The existing framework for environmental risk assessment of chemicals used in regulatory contexts, where exposure levels are combined with hazards, e.g. as outlined in the Guidelines of the European Chemicals Agency (ECHA) (EC, 2003), is yet to be applied to chemicals in marine plastic particles, including MP (Koelmans et al., 2017). Koelmans et al. (2017) further argue that the risk assessment of plastic debris should be divided into a risk component from the plastic particle itself and a separate risk component of the associated chemicals.

In order to quantify exposure to residual additives in plastics, including its variation in space and time, and to assess exposure levels relative to effect endpoints, it is necessary to know the type, size and number of plastic particles and associated chemicals per unit volume of water and to estimate uptake by and bioavailability to marine organisms. In order to assess the indirect exposure, leaching and exposure via environmental media needs to be estimated. However, little is known about the concentration of a certain residual additive in a plastic product, its environmental behaviour and the actual exposure of marine organisms. An additional element of complexity is introduced by varying degrees of physical, chemical or biological weathering of the plastic product with potential implications for its leaching behaviour and/or bioavailability (GESAMP, 2020; Besseling et al., 2019).

A chemical risk assessment procedure is comprised of the steps in Fig. 1. Each step is associated with an uncertainty that is carried along in subsequent steps and is reflected in the final risk estimate. The topics that will be considered in this review are marked in red. MP as vectors for sorption of chemicals from the surrounding environment and the physical effects of plastic particles themselves are not considered. For this we refer to Besseling et al. (2019), Beiras and Schönemann (2020), Everaert et al. (2018), Horodytska et al. (2020), Adam et al. (2019) and Burns and Boxall (2018).

2. Residual plastic additives

2.1. Residual additives and their amounts in applied plastic material

Several thousand different additives are used in the polymer industry. Polymerisation additives become part of the polymer (e.g. cross linking agents, curing agents, inhibitors, initiators), or are necessary to preserve the stability of the polymer. Other additives are combined with a polymer to form a mixture with the particular properties needed for the formation of articles. These latter "inherent" additives are manifold and include stabilizers, plasticizers, flame retardants, antioxidants and photostabilizers, among others (EC, 2012). Unlike plasticizers and flame retardants, which have been relatively well investigated, antioxidants and photostabilizers have rarely been studied with regard to their environmental risk, even though many of them are highly hydrophobic and not readily biodegradable. Some of these chemicals may be found at relatively high concentrations (10–15 wt-%) in the plastic material and be potentially bioavailable after ingestion by organisms (Andrady, 2017).

Polymerisation reactions are rarely 100% complete and therefore unreacted monomers, oligomers and in some cases reaction by-products are present after manufacture of some types of plastic. The proportion of unreacted monomers or by-products can vary greatly depending on type of polymer, polymerisation technique and techniques for reducing the levels of these constituents, as well as the design and application of the plastic material. In a review by Araujo et al. (2002), the proportion of unreacted monomers or by-products varied from no or very low levels (1 ppm; i.e. 0. 0001 wt-%) to up to 40,000 ppm (i.e. 4 wt-%). Examples of unreacted monomers are vinylchloride and styrene. The latter has been found in concentrations up to 0.6 wt-% in polystyrene granules (Peng et al., 2017; Garrigos et al., 2004).

Formation of hazardous degradation products can occur during the manufacture process, for example methylene dianiline (MDA) formed from methylene diphenyl diisocyanate (MDI) used in the production of polyurethane foam (PUR) (Lewandowski et al., 2005). Furthermore, isocyanates can be formed from work processes where PUR products are heated to temperatures above 150 °C, e.g. when welding or soldering surfaces with PUR coatings. Tian et al. (2021) found that a chemical derived from the antioxidant 6PPD (Table 1b) in tire tread leachate was the primary causal toxicant during urban runoff mortality syndrome events across the U.S. West Coast. Considering the potential release to the marine environment only MDA and 6PPD are included here in a risk assessment context, due to limited data material.

Identities and used amounts of potential residual compounds in specific polymers are compiled in Table 1a for monomers and Table 1b for potential residual additives and degradation products, including some observations on their migration behaviour. A number of compounds with unknown amounts are listed in e.g. ECHA (2021), but only additives with known amounts are included here. Compounds with restrictions or bans, e.g. the flame retardants decabrominated diphenyl

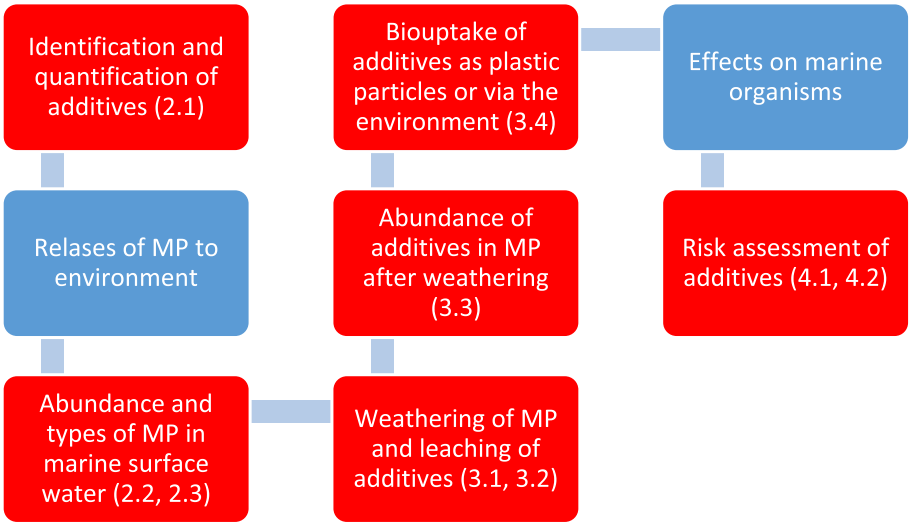

Fig. 1. Conceptual procedure for risk assessment of additives in microplastic (MP) particles in the marine environment. Red blocks are included in this review. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Table 1a
Potential residual unreacted monomers, with typical use amounts in wt-% (Lithner et al., 2011).

Monomer (typical amount in polymer/ plastic, wt-%)	Polymer type (see Table SI-1 for full polymer names)
Acrylamide (8%)	PAN co-monomer
Ethylene oxide (4–7%)	PUR, POM *co*-polymer
Propylene oxide (31–58%)	PUR
1,3-Butadiene (8–20%)	ABS, HIPS
4,4′-Methylenedianiline (MDA) (18%)	Epoxy
Acrylonitrile (22–100%)	PAN and its co-monomers, SAN, ABS
Epichlorohydrin (30–37%)	Epoxy
Vinylchloride (50–100%)	PVC
m-Phenylenediamine (35%)	MPD-I
Toluene-diisocyanate (TDI) (29%)	PUR
p-Phenylenediamine (34%)	PPD-T
4,4′-Methylenediphenyl diisocyanate (MDI) (24–52%)	PUR, TPU
Phthalic anhydride (31%)	UP
Maleic anhydride (21%)	UP
Formaldehyde (18–100%)	POM, PF, MF, UF
Phenol (61–72%)	PF
Bisphenol A (43–70%)	Epoxy, PC
1,4-Dichlorobenzene (65%)	PPS
Phosgene (30%)	PC
Methyl methacrylate (30–100%)	PMMA, UP
Hexamethylenetetramine (10%)	PF one type
2,6-Xylenol (100%)	PPO
Sodium sulphide (35%)	PPS
Acrylic acid (100%)	PAA
Pentane (7%)	EPS
Hexamethylenediamine (37–44%)	Nylon 6.6
Vinylidene chloride (40%)	Modacrylic
Trioxymethylene (94–96%)	POM co-polymers
ε-Caprolactam (100%)	Nylon 6
Styrene (30–100%)	PS, EPS, HIPS, SAN, ABS, UP
Cyclopentane (4%)	PUR
Ethylene (80–100%)	HDPE, LDPE, LLDPE, EVA
Adipic acid (35–56%)	Nylon 6.6, TPU
4,4′-Diamino diphenyl sulfone (DDS) (22%)	Epoxy
Ethylene glycol (35–39%)	TPU, PET
1-Butene (10%)	LLDPE
1,3-Dioxolane (6%)	POM co-polymer
Propylene (100%)	PP
Vinyl acetate (8–100%)	PAN co-monomer, EVA, PVAc
11-Aminoundecanoic acid (100%)	Nylon 11
Melamine (41%)	MF
Dicyandiamide (3%)	Epoxy
Dimethyl terephthalate (61–62%)	PET, PBT
Propylene glycol (18%)	UP
Sorbitol	PUR
Terephthalic acid (63%)	PET
Urea (50%)	UF
Vinylidene fluoride (100%)	PVDF
Lactic acid (100%)	PLA
Diphenyl carbonate (50%)	PC
1-Hexene (10%)	LLDPE
1-Octene (10%)	LLDPE
Lauryl lactam (100%)	Nylon 12
1,4-Butanediol (6–16%)	TPU, PBT
HCF-134a	PUR
Isophthaloyl chloride (65%)	MPD-I
Sebacic acid (63%)	Nylon 6.10
Terephthaloyl chloride (66%)	PPD-T
Tetrafluoroethylene (100%)	PTFE

ether (Deca-BDE) and hexabromocyclododecane (HBCDD), are omitted in the tables of ECHA (2021), but included here as they may still occur in plastic materials of long lifetimes as well as recycled plastic. Additional information can be found in Flick (2004), Harper and Petrie (2003) and Zweifel et al. (2009), the additive database SpecialChem (2020), manufacturers' web sites, e.g. BASF (2020), and suppliers of analytical reference standards, e.g. AccuStandard (2018). However, additive amounts in polymers are not given in these references, and the information is therefore not included in Tables 1a and 1b.

2.2. Marine microplastic levels

Quantified levels of plastic particles in the marine environment are necessary in determining the exposure of marine organisms to additives. The concentration and distribution of plastic in the water surface and water column compartments are highly variable due to seasonal changes in river outputs, ocean currents, mechanisms of degradation and fragmentation, changes in litter characteristics and movement to and from other compartments (GESAMP, 2019). In addition, variations can be related to sampling and detection methods. As an example Song et al. (2014) reported that different sampling devices produce differences up to four orders of magnitude in floating MP concentrations. Goldstein et al. (2012) reported that 98.5% of the particles caught in a 333 μm mesh would also have been caught in a 505 μm mesh, and there were no significant differences in particle size spectra using one size or another. On the other hand, Magnusson and Norén (2011) found 2500 and 500 times higher concentration of fibers and particles, respectively, when filtering with 10 μm filters compared to 300 μm. A common challenge in any sampling effort is therefore for the collected information to be as representative as possible. Lacks of standardized methods impede comparisons although several standards and recommended protocols have recently emerged for sampling MPs (Karlsson et al., 2020). Surface water samples of floating MP are commonly collected using nets such as manta trawls, and water column samples of MP can be collected with pumps (GESAMP, 2019).

Karlsson et al. (2020) performed a study comparing different sampling techniques, i.e. manta trawl and in situ filtering pumps, during realistic, but controlled, field tests. Performing identical MP analyses of all replicates allowed an assessment of the differences between the methods with respect to precision, concentrations, and composition. The study concluded that some of the challenges/suggestions regarding sampling of MP were that smaller particle fractions, i.e. below 100 μm, must be quantified with supplementary chemical techniques. Furthermore, caution is recommended when using an automated library search, e.g. in plastic reference libraries, due to uncertainties in analysing polymer spectra. In general, high sample volumes and replicates are necessary to provide quantitative and compositional data (Karlsson et al., 2020). Further, Tamminga et al. (2019) argue that sampling with neuston nets, such as manta trawls, and pump sampling are complementary techniques, as they cover different parts of the overall MP pollution in terms of particles sizes and shapes. In addition to the sampling method the filter size is a determining factor in the MP abundance. Historically, filter/sieve sizes span from typically 10 μm to approx. 500 μm.

Table 2 shows a compilation of oceanic MP data, only including samples collected with neuston nets, preferably manta trawl, for surface water, and filter pumps for water column sampling. Furthermore, only studies published after 2010 are considered. Data for fibers are also included, as a number of studies report that fibers constitute most of the MP found in marine waters (Lusher et al., 2015; Cole et al., 2014; Beer et al., 2018).

Beiras and Schönemann (2020) performed a comprehensive review on plastic particle concentrations in surface and sub-surface waters of the global ocean, most of which are included in Table 2. To allow data comparability, surface sampling techniques were limited to manta trawl or neuston nets. Mesh sizes ranged from 200 to 505 μm, and reported mean concentrations of plastics in surface waters ranged from 0.00012–7.250 pieces/m^3, and from 0.01–34.09 μg/L. Similar amounts, in mass units, were found in sub-surface waters. Either recorded in numbers or mass units, concentrations frequently showed a log-normal distribution. The variance in plastic concentrations was large and similar in all datasets, with an average coefficient of variation of 201% (Beiras and Schönemann, 2020). In order to identify local characteristics that are not uniform across the entire ocean the location within a specific ocean rather than the ocean itself is more useful to predict plastic concentrations (Law et al., 2010). In coastal regions, plastic concentrations

Table 1b
Potential residual additives, including degradation products, with typical amounts of use in wt-%, and their use in polymers. ECHA (2021) information was retrieved in September 2021. Some observations on their migration behaviour are also included.

Function of additive	Additive (typical amount in polymer/plastic, wt-%)	Polymer type (see Table SI-1 for full polymer names)	Reference
Light stabilizers	Octabenzone (0.2–5%)	Polyolefins; PUR; PVC (soft); ABS; PVC (rigid); PMMA; PC; (E)PS	ECHA (2021)
	2-(2H-benzotriazol-2-yl)-p-cresol (0.0015–0.5%)	Polyolefins; PVC (soft); ABS; PVC (rigid); PMMA; (E)PS	ECHA (2021)
	2-(2H-benzotriazol-2-yl)-4-(1,1,3,3-tetramethylbutyl)phenol (0.3%)	PC	ECHA (2021)
	Bumetrizole (0.3–1%)	Polyolefins; PUR; PVC (soft); ABS; PVC (rigid); PC; (E)PS	ECHA (2021)
	N-(2-ethoxyphenyl)-N′-(2-ethylphenyl)oxamide (0.7%)	PVC (soft); ABS; PVC (rigid); PA; PC; (E)PS	ECHA (2021)
	Bis(1,2,2,6,6-pentamethyl-4-piperidyl) [[3,5-bis(1,1-dimethylethyl)-4-hydroxyphenyl]methyl]butylmalonate (1%)	PMMA	ECHA (2021)
	3,5-Di-tert-butyl-4-hydroxybenzoic acid, hexadecyl ester (0.2%)	Polyolefins	ECHA (2021)
	2-(2H-benzotriazol-2-yl)-4,6-bis(1-methyl-1-phenylethyl)phenol (0.2–5%)	Polyolefins; PUR; PVC (soft); ABS; PVC (rigid); PET; PMMA; PA; PC; (E)PS	ECHA (2021)
	2,2′-Methylenebis(6-(2H-benzotriazol-2-yl)-4-(1,1,3,3-tetramethylbutyl)phenol) (0.2–6%)	PMMA; PC	ECHA (2021)
	A mixture of: bis(2,2,6,6-tetramethyl-1-octyloxypiperidin-4-yl)-1,10-decanedioate; 1,8-bis[(2,2,6,6-tetramethyl-4-((2,2,6,6-tetramethyl-1-octyloxypiperidin-4-yl)-decan-1,10-dioyl)piperidin-1-yl)oxy]octane (0.2–0.5%)	PVC (soft); PVC (rigid); PMMA	ECHA (2021)
	2-(4,6-Diphenyl-1,3,5-triazin-2-yl)-5-((hexyl)oxy)phenol (6%)	PVC (soft); PVC (rigid); PET; PMMA; PC	ECHA (2021)
	N,N′-1,6-hexanediylbis(N-(2,2,6,6-tetramethyl-piperidin-4-yl) formamide) (0.5–0.8%)	Polyolefins; ABS; PMMA; PA; (E)PS	ECHA (2021)
	Reaction mass of Bis(1,2,2,6,6-pentamethyl-4-piperidyl) sebacate and Methyl 1,2,2,6,6-pentamethyl-4-piperidyl sebacate (0.2–0.5%)	PUR; ABS; PMMA; (E)PS	ECHA (2021)
	Antioxidants and UV-stabilizers: Bisphenol A (BPA); Cadmium and Lead compounds; Nonylphenol compounds; Octylphenol; 1,3,5-Tris (oxiran-2-ylmethyl)-1,3,5-triazinane-2,4,6-trione (TGIC)/1,3,5-tris[(2S and 2R)- 2,3-epoxypropyl]-1,3,5- triazine-2,4,6-(1H,3H,5H)- trione (β-TGIC), Butylated hydroxytoluene (BHT), 2- and 3-t-butyl-4 hydroxyanisole (BHA), tetrakismethylene-(3,5-di-t-butyl-4- hydroxyhydrocinnamate) methane (Irganox 1010), and bisphenolics such as Cyanox 2246 and 425, Tris-nonyl-phenyl phosphate (TNPP), tris (2, 4-di-tert-butylphenyl) phosphite, (Irgafos 168) (0.05–3%)	The amount depends on the chemical structure of the additive and of the plastic polymer. Phenolic antioxidants are used in low amounts and phosphites in high. Lowest amounts in polyolefins (LLDPE, HDPE), higher in HIPS and ABS	Hahladakis et al. (2018) recreated from Hansen et al. (2013)
Heat stabilizers	Dibutyltin dilaurate (3%)	Polyolefins; PVC (soft); PVC (rigid)	ECHA (2021)
	Triphenyl phosphite (3%), Zinc dilaurate (2%), Pentalead tetraoxide sulphate (2%), Tetralead trioxide sulphate (2%), Dioxobis(stearato) trilead (2%), 2,2-dioctyl-1,3,2-oxathiastannolan-5-one (2%), Methyl (Z,Z)-8,8-dibutyl-3,6,10-trioxo-2,7,9-trioxa-8-stannatrideca-4,11-dien-13-oate (2%), 2-ethylhexyl 10-ethyl-4,4-dioctyl-7-oxo-8-oxa-3,5-dithia-4-stannatetradecanoate (2%), 2-ethylhexyl diphenyl phosphite (2%), Triisodecyl phosphite (2%), Diisodecyl phenyl phosphite (3%), Isodecyl diphenyl phosphite (3%), 2-ethylhexyl 10-ethyl-4-[[2-[(2-ethylhexyl)oxy]-2-oxoethyl]thio]-4-octyl-7-oxo-8-oxa-3,5-dithia-4-stannatetradecanoate (2%), Zinc bis[12-hydroxyoctadecanoate] (2%), 2-ethylhexyl 10-ethyl-4-[[2-[(2-ethylhexyl)oxy]-2-oxoethyl]thio]-4-methyl-7-oxo-8-oxa-3,5-dithia-4-stannatetradecanoate (2%), 2-ethylhexyl 10-ethyl-4,4-dimethyl-7-oxo-8-oxa-3,5-dithia-4-stannatetradecanoate (4%), Sulfurous acid, lead salt, dibasic (2%), Ethyl 9,9-dioctyl-4,7,11-trioxo-3,8,10-trioxa-9-stannatetradeca-5,12-dien-14-oate (2%), Triisotridecyl phosphite (2%), Fatty acids, C16–18, lead salts (2%), Fatty acids, C16–18, zinc salts (2%)	PVC (soft); PVC (rigid)	ECHA (2021)
	N,N′,N″,N‴-tetrakis(4,6-bis(butyl-(*N*-methyl-2,2,6,6-tetramethylpiperidin-4-yl)amino)triazin-2-yl)-4,7-diazadecane-1,10-diamine (1.5%)	Polyolefins; PUR	ECHA (2021)
	Cadmium and lead compounds; Nonylphenol (barium and calcium salts) (0.5–3%)	Mainly used in PVC. Based on Pb, Sn, Ba, Cd and Zn compounds. Pb is the most efficient and it is used in lower amounts	Hahladakis et al. (2018) recreated from Hansen et al. (2013)
Other stabilizers	Copper iodide (0.5%)	PA	ECHA (2021)
	Magnesium carbonate (1%)	Polyolefins; PA	ECHA (2021)
	Fatty acids, C14–18 and C16–18-unsatd. (1%)	Polyolefins	ECHA (2021)
	N,N′-ethylenedi(stearamide) (0.3–0.5%)	Polyolefins; ABS; (E)PS	ECHA (2021)

(*continued on next page*)

Table 1b (*continued*)

Function of additive	Additive (typical amount in polymer/plastic, wt-%)	Polymer type (see Table SI-1 for full polymer names)	Reference
	Bis(2,2,6,6-tetramethyl-4-piperidyl) sebacate (0.2–1%)	Polyolefins; ABS; PMMA; PA; PC; (E)PS	ECHA (2021)
	[Carbonato(2-)]hexadecahydroxybis (aluminum)hexamagnesium (0.5%)	Polyolefins; PUR; ABS; PA; PC	ECHA (2021)
	Calcium oxide (0.1%)	Polyolefins; PUR; PVC (rigid)	ECHA (2021)
Antioxidants	6,6′-Di-tert-butyl-4,4′-thiodi-m-cresol (0.001%)	Polyolefins	ECHA (2021)
	6,6′-Di-tert-butyl-2,2′-methylenedi-p-cresol (0.5%)	Polyolefins; ABS	ECHA (2021)
	Di-stearyl-pentaerythrityl-di-phosphite (0.2%)	Polyolefins	ECHA (2021)
	2,4-Bis(octylthiomethyl)-6-methylphenol (0.015–0.2%)	Polyolefins; (E)PS	ECHA (2021)
	1,3,5-Tris(3,5-di-tert-butyl-4-hydroxybenzyl)-1,3,5-triazine-2,4,6 (1H,3H,5H)-trione (0.4–0.8%)	Polyolefins; ABS	ECHA (2021)
	Dioctadecyl 3,3′-thiodipropionate (0.25–3%)	Polyolefins; ABS; PET; PA	ECHA (2021)
	3,9-Bis(2,4-di-tert-butylphenoxy)-2,4,8,10-tetraoxa-3,9-diphosphaspiro[5.5]undecane (0.0015–1%)	Polyolefins; ABS; PET; PA; PC; (E)PS	ECHA (2021)
	3,3′,3″,5,5′,5″-Hexatert-butyl-α,α′,α″-(mesitylene-2,4,6-triyl)tri-p-cresol (0.5–2.5%)	Polyolefins; PA	ECHA (2021)
	Reaction products of phosphorous trichloride, with 1,1′-biphenyl and 2,4-bis(1,1- dimethylethyl)phenol (0.1–0.25%)	Polyolefins; PET; PMMA; PA; PC	ECHA (2021)
	4,4′,4″-(1-Methylpropanyl-3-ylidene)tris[6-tert-butyl-m-cresol] (0.5–1%)	Polyolefins; PVC (soft); ABS; PVC (rigid); (E)PS	ECHA (2021)
	Octadecyl 3-(3,5-di-tert-butyl-4-hydroxyphenyl)propionate (0.002–4%)	Polyolefins; PVC (soft); ABS; PVC (rigid); PMMA; PC; (E)PS	ECHA (2021)
	Tris(2,4-ditert-butylphenyl) phosphite (0.004–5%)	Polyolefins; PUR; ABS; PET; PMMA; PA; PC; (E)PS	ECHA (2021)
	2′,3-Bis[[3-[3,5-di-tert-butyl-4-hydroxyphenyl]propionyl]] propionohydrazide (0.002–3%)	Polyolefins; PUR; PVC (soft); PA; PC	ECHA (2021)
	Pentaerythritol tetrakis(3-(3,5-di-tert-butyl-4-hydroxyphenyl) propionate) (0.002–0.5%)	Polyolefins; PUR; PVC (soft); PVC (rigid); PET; PA; PC; (E)PS	ECHA (2021)
	Di-octadecyl-disulphide (1%)	Polyolefin	ECHA (2021)
	Ethylenebis(oxyethylene) bis[3-(5-tert-butyl-4-hydroxy-m-tolyl) propionate] (0.005–3%) (1%)	PUR; PVC (soft); ABS; PVC (rigid); PMMA; PA; (E)PS	ECHA (2021)
	6,6′-Di-tert-butyl-4,4′-butylidenedi-m-cresol (0.5%)	PVC (rigid); PA	ECHA (2021)
	C18H24N2 anti-ozonant “6PPD” [N-(1,3-dimethylbutyl)-*N*′-phenyl-phenylenediamine] (0.4% to 2%)	Passenger and commercial vehicle tire formulations	Tian et al. (2021)
Nucleating agents	Sodium benzoate (0.2%), 2,2′-methylene bis-(4,6-di-tert.butylphenyl) sodium phosphate (0.2%)	Polyolefin	ECHA (2021)
Pigments	3-Hydroxy-N-(o-tolyl)-4-[(2,4,5-trichlorophenyl)azo]naphthalene-2-carboxamide (2%)	(E)PS	ECHA (2021)
	Tricobalt tetraoxide (1%), Aluminum oxide (0.25%)	PA	ECHA (2021)
	Barium 4-[(5-chloro-4-methyl-2-sulphonatophenyl)azo]-3-hydroxy-2-naphthoate (2%)	Polyolefins	ECHA (2021)
	Copper (tetrachloro-29H,31H-phthalocyaninato(2-)-N29,N30,N31, N32) (0.05%)	Polyolefins	ECHA (2021)
	N-(2,3-dihydro-2-oxo-1H-benzimidazol-5-yl)-3-oxo-2-[[2-(trifluoromethyl)phenyl]azo]butyramide (2%)	Polyolefins; ABS; (E)PS	ECHA (2021)
	3,6-Bis-biphenyl-4-yl-2,5-dihydro-pyrrolo[3,4-c]-pyrrole-1,4-dione (2%)	Polyolefins; ABS; PET; (E)PS	ECHA (2021)
	2,2′-[(3,3′-Dichloro[1,1′-biphenyl]-4,4′-diyl)bis(azo)]bis[N-(2-methoxyphenyl)-3-oxobutyramide] (2%)	Polyolefins; PVC (soft)	ECHA (2021)
	Calcium bis[4-[[1-[[(2-methylphenyl)amino]carbonyl]-2-oxopropyl] azo]-3-nitrobenzenesulphonate] (2%)	Polyolefins; PVC (soft); ABS; (E)PS	ECHA (2021)
	2-[[1-[[(2,3-Dihydro-2-oxo-1H-benzimidazol-5-yl)amino]carbonyl]-2-oxopropyl]azo]benzoic acid (2%), Calcium bis[4-[[1-[[(2-chlorophenyl)amino]carbonyl]-2-oxopropyl]azo]-3-nitrobenzenesulphonate] (2%)	Polyolefins; PVC (soft); ABS; PVC (rigid); (E)PS	ECHA (2021)
	2,2′-[Ethylenebis(oxyphenyl-2,1-eneazo)]bis[N-(2,3-dihydro-2-oxo-1H-benzimidazol-5-yl)-3-oxobutyramide] (2%), 3,6-Bis(4-*tert*-butylphenyl)-2,5-dihydropyrrolo[3,4-*c*]pyrrole-1,4-dione (2%)	Polyolefins; PVC (soft); ABS; PVC (rigid); PC; (E)PS	ECHA (2021)

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Table 1b (*continued*)

Function of additive	Additive (typical amount in polymer/plastic, wt-%)	Polymer type (see Table SI-1 for full polymer names)	Reference
	Calcium 4-[(5-chloro-4-methyl-2-sulphonatophenyl)azo]-3-hydroxy-2-naphthoate (2%), N,N′-(2-chloro-1,4-phenylene)bis[4-[(4-chloro-2-nitrophenyl)azo]-3-hydroxynaphthalene-2-carboxamide] (2%)	Polyolefins; PVC (soft); ABS; PVC (rigid); PET; PA; PC; (E)PS	ECHA (2021)
	2-[(2-Methoxy-4-nitrophenyl)azo]-N-(2-methoxyphenyl)-3-oxobutyramide (2%)	Polyolefins; PVC (soft); ABS; PVC (rigid); PET; PC; (E)PS	ECHA (2021)
	Copper, [29H,31H-phthalocyaninato(2-)-N29,N30,N31,N32]-, brominated chlorinated (2%)	Polyolefins; PVC (soft); ABS; PVC (rigid); PET; PMMA; PA; PC; (E)PS	ECHA (2021)
	Copper, [29H,31H-phthalocyaninato(2-)-N29,N30,N31,N32]-, chlorinated (2%)	Polyolefins; PVC (soft); PVC (rigid); (E)PS	ECHA (2021)
	Zinc sulphide (2–10%)	Polyolefins; PUR; ABS; PET; PMMA; PA; PC; (E)PS	ECHA (2021)
	Perylene-3,4:9,10-tetracarboxydiimide (2%)	Polyolefins; PUR; ABS; PVC (rigid); PET; PMMA; PA; PC; (E)PS	ECHA (2021)
	Reaction mass of melamine and Nickel, 5,5′-azobis-2,4,6(1H,3H,5H)-pyrimidinetrione complexes (2%)	Polyolefins; PUR; PVC (soft); ABS; PVC (rigid); PET; PA; PC	ECHA (2021)
	4,4′-[(3,3′-Dichloro[1,1′-biphenyl]-4,4′-diyl)bis(azo)]bis[2,4-dihydro-5-methyl-2-phenyl-3H-pyrazol-3-one] (2%), 2,2′-[(3,3′-dichloro[1,1′-biphenyl]-4,4′-diyl)bis(azo)]bis[N-(2,4-dimethylphenyl)-3-oxobutyramide] (2%), Copper chlorophthalocyanine (2%)	Polyolefins; PUR; PVC (soft); ABS; PVC (rigid); PET; PA; PC; (E)PS	ECHA (2021)
	N,N′-(2,5-dichloro-1,4-phenylene)bis[4-[(2,5-dichlorophenyl)azo]-3-hydroxynaphthalene-2-carboxamide] (2%)	Polyolefins; PUR; PVC (soft); ABS; PVC (rigid); PET; PC; (E)PS	ECHA (2021)
	6,15-Dihydroanthrazine-5,9,14,18-tetrone (2%), 29H,31H-phthalocyaninato(2-)-N29,N30,N31,N32 copper (0.5–2%), Calcium carbonate (5%), 29H,31H-Phthalocyanine (2%), 5,12-dihydro-2,9-dimethylquino[2,3-*b*]acridine-7,14-dione (2%), 5,12-dihydroquino[2,3-*b*]acridine-7,14-dione (2%), Chromium (III) oxide (1%), Diiron trioxide (1%), Triiron tetraoxide (1%), Titanium dioxide (rutil) (5%), Polychloro copper phthalocyanine (1%), Carbon black (2.5–40%), Lead sulfochromate yellow (1%), 2-[(4-methyl-2-nitrophenyl)azo]-3-oxo-*N*-phenylbutyramide (2%), 2,9-bis[4-(phenylazo)phenyl]anthra[2,1,9-def:6,5,10-d′e′f′]diisoquinoline-1,3,8,10(2H,9H)-tetrone (2%), 2,9-dichloro-5,12-dihydroquino[2,3-*b*]acridine-7,14-dione (2%), *N*,*N*′-phenylene-1,4-bis[4-[(2,5-dichlorophenyl)azo]-3-hydroxynaphthalene-2-carboxamide] (2%), 4,4′-diamino[1,1′-bianthracene]-9,9′,10,10′-tetraone (1–2%), 2,9-bis(3,5-dimethylphenyl)anthra[2,1,9-def:6,5,10-d′e′f′]diisoquinoline-1,3,8,10(2H,9H)-tetrone (2%), N-(4-chloro-2,5-dimethoxyphenyl)-3-hydroxy-4-[[2-methoxy-5-[(phenylamino)carbonyl]phenyl]azo]naphthalene-2-carboxamide (2%), N,N′-(2-chloro-1,4-phenylene)bis[4-[(2,5-dichlorophenyl)azo]-3-hydroxynaphthalene-2-carboxamide] (2%), 3,3′-[(2,5-dimethyl-*p*-phenylene)bis[imino(1-acetyl-2-oxoethylene)azo]]bis[4-chloro-N-(5-chloro-o-tolyl)benzamide] (2%), Calcium 3-hydroxy-4-[(4-methyl-2-sulphonatophenyl)azo]-2-naphthoate (2%), 2,9-dimethylanthra[2,1,9-def:6,5,10-d′e′f′]diisoquinoline-1,3,8,10(2H,9H)-tetrone (2%), 3,3′-[(2-chloro-5-methyl-*p*-phenylene)bis[imino(1-acetyl-2-oxoethylene)azo]]bis[4-chloro-N-(3-chloro-o-tolyl)benzamide] (2%), 4-[(2,5-dichlorophenyl)azo]-3-hydroxy-*N*-phenylnaphthalene-2-carboxamide (2%), 2,2′-[(3,3′-dichloro[1,1′-biphenyl]-4,4′-diyl)bis(azo)]bis[3-oxo-*N*-phenylbutyramide] (2%), Barium sulfate (50%), Antimony nickel titantium oxide yellow (1%), Cadmium zinc sulfide yellow (5%), Ammonium manganese(3+) diphosphate (5%), Iron manganese trioxide (0.5%), Pentacalcium hydroxide tris(orthophosphate) (5%), 2-[(4-chloro-2-nitrophenyl)azo]-N-(2,3-dihydro-2-oxo-1H-benzimidazol-5-yl)-3-oxobutyramide (2%), Lead chromate molybdate sulfate red (5%), Chromium iron oxide (5%), Ultramarine Violet (0.5%), Titanium dioxide (5–20%), Bismuth vanadium tetraoxide (0.5%), [1,3,8,16,18,24-hexabromo-2,4,9,10,11,15,17,22,23,25-decachloro-29H,31H-phthalocyaninato(2-)-N29,N30,N31,N32]copper (2%), 4,4′-[(3,3′-dichloro[1,1′-biphenyl]-4,4′-diyl)bis(azo)]bis[2,4-dihydro-5-methyl-2-(p-tolyl)-3H-pyrazol-3-one] (2%), Tin dioxide (5%), Ammonium iron(3+) hexakis(cyano-C) ferrate(4-) (0.5%), [N,N,N′,N′,N″,N″-hexaethyl-29H,31H-phthalocyaninetrimethylaminato(2-)-N29,N30,N31,N32]copper (2%), 3,4,5,6-tetrachloro-N-[2-(4,5,6,7-tetrachloro-2,3-dihydro-1,3-dioxo-1H-inden-2-yl)-8-quinolyl]phthalimide (2%), Iron hydroxide oxide yellow (5%), Cadmium sulfoselenide red (5%), Methyl 4-[[(2,5-	Polyolefins; PUR; PVC (soft); ABS; PVC (rigid); PET; PMMA; PA; PC; (E)PS	ECHA (2021)

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Table 1b (*continued*)

Function of additive	Additive (typical amount in polymer/plastic, wt-%)	Polymer type (see Table SI-1 for full polymer names)	Reference
	dichlorophenyl)amino]carbonyl]-2-[[2-hydroxy-3-[[(2-methoxy-phenyl)amino]carbonyl]-1-naphthyl]azo]benzoate (2%), Cobalt zinc aluminate blue spinel (5%), Zinc iron chromite brown spinel (5%), Chrome antimony titanium buff rutile (1%), Copper chromite black spinel (0.5%), Chrome tungsten titanium buff rutile (0.5%), Manganese ferrite black spinel (5%), Iron cobalt chromite black spinel (5%), Cobalt chromite blue green spinel (5%), Zirconium praseodymium yellow zircon (5%), Zinc ferrite brown spinel (5%), Manganese antimony titanium buff rutile (5%), Hematite, chromium green black (5%), Bentonite, acid-leached (5%), Nickel iron chromite black spinel (5%), 5-[(2,3-dihydro-6-methyl-2-oxo-1H-benzimidazol-5-yl)azo]barbituric acid (2%), 3,3′-[(2-chloro-5-methyl-*p*-phenylene)bis[imino(1-acetyl-2-oxoethylene)azo]]bis[4-chloro-N-[2-(4-chlorophenoxy)-5-(trifluoromethyl)phenyl]benzamide] (2%), Copper, [29H,31H-phthalocyaninato(2-)-N29,N30,N31,N32]-, [[3-(1-methylethoxy) propyl]amino]sulfonyl derivs. (2%), Silicic acid, aluminum sodium salt, sulfurized (5%), Silicic acid, zirconium salt, cadmium pigment-encapsulated (5%), Cobalt aluminate blue spinel (5%), 3, 6 - bis (4 - chlorophenyl)- 2, 5 - dihydro - 1, 4 - diketo pyrrolo [3, 4 - c] pyrrole; Pyrrolo[3,4-*c*]pyrrole-1,4-dione, 3,6-bis(4-chlorophenyl)-2,5-dihydro- (2%), Reaction mass of willemite, white and zinc iron chromite brown spinel (5%)		
	Calcium 4,5-dichloro-2-[[4,5-dihydro-3-methyl-5-oxo-1-(3-sulphonatophenyl)-1H-pyrazol-4-yl]azo]benzenesulphonate (2%)	Polyolefins; PUR; PVC (soft); ABS; PVC (rigid); PET; PMMA; PC; (E)PS	ECHA (2021)
	5,5′-(1H-isoindole-1,3(2H)-diylidene)dibarbituric acid (2%)	Polyolefins; PUR; PVC (soft); PVC (rigid)	ECHA (2021)
	Tetramethyl 2,2′-[1,4-phenylenebis[imino(1-acetyl-2-oxoethane-1,2-diyl)azo]]bisterephthalate (2%)	Polyolefins; PUR; PVC (soft); PVC (rigid); PET; PMMA; (E)PS	ECHA (2021)
	Barium bis[2-chloro-5-[(2-hydroxy-1-naphthyl)azo]toluene-4-sulphonate] (2%)	Polyolefins; PVC (soft); PVC (rigid)	ECHA (2021)
	N-(2,3-dihydro-2-oxo-1H-benzimidazol-5-yl)-3-hydroxy-4-[[2-methoxy-5-[(phenylamino)carbonyl]phenyl]azo]naphthalene-2-carboxamide (2%)	PUR; PVC (soft); ABS; PVC (rigid); PET	ECHA (2021)
	2,2′-[(3,3′-Dichloro[1,1′-biphenyl]-4,4′-diyl)bis(azo)]bis[N-(2-methylphenyl)-3-oxobutyramide] (2%)	PUR; PVC (soft); PVC (rigid)	ECHA (2021)
	Strontium 4-[(5-chloro-4-methyl-2-sulphonatophenyl)azo]-3-hydroxy-2-naphthoate (1:1) (2%)	PVC (soft); PVC (rigid)	ECHA (2021)
	Soluble (e.g. azocolorants) (0.25–5%)	They migrate easily and are used in highly transparent plastics. They are expensive, with limited light and heat resistance. Mostly used in PS, PMMA and cellulose plastics to give a bright transparent colour	Hahladakis et al. (2018) recreated from Hansen et al. (2013)
	Organic pigments (insoluble): Cobalt(II) diacetate (0.001–2.5%)	They are insoluble with low migration tendency	Hahladakis et al. (2018) recreated from Hansen et al. (2013)
	Inorganic pigments: Cadmium compounds; Chromium compounds; Lead compounds. Zinc sulphide, zinc oxide, iron oxide, cadmium-manganese based compounds, chromium based compounds, ultramarine and titanium dioxide (0.01–10%)		Hahladakis et al. (2018) recreated from Hansen et al. (2013)
Antistatics	Multi-Walled Carbon Nanotubes (MWCNT), synthetic graphite in tubular shape (10%)	PA	ECHA (2021)
	Stearic acid, monoester with glycerol (1%)	Polyolefins	ECHA (2021)
	Disodium tetraborate, anhydrous (5%)	Polyolefins; PVC (soft)	ECHA (2021)
	Zinc oxide (5%)	Polyolefins; PUR; PVC (soft); ABS; PVC (rigid); PET; PMMA; PA; PC; (E)PS	ECHA (2021)
	Not specified (0.1–1%)	Most types are hydrophilic with the potential to migrate to water	Hahladakis et al. (2018) recreated from Hansen et al. (2013)
Flame retardants	Boehmite (Al(*OH*)O) (5%)	PA	ECHA (2021)
	Calcium phosphinate (1%), Dechlorane plus = 1,2,3,4,7,8,9,10,13,13,14,14-dodecachloro-1,4,4a,5,6,6a,7,10,10a,11,12,12a-dodecahydro-1,4,7,10-dimethanodibenzo[*a*,*e*]cyclooctene (3%), Polyphosphoric acids, ammonium salts (30%)	Polyolefins	ECHA (2021)
	2,2′,6,6′-Tetrabromo-4,4′-isopropylidenediphenol (5–10%), Magnesium hydroxide (4–8%)	Polyolefins; ABS	ECHA (2021)

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Table 1b (*continued*)

Function of additive	Additive (typical amount in polymer/plastic, wt-%)	Polymer type (see Table SI-1 for full polymer names)	Reference
	Hexaboron dizinc undecaoxide (0.3–0.4%)	Polyolefins; PA	ECHA (2021)
	Diantimony trioxide (8%)	Polyolefins; PVC (soft); ABS; PA	ECHA (2021)
	Aluminum hydroxide (0.25–50%)	Polyolefins; PUR; PA	ECHA (2021)
	Triethyl phosphate (10%), Melamine (25%), Dimethyl propylphosphonate (15%), 2,2-bis(chloromethyl)trimethylene bis(bis(2-chloroethyl)phosphate) (12%)	PUR	ECHA (2021)
	1,3,5-Triazine-2,4,6(1H,3H,5H)-trione, compound with 1,3,5-triazine-2,4,6-triamine (1:1) (5–30%)	PUR; PA	ECHA (2021)
	Phenol, isopropylated, phosphate (3:1) (15–35%), 1,1′-(ethane-1,2-diyl)bis[pentabromobenzene] (15–35%), Alkanes, C14–17, chloro (medium-chain chlorinated paraffins (MCCP)) (15%), 1,1′-(isopropylidene)bis[3,5-dibromo-4-(2,3-dibromo-2-methylpropoxy)benzene] (15%), Reaction product of tris(2-chloropropyl) phosphate and tris(2-chloro-1-methylethyl) phosphate and Phosphoric acid, bis(2-chloro-1-methylethyl) 2-chloropropyl ester and Phosphoric acid, 2-chloro-1-methylethyl bis(2-chloropropyl) ester (15%)	PUR; PVC (soft)	ECHA (2021)
	Short, medium, long chain chlorinated paraffins (SCCP/MCCP/LCCP) Boric acid, Brominated flame retardants with antimony (Sb) as synergist (e.g. Polybrominated diphenyl ethers (PBDEs), Decabromodiphenylethane, Tetrabromobisphenol A (TBBPA)) (3–25%), Phosphorous flame retardant (e.g. Tris(2-chloroethyl)phosphate (TCEP) Tris(2-chlorisopropyl)phosphate (TCPP))		Hahladakis et al. (2018) recreated from Hansen et al. (2013)
	Hexabromocyclododecane (HBCDD) (0.7–3%)		Hahladakis et al. (2018) recreated from Hansen et al. (2013)
	Hexabromocyclododecane (HBCDD) (1.1E-05–0.096%), Tetrabromobisphenol A (TBBPA) (3.8E-07–5.5E-05%), 1,2-bis(2,4,6-tribromophenoxy)ethane (BTBPE) (4.4E-06–2.2E-05%), Decabromodiphenylethane (DBDPE) (2.2E-05–4.2E-04%)	EPS, XPS, extruded PS	Rani et al. (2014)
	Hexabromocyclododecane (HBCDD) (5.2E-04-0.87%)	EPS	Jang et al. (2017)
Plasticizers Other functions	Triphenyl phosphate (2%)	ABS	ECHA (2021)
	Fatty acids, C16–18, C12–18-alkyl esters (0.5%)	ABS; PC	ECHA (2021)
	Amides, C16-C18 (even), N,N′-ethylenebis (1%)	Polyolefins; PVC (soft); PA; (E)PS	ECHA (2021)
	Isodecyl diphenyl phosphate (10%)	PUR	ECHA (2021)
	N-butylbenzenesulphonamide; N-butylbenzenesulfonamide (10–15%)	PUR; PA	ECHA (2021)
	Tributyl-O-Acetyl citrate (10–35%), Triethyl citrate (10–35%), Tributyl citrate (10–35%), Dibutyl phthalate (10–35%), Bis(2-ethylhexyl) adipate (10–35%), Dibutyl adipate (10–35%), Dimethyl sebacate (10–35%), Dibutyl sebacate (10–35%), Dihexyl adipate (10–35%), Decanedioic acid, 1,10-bis (2-ethylhexyl) ester (10–35%), Di-allyl phthalate (10–35%), Di-n-butyl terephthalate (10–35%), 1,3-Propanediol, 2,2-dimethyl-, 1,3-dibenzoate (10–35%), Linseed oil, epoxidized (10–35%), 2,2-bis[[(1-oxopentyl)oxy]methyl] propane-1,3-diyl divalerate (10–35%), Bis(tridecyl) adipate (10–35%), Diisotridecyl adipate (10–35%), Di-isotridecyl phthalate (10–35%), Diisononylphthalate (10–35%), Diisononyladipate (10–35%), Bis(2-propylheptyl) phthalate (10–35%), 1,2-Benzenedicarboxylic acid, di-C11–14-branched alkyl esters, C13-rich (10–35%), Di-C8–10-branched alkyl esters, C9-rich (10–35%), Di-C9–11-Branched alkyl esters, C10-rich; Di-isodecyl phthalate (10–35%), 1,2-Benzenedicarboxylic acid, di-C16–18-alkyl esters (10–35%), 1,2,4-Benzenetricarboxylic acid, tri-C9–11-alkyl esters (10–35%), 1,2-Cyclohexanedicarboxylic acid, diisononyl ester, reaction products of hydrogenation of di-isononylphthalates (n-butenes based); Di-isononyl cyclohexanoate (10–35%), Nonylbenzoate, branched and linear (10–35%), Reaction mass of benzyl 2-ethylhexyl adipate, bis (2-ethylhexyl) adipate, dibenzyl adipate (10–35%), bis(decyl and/or dodecyl) benzene-1,2-dicarboxylate (10–35%), Dodecanoic acid, ester with 1,2,3-propanetriol, acetylated (10–35%)	PUR; PVC (soft)	ECHA (2021)

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Table 1b (*continued*)

Function of additive	Additive (typical amount in polymer/plastic, wt-%)	Polymer type (see Table SI-1 for full polymer names)	Reference
	Di-2-ethylhexyl phthalate (2–35%)	PUR; PVC (soft); ABS; (E)PS	ECHA (2021)
	Triacetin (10%), Tris(2-ethylhexyl) benzene-1,2,4-tricarboxylate (35%), Isosorbide diesters (30%)	PVC (soft)	ECHA (2021)
	Short. medium and long chain chlorinated paraffins (SCCP/MCCP/LCCP); Diisoheptylphthalate (DIHP); DHNUP; Benzyl butyl phthalate (BBP); Bis (2-ethylhexyl)phthalate (DEHP): Bis(2-methoxyethyl) phthalate (DMEP): Dibutyl phthalate (DBP); dipentyl phthalate (DPP), di-(2-ethylhexyl) adipate (DEHA), di-octyladipate (DOA), diethyl phthalates (DEP), diisobutylphthalate (DiBP); Tris(2 chloroethyl) phosphate (TCEP); dicyclohexyl phthalate (DCHP), butyl benzyl phthalate (BBP), diheptyl adipate (DHA), heptyl adipate (HAD), and heptyl octyl adipate (HOA) (10–70%)	Approx. 80% is used in PVC and 20% in cellulose plastic	Hahladakis et al. (2018) recreated from Hansen et al. (2013)
	Sorbitan tristearate (0.5%)	Polyolefins	ECHA (2021)
	Palmitic acid (1%), Stearic acid (1%), (*Z*)-docos-13-enamide (0.1%), octadecanamide (0.2–3%), Lauric acid (1%), Reaction products resulting from the esterification of Sorbitol with C8–18 (even) and C18 unsaturated fatty acids in the ratio of 1:1 (4%)	Polyolefins	ECHA (2021)
	Glycerides, C16–18 mono- and di- (1%)	Polyolefins; ABS; PVC (rigid); (E)PS	ECHA (2021)
	Glycerides, C16–18 mono- (0.3–0.8%)	Polyolefins; ABS; PVC (rigid); PMMA; PC; (E)PS	ECHA (2021)
	Kaolin (15–20%)	Polyolefins; PVC (rigid); PA	ECHA (2021)
	Fatty acids, C16–18 (1%)	Polyolefins; PVC (soft); ABS; PVC (rigid); PET; PMMA	ECHA (2021)
	Benzenamine, reaction products with aniline hydrochloride and nitrobenzene, hydrochlorides (1%)	Polyolefins; PUR; PVC (soft); ABS; PVC (rigid); PET; PMMA; PA; PC; (E)PS	ECHA (2021)
	Azodicarbonamide (0.1%)	Polyolefins; PVC (rigid)	ECHA (2021)
	N,N′-hexane-1,6-diylbis[3-(3,5-di-tert-butyl-4-hydroxyphenylpropionamide)] (0.5%)	PUR; PA	ECHA (2021)
	(*Z*)-N-octadecyldocos-13-enamide (5%)	PVC (soft)	ECHA (2021)
	Zinc distearate (0.5–1%)	PVC (soft); PVC (rigid); PET; PA	ECHA (2021)
	Slip agents: Fatty acid amides (primary erucamide and oleamide), fatty acid esters, metallic stearates (for example, zinc stearate), and waxes (0.1–3%)	The amounts are dependant on the chemical structure of the slip agent and the plastic polymer type	Hahladakis et al. (2018) recreated from Hansen et al. (2013)
	Lubricants (0.1–3%)		Hahladakis et al. (2018) recreated from Hansen et al. (2013)
	Curing agents: 4,4′- Diaminodiphenylmethane (MDA); 2,2′-dichloro-4,4′- methylenedianiline (MOCA); Formaldehyde – reaction products with aniline; Hydrazine; 1,3,5-Tris(oxiran-2-ylmethyl)- 1,3,5-triazinane-2,4,6-trione (TGIC)/1,3,5-tris[(2S and 2R)- 2,3-epoxy-propyl]-1,3,5- triazine-2,4,6-(1H,3H,5H)- trione (β-TGIC) (0.1–2%)	Peroxides and other crosslinkers, catalysts, accelerators	Hahladakis et al. (2018) recreated from Hansen et al. (2013)
	Blowing agents: Azodicarbonamide, benzene disulphonyl hydrazide (BSH), pentane, CO_2 (amount depends on the density of the foam and the potential gas production of the agent)		Hahladakis et al. (2018) recreated from Hansen et al. (2013)
	Biocides: Arsenic compounds; Organic tin compounds; triclosan (0.001–1%)	Soft PVC and foamed polyurethanes are the major consumers of biocides. They vary in chemical structures and include chlorinated nitrogensulphur heterocycles and compounds based on Sn, Hg, As, Cu and Sb, e.g. tributyltin and 10,10′-oxybisphenoarsine.	Hahladakis et al. (2018) recreated from Hansen et al. (2013)
	Special effects: Al and Cu powder, lead carbonate or bismuthoxichloride and substances with fluorescence. Substances with fluorescence might migrate, the former not		Hahladakis et al. (2018) recreated from Hansen et al. (2013)
	Fillers: Calcium carbonate, talk, clay, zinc oxide, glimmer, metal powder, wood powder, asbest, barium sulphate, glass microspheres, silicious earth (up to 50%)		Hahladakis et al. (2018) recreated from Hansen et al. (2013)
	Reinforcements: Glass fibers, carbon fibers, aramide fibers. (15–30% for glass, due to is high density)		Hahladakis et al. (2018) recreated from Hansen et al. (2013)

showed very high variability, with concentrations highly above average in areas influenced by riverine discharges such as the Yangtze estuary (Zhao et al., 2014), or by surface currents that could be responsible for the dispersal of plastics away from the source area on land (Yamashita and Tanimura, 2007). Regarding variability for the same site, rough wind conditions were reported to reduce the concentration of floating plastic particles (Collignon et al., 2012; Reisser et al., 2013; Suaria et al., 2016), as could be expected in vertically mixed compared to stratified water columns, which was observed for earlier studies (Kukulka et al., 2012).

Table 2
Studies (published since 2010) on abundance of microplastic particles in surface and sub-surface water, only sampling with neuston nets, e.g. manta trawls, and pumps for surface and sub-surface water, respectively. Within each region the studies are sorted in descending order after lower detection limit (net/filter size).

Site/region (country)	Abundance Mean (±sd) or min-max (per m^3)	Sampling method; lower detection limit (net/filter/sieve size) (μm)	Reference
Baltic Sea Region and other Nordic waters:			
Surface water:			
Stockholm Archipelago (Sweden)	0.19–7.73	Manta trawl: >335 μm	Gewert et al. (2017)
S and S-W of Svalbard (Norway)	0.0–1.31; 0.34 ± 0.31	Manta trawl: >333 μm	Lusher et al. (2015)
Skagerrak/ Kattegat, Baltic Sea and Gulf of Bothnia	0.04	Manta trawl: >333 μm	Schönlau et al. (2020)
Gulf of Finland (Finland)	0.3–2.1 (manta), 0–3.4 (pump 300 μm), 0–8.2 (pump 100 μm)	Manta trawl: >333 μm. Submersible pump: >100 μm and >300 μm	Setälä et al. (2016)
Ryaverket (Sweden)	0.9–2.9	Manta trawl: >330 μm	Magnusson et al. (2016)
Gullmarsfjord (Sweden)	0.41	Manta trawl: >330 μm	Magnusson et al. (2016)
Gulf of Finland, Northern Baltic proper, Gulf of Riga	<4	Manta trawl: >330 μm	Unpublished, Kati Lind & Inga Lips, data from 2016
Archipelago Sea (Finland)	0.73; 0.48; 0.25 ± 0.07	Manta trawl: >300 μm	Magnusson (2014)
Gullmarsfjord (Sweden)	0.18–0.92	Manta trawl: >300 μm	Karlsson et al. (2020)
Danish Seas, South Funen Archipelago (Denmark)	0.07 ± 0.02; Concentration in low-volume bulk samples is not comparable to manta trawl	Manta trawl, low-volume bulk sampler: >300 μm	Tamminga et al. (2018)
Arkona Basin/ Bornholm Basin	0.0–8.0 (particles); 0.0–35.0 (fibers)	Manta trawl: >300 μm	Norén et al. (2015)
Kattegat/The Sound/Arkona Basin/Baltic Sea	10 μm: 4000 (fibers max), 32,000 (particles mean); 300 μm: 0–8 (fibers) and 0–2.5 (particles)	Manta trawl: ≥10 μm and ≥300 μm	Magnusson and Norén (2011)
North Sea (Denmark)	0.39	Manta trawl: >100 μm.	Mintenig (2014)
Kattegat (Denmark)	3.54	Manta trawl: >100 μm.	Mintenig (2014)
Belt Sea (Denmark)	1.44	Manta trawl: >100 μm.	Mintenig (2014)
Gulf of Finland (Finland)	10–650 (fibers); 500–9400 (particles)	Pump: >20 μm	Talvitie et al. (2015)
Sub-surface water:			
Skagerrak (Sweden)	10.3 ± 17.0; 0.84 ± 0.3	Pump (0.1–1.5 m): >300 μm	Norén et al. (2014)
Gullmarsfjord (Sweden)	0–0.4	Pump (5–10 cm): >300 μm	Karlsson et al. (2020)
Skagerrak/ Kattegat, Baltic Sea and Gulf of Bothnia	300 μm: 0.10; 50 μm: 3.82	Pump (10–20 cm): 500, 300 and 50 μm	Schönlau et al. (2020)
S and S-W of Svalbard	0.0–11.5; 2.68 ± 2.95	Pump (6 m): >250 μm	Lusher et al. (2015)
Baltic Sea (Russian economic zone)	32.2 ± 50.4	Pump (up to 100 m): >174 μm	Zobkov et al. (2019)
Other European Waters incl. the Arctic Ocean:			
Surface water:			
Western Mediterranean Sea	112,000 pieces/km^2	Manta trawl: >780 μm	Schmidt et al. (2018)
Gulf of Oristano, Sardinia, Italy	0.010–0.35	Manta trawl: >500 μm	de Lucia et al. (2014)
Arctic Ocean, Northern Atlantic Ocean and the Baltic Sea	0.06 ± 0.04	Manta trawl: >335 μm	Hänninen et al. (2021)
Arctic Ocean	0.0059–2.458	Manta trawl: >330 and 500 μm	Cozar et al. (2017)
Northwest Mediterranean Sea	0.1–8.92	Manta trawl: >333 μm	Collignon et al. (2012)
NW of Sardinia (Mediterranean Sea)	0.04–1.69	Manta trawl: >333 μm	Panti et al. (2015)
Northwestern Mediterranean	0.216–5.784	Manta trawl: >333 μm	Pedrotti et al. (2016)
Aegean-Levantine Sea	7.68 ± 2.38	Manta trawl: >333 μm	van der Hal et al. (2017)
Whole Mediterranean	62,000 pieces/km^2	Manta trawl: >333 μm	Ruiz-Orejón et al. (2016)
Western Mediterranean Sea	0.26 ± 0.33	Manta trawl: >330 μm	Baini et al. (2018)
Aegean-Levantine Sea	140,418 ± 120,671 pieces/km^2	Manta trawl: >330 μm	Güven et al. (2017)
Western Mediterranean Sea	129,682 pieces/km^2	Manta trawl: >330 μm	Faure et al. (2015)
Plymouth Sound	0.64 (0.0–1.24)	Manta trawl: >300 μm	Green et al. (2018)
North Atlantic	0.035–0.870	Neuston net: >300 μm	Poulain et al. (2019)
Adriatic Sea	472,000 ± 201,000 pieces/km^2	Neuston net: >300 μm	Gajšt et al. (2016)
Mediterranean Sea	0.020–11.30	Neuston net: >200 μm	Suaria et al. (2016)
Mediterranean Sea	1.219	Neuston net: >200 μm	Cozar et al. (2015)
Ligurian (Mediterranean) Sea	0.94 ± 2.55	Neuston/ plankton net: >200 μm	Fossi et al. (2012)
Sardinian (Mediterranean) Sea	0.13 ± 0.27	Neuston/ plankton net: >200 μm	Fossi et al. (2012)
Bay of Calvi, Corsica, France	0.013–3.438	Neuston net: >200 μm	Collignon et al. (2014)
Sub-surface water:			
Arctic/North Pacific	1.72 ± 0.93	Pump (0–50 m): >500 μm	Amélineau et al. (2016)
Atlantic Ocean	148 ± 131	Pump (3 m): >300 μm and 10 μm	Enders et al. (2015)
Open ocean sub-surface water in Northeast Atlantic Ocean	2.46 ± 2.43	Intake water (3 m): >250 μm,	Lusher et al. (2014)
Arctic central basin	0.7	Pump (8.5 m): >250 μm	Kanhai et al. (2018)
Atlantic	2782 ± 3393	Pump (10–240 m): >25 μm	Pabortsava and Lampitt (2020)
West Greenland	142 ± 85.83	Pump (5 m): >10 μm	Rist et al. (2020)

(*continued on next page*)

Table 2 (*continued*)

Site/region (country)	Abundance Mean (±sd) or min-max (per m^3)	Sampling method; lower detection limit (net/filter/sieve size) (μm)	Reference
Western Atlantic Ocean:			
Surface water:			
Fernando de Noronha, Abrolhos and Trindade, Brazil (south Atlantic)	0.03	Manta trawl: >300 μm	Ivar do Sul et al. (2014)
Sub-surface water:			
St. Peter and St. Paul Archipelago, Brazil (equatorial Atlantic)	0.01	Sub-surface plankton tows (0.6 m): >300 μm	Ivar do Sul et al. (2013)
Pacific Ocean and Asia:			
Surface water:			
Bering Sea (North Pacific)	0.0022–3.14	Manta trawl: >505 μm	Doyle et al. (2011)
Great Barrier Reef (Australia)	0.040–0.470	Plankton net: >355 μm	Jensen et al. (2019)
Tokyo Bay, Suruga Bay, Ise Bay, Seto Inland Sea (Asia)	0.03–0.075	Neuston net: >350 μm	Isobe et al. (2015)
Southern Ocean	0.031	Neuston net: >350 μm	Isobe et al. (2017)
Louisiana coast, northern Gulf of Mexico	11.1 ± 2.8	Neuston net: >335 μm	Mauro et al. (2017)
Arctic/North Pacific	0.001–49.27	Neuston net: >335 μm	Law et al. (2014)
Australian coast (south Pacific)	0.0026–0.288	Manta trawl: >333 μm and Neuston net: >335 μm	Reisser et al. (2013)
Yangtze Estuary, east China Sea	4137.3 ± 2461.5 (estuarine); 0.167 ± 0.138 (sea)	Pump (estuarine) (1 m): >32 μm; Neuston net (sea): >333 μm	Zhao et al. (2014)
North Pacific central gyre	85,184 pieces/km^2 (0.017)[a]	Manta trawl: >333 μm	Carson et al. (2013)
North Pacific subtropical gyre	21,000; 448,000 pieces/km^2 (0.0042–0.089)[a]	Manta trawl: >333 μm	Goldstein et al. (2013)
South Pacific subtropical gyre	26,898 particles/km^2 (0.0054)[a]	Manta trawl: >330 μm	Eriksen et al. (2013)
Geoje Island, South Korea	47 ± 192	Manta trawl: >330 μm	Song et al. (2014)
Nakdong Rivermouth, Southern Sea of Korea	57–62 before rainy season (May); 0.64–860 after rainy season (July)	Manta trawl: >330 μm	Kang et al. (2015)
Arctic/North Pacific	0.018–0.310	Manta trawl: >330 μm	Mu et al. (2019)
Sub-surface water:			
North Pacific	4.6 ± 3.1	Pump (5–1000 m): >100 μm	Choy et al. (2019)
Vancouver Island (eastern north Pacific, Canada)	2080 ± 2190	Intake water (4.5 m), copper sieves with pore sizes: 250 μm, 125 μm, and 62.5 μm.	Desforges et al. (2014)

[a] Lusher et al. (2015) converted values in km^2 to m^3 from: km^2 to m^2 by division by 1,000,000 followed by multiplication by 0.2 m. m^2 to m^3 by 0.2 multiplication.

From Table 2 the reported mean particle abundance in surface water was 1.3 and 1.8 particles/m^3, for mesh sizes >300 μm and >100 μm, respectively. These numbers are comparable, possibly because the sampling sites with use of mesh sizes >100 μm were in the Danish North Sea, Kattegat and the Belt Sea, which can be considered less polluted than the Baltic Sea. As mentioned previously measured abundances in Kattegat/The Sound/Arkona Basin/Baltic Sea with mesh size >10 μm were significantly higher (32,000 particles/m^3) (Magnusson and Norén, 2011) than findings with mesh sizes >100 μm and >300 μm. In the Mediterranean Sea measured abundances in the surface water with trawl mesh sizes >200 μm and >300 μm were 2.2 particles/m^3, which is comparable with the Nordic findings. Surface water measured with trawl mesh sizes >300 μm at various sites in the Pacific Ocean had a mean value of 7.1 particles/m^3, which is higher than the Nordic waters and the Mediterranean Sea. However, omitting single high values at Geoje Island, South Korea (47 particles/m^3) and Nakdong Rivermouth, Southern Sea of Korea (60 particles/m^3) the mean abundance was 4.2 particles/m^3.

3. Influence of weathering on residual additive concentrations and bioavailability

3.1. Migration and leaching of residual additives

Additives are typically not chemically bound to the polymer, but only weakly associated via van der Waals forces (Zhang and Chen, 2014). Therefore, they have the ability to leach out of the polymer matrix into the surrounding environment (Schrank et al., 2019). During the fragmentation and weathering processes, the surface area of the particle increases, which may also increase the leaching rates (Kwon et al., 2017).

Hahladakis et al. (2018) performed a review of the migration and release potential of various additives present in plastic food contact materials, and Yusà et al. (2020) performed a risk assessment of plastic additives in LDPE to check for the safety of their migration in food contact materials. Analogously, the direct exposure of marine organisms to residual plastic additives, via ingestion of particles, can be divided into four major steps (Ferrara et al., 2001): i) diffusion of chemicals through the polymers, ii) desorption of the molecules from the polymer surface, iii) sorption of the chemicals at the plastic–stomach/gut interface, and iv) absorption of the chemicals in the stomach/gut. The extent of migration and leaching depends on the characteristics of the plastic materials (e.g. initial concentration of the chemicals in the plastic, thickness of the plastic item, crystallinity of the plastic, surface structure of the plastic item), the characteristcs of the chemical and environmental conditions, such as temperature (Hahladakis et al., 2018). Migration will decrease with time as the concentration gradient between the plastic material and the surroundings becomes smaller (Hansen et al., 2014). A summary of the migration behaviour of some chemical additives is shown in Fig. 2 (Hansen et al., 2013, 2014).

Experimental studies on migration and leaching of residual plastic additives are scarce. Hansen et al. (2014) states that the migration rate of the plasticizer DEHP is likely to be in the range of 0.1–1% per year or below. Moreover, a study specifically addressing leaching to water of ΣHBCDD from PS foam macrodebris, larger than 2 cm with outer 2–3 mm removed, collected on beaches showed that leaching of ΣHBCDD occurred at rates of over 150 ng/g/d for more than 30 days (Aminot et al., 2020). In a study by Capolupo et al. (2020) the chemical composition of aqueous seawater leachates from a suite of PP, PET, PS, PVC (cryomilled, 1000 mm sieved fraction) and car tire rubber (CTR)-derived granulate (1–2 mm), was assessed. A number of organic and metal compounds were detected in the leachates, including plasticizers, antioxidants, antimicrobials, lubricants, and vulcanizers. CTR and PVC materials and their corresponding leachates had the highest content of tentatively identified organic additives, while PET had the lowest. The metal content also varied between polymer leachates. Notably high

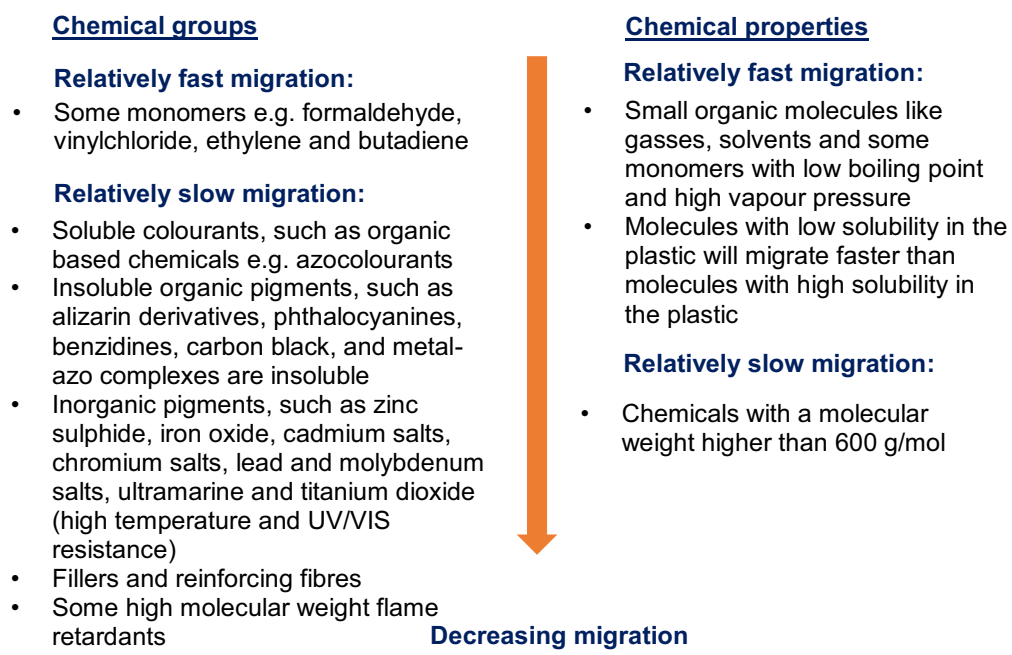

Fig. 2. Summary of the migration potential of certain residual additives in plastics under ambient environmental conditions (Hansen et al., 2013, 2014).

concentrations in the leachates were 1460 ± 35 μg/L benzothiazole in CTR, 510 ± 218 μg/L phthalide in PVC, 166 ± 3 μg/L acetophenone in PP, 13.0 ± 0.3 μg/L cobalt in CTR, 8.7 ± 0.2 μg/L cobalt in PET, 5138 ± 128 μg/L zinc in CTR, 6357 ± 140 μg/L in PVC, and 9.0 ± 0.3 μg/L lead in PP. Fikarová et al. (2019) found that out of the eight regulated phthalates contained in the materials, only the two most polar compounds, namely, dimethyl phthalate and diethyl phthalate as well as bisphenol A (BPA), leached significantly into seawater in less than 2 h, with bioaccessibility percentages of 51–100%. However, these studies did not address the effect of weathering on the migration and leaching of additives. Halsband et al. (2020) investigated the organic chemical and metal additive content of in crumb rubber granulate materials from a supplier and material collected from outdoor sports fields that had been in the environment for an unknown period of time. Leachates were investigated over a 30-day period in seawater and revealed that the most abundant leachate components were benzothiazole and Zn, Fe, Co, as well as detectable levels of PAHs and phenolic compounds. While organic chemical concentrations in the leachates stabilized within days, metals continued to leach out over the 30-day period.

3.2. Weathering

In the marine environment, plastic is degraded due to different processes such as solar UV-induced photo-oxidation, hydrolysis, mechanical and thermal degradation as well as biodegradation (Gewert et al., 2015; Andrady, 2015; Jahnke et al., 2017). These processes eventually fragment the plastic particles resulting in MP or even nanoparticles. For example exposure to UV radiation and the presence of atmospheric oxygen will initiate a photo-oxidative degradation, which changes the polymer structure, e.g. by breaking or cross-linking the polymer chains or forming new chemical functional groups, with implications for the material properties (Gewert et al., 2015). Chen et al. (2019) found that stresses such as solar irradiation increased the release of endocrine disrupting compounds (EDCs) compared with no treatment, possibly related to more effective diffusion due to the increase in plastic surface area.

Compared to other environmental compartments, the degradation of plastic is particularly slow in the marine environment as it is limited to solar radiation and slow thermal oxidation (Lithner et al., 2011). However, degradation times vary depending on the type of plastics, the presence of additives and environmental conditions. As an example PUR has relatively low resistance to biodegradation and is degraded by heat, oxidation, light, hydrolysis, mechanical shear and various pollutants (Lithner et al., 2011). Photo-initiated oxidative degradation of PE, PP and PS leads to a decrease in the molecular weight and formation of carboxylic end groups, and UV-light is particularly effective at initiating dechlorination of PVC. Polymer fragments with lower molecular weight formed by chain scission after UV-initiation can then be biodegraded. Plastics that contain heteroatoms in the main chain, like PET and PU, can be degraded by hydrolysis, photo-oxidation, and biodegradation, also leading to the formation of smaller fragments and carboxylic end groups (Gewert et al., 2015). Andrady (1990) found that exposure in seawater reduced the tensile strength of expanded polystyrene by over 60% in 4 months. However, in some situations, the time for complete degradation could be several hundred years (Lithner et al., 2011).

3.3. Additives in weathered plastic

Empirical data of residual additives in plastic materials are rare, and even less information is available for weathered plastic particles. A comparison between the amounts in the original plastic products and the weathered plastics would give an understanding of the leaching potential as a part of the weathering processes. In a chemical risk assessment the concentrations of additive residuals in environmental plastic particles and particles ingested by marine species are needed for correct determination of exposure levels. The available data are compiled in Table 3.

Concentrations of nonylphenols (NP) and octylphenols (OP) found in PE and PP particles collected from 18 beaches in Japan and Malaysia and four Japanese coasts (Mato et al., 2001, 2002) varied among the sampling sites. Other studies showed large piece-to-piece variation in the concentrations of residual additives as well. For example, PBDEs were detected in all the fragment PE and PP samples analyzed from the open ocean and remote and urban beaches, while NP and OP were detected in many but not all samples from the open ocean and remote beaches (Hirai et al., 2011). In most locations including urban coasts, only trace concentrations (<1 ng/g) of BPA were detected, but sporadic high concentrations were detected in some plastic fragment samples from remote

Table 3
Chemical additives and their concentrations in weathered and applied (new) plastic particles. DF = detection frequency. The chemicals reported here are potential additives, as they may also be sorbed from the surrounding marine environment from sources other than plastic. n.d.: not detected

Chemical	Type (potential additive)	Plastic type	Conc. in weathered plastic (ng/g plastic)	Sample/site information	Reference
Nonylphenols (NP)	Unreacted raw material	PE and PP	18–17,000	PE and PP pellets collected from 18 beaches in Japan and Malaysia	Mato et al. (2002)
Octylphenols (OP)	Unreacted raw material	PE and PP	0–41	PE and PP pellets collected from 18 beaches in Japan and Malaysia	Mato et al. (2002)
NP	Unreacted raw material	PP	13–16,000	PP resin pellets collected from four Japanese coasts	Mato et al. (2001)
PBDEs	Flame retardant	PE and PP	0.02–9909 (total PBDEs)	Plastic fragments (approx. 10 mm) from open ocean and from remote and urban beaches	Hirai et al. (2011)
Deca-BDE (BDE-209)	Flame retardant	PE and PP	0.1–9907	Plastic fragments (approx. 10 mm) from open ocean and from remote and urban beaches	Hirai et al. (2011)
NP	Unreacted raw material	PE and PP	0.3–3940	Plastic fragments (approx. 10 mm) from open ocean and from remote and urban beaches	Hirai et al. (2011)
OP	Unreacted raw material	PE and PP	0.1–153	Plastic fragments (approx. 10 mm) from open ocean and from remote and urban beaches	Hirai et al. (2011)
Bisphenol A	e.g. UV-stabilizer	PE and PP	0–730	Plastic fragments (approx. 10 mm) from open ocean and from remote and urban beaches	Hirai et al. (2011)
DEHP, DnBP, DEP, DiBP, 2,4-di-tert-butylphenol (2,4-DTBP)	Plasticizers, antioxidant	PE, PP	Qualititive identification	Ten particles extracted from two sediment samples from Norderney, a North Sea island	Fries et al. (2013)
DnBP, DiBP, DEP, DMP	Plasticizers	PS	Qualititive identification	Ten particles extracted from two sediment samples from Norderney, a North Sea island	Fries et al. (2013)
DEHP, DiBP, DEP	Plasticizers	Polyamide-6 (PA-6)	Qualititive identification	Ten particles extracted from two sediment samples from Norderney, a North Sea island	Fries et al. (2013)
UV-326, UV-327, UV-328, UV-320, UvinualMC80	UV stabilizers	PE, PP, PET, PC, Poly(acrylic/styrene)	Qualitative screening	Plastic debris (n = 19) from coastal beaches along with new similar plastics (n = 25)	Rani et al. (2015)
Irganox 1076, DEHP, antioxidant no 33, din-octylisophthalate, diisooctyl phthalate, hexanoic acid 2-ethyl-hexadecyl ester, hydrocarbons	Antioxidants, plasticizers	PE, PP, PET, PC, Poly(acrylic/styrene)	Qualitative screening	Plastic debris (n = 19) from coastal beaches along with new similar plastics (n = 25)	Rani et al. (2015)
UV-328	Benzotriazole UV stabilizer	PE, PP, PET	n.d–1600 (DF = 97%) new: 70 ± 160	29 pieces of marine plastic debris from beaches along with their corresponding new plastic products (n = 27)	Rani et al. (2017)
UV-326	Benzotriazole UV stabilizer	PE, PP, PET	n.d.–82,000 (DF = 100%) new: 10,000 ± 36,000	29 pieces of marine plastic debris from beaches along with their corresponding new plastic products (n = 27)	Rani et al. (2017)
UV-327	Benzotriazole UV stabilizer	PE, PP, PET	n.d.–20,000 (DF = 66%) new: 2100 ± 7400	29 pieces of marine plastic debris from beaches along with their corresponding new plastic products (n = 27)	Rani et al. (2017)
UV-320	Benzotriazole UV stabilizer	PE, PP, PET	n.d.-53,000 (DF = 90%) new: 4100 ± 15,000	29 pieces of marine plastic debris from beaches along with their corresponding new plastic products (n = 27)	Rani et al. (2017)
Butylated hydroxytoluene (BHT)	Antioxidant	PE, PP, PET	n.d-29,000 (DF = 93%) new: 20–50,000	29 pieces of marine plastic debris from beaches along with their corresponding new plastic products (n = 27)	Rani et al. (2017)
Irganox 1010	Antioxidant	PE, PP, PET	20–155,000 (DF = 100%) new: 230–700,000	29 pieces of marine plastic debris from beaches along with their corresponding new plastic products (n = 27)	Rani et al. (2017)
Irganox 1076	Antioxidant	PE, PP, PET	50,000–100,000 (DF = 90%) new: n.d-2,130,000	29 pieces of marine plastic debris from beaches along with their corresponding new plastic products (n = 27)	Rani et al. (2017)
2,4- DTBP	Antioxidant	PE, PP, PET	30–16,000 (DF = 100%) new: 31–28,000	29 pieces of marine plastic debris from beaches along with their corresponding new plastic products (n = 27)	Rani et al. (2017)
BPA	Antioxidants and UV-stabilizers	Each sample was composed of multiple small pieces of plastic	1.4–4.9 (DF = 63%)	Plastic debris sampled from manta trawls in the Atlantic Ocean	Rochman et al. (2014b)
Alkylphenols (4-nonylphenol, 4noctylphenol)	Unreacted raw material, antioxidant and plasticizer	Each sample was composed of multiple small pieces of plastic	22.3–341.5 (DF = 100%)	Plastic debris sampled from manta trawls in the Atlantic Ocean	Rochman et al. (2014b)
Alkylphenols ethoxylates (4-nonylphenol monoethoxylate, 4-nonylphenol Diethoxylate)	Antioxidants and UV-stabilizers	Each sample was composed of multiple small pieces of plastic	0.8–97.6 (DF = 100%)	Plastic debris sampled from manta trawls in the Atlantic Ocean	Rochman et al. (2014b)

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Table 3 (*continued*)

Chemical	Type (potential additive)	Plastic type	Conc. in weathered plastic (ng/g plastic)	Sample/site information	Reference
PBDEs	Flame retardants	Each sample was composed of multiple small pieces of plastic	0.1–4.6 (DF = 100%)	Plastic debris sampled from manta trawls in the Atlantic Ocean	Rochman et al. (2014b)
Tentative identification of plastic additives	Plasticizers, antioxidants, UV-stabilizer and cross-linking agents	LDPR, PP, HIPS, PET, polyester and nylon fibre	Qualitative chemical fingerprints	Analysis of 21 samples: plastic pellets from polymer manufacturers, microbeads from personal care products, MP collected from the aquatic environment, and synthetic fibers	Zhang et al. (2020)
HBCDD (α-, β-, and γ-isomers)	Flame retardant	PS	>100,000 (sum of isomers) – 1,940,000 (DF = 50%)	16 foam-like PS debris, collected along the French Riviera coastline	Aminot et al. (2020)
Tetrabromobisphenol A (TBBPA)	Flame retardant	PS	Approx. 1,000,000	16 foam-like PS debris, collected along the French Riviera coastline	Aminot et al. (2020)
Tribromophenol (TBP)	Flame retardant	PS	Approx. 1,000,000	16 foam-like PS debris, collected along the French Riviera coastline	Aminot et al. (2020)
BPA	Plasticizer	PS	4,565,000 (DF = 19%)	16 foam-like PS debris, collected along the French Riviera coastline	Aminot et al. (2020)
HBCDD	Flame retardant	EPS	n.d.-4,680,000	EPS debris and MP in South Korea and in 12 other countries in the Asia-Pacific region	Jang et al. (2017)
BPA	Stabilizer, antioxidant, plasticizer	PE	475,100 ± 881,500 (DF = 75%)	MP and mesoplastic sampled from the North Pacific Subtropical Gyre by a manta trawl	Chen et al. (2019)
NP	Stabilizer, antioxidant	PE	3700 ± 7700 (DF = 49%)	MP and mesoplastic sampled from the North Pacific Subtropical Gyre by a manta trawl	Chen et al. (2019)
Bisphenol S	Replacement for BPA	PE	7300 ± 25,900 (DF = 68%)	MP and mesoplastic sampled from the North Pacific Subtropical Gyre by a manta trawl	Chen et al. (2019)
OP	Antioxidant	PE	2500 ± 8700 (DF = 63%)	MP and mesoplastic sampled from the North Pacific Subtropical Gyre by a manta trawl	Chen et al. (2019)
UV-326	UV stabilizers	PE	23,000–180,000 (DF = 1.0%)	194 plastic pieces ingested by three different species of seabirds	Tanaka et al. (2019)
UV-327	UV stabilizers	PE, PEPD	5800–860,000 (DF = 2.1%)	194 plastic pieces ingested by three different species of seabirds	Tanaka et al. (2019)
UV-328	UV stabilizers	PP	1100–1400 (DF = 1.0%)	194 plastic pieces ingested by three different species of seabirds	Tanaka et al. (2019)
Benzophenone (BP-12)	UV stabilizers	PE	1,700,000 (DF = 0.5%)	194 plastic pieces ingested by three different species of seabirds	Tanaka et al. (2019)
HBCDD	Flame retardant	PE, PP	3000–29,000 (DF = 1.5%)	194 plastic pieces ingested by three different species of seabirds	Tanaka et al. (2019)
PBDEs (sum of BDE-202, −197, −203, −196, −208, −207, −206, and −209)	Flame retardant	PP	1100,000 (DF = 0.5%)	194 plastic pieces ingested by three different species of seabirds	Tanaka et al. (2019)
Styrene oligomers (sum of ST2, ST3, ST4, and ST5)	Impurities from the manufacture of PS	PE, PS, PEPD	n.d.–3,200,000 (DF = 2.1%)	194 plastic pieces ingested by three different species of seabirds	Tanaka et al. (2019)
15 organic additives in text	Plasticizers, antioxidants, UV stabilizers, flame retardants, and preservatives	PE, PP, PS	Qualitative screening	MP reference mixture (PTX001) and a marine litter derived PS foam sample	Kühn et al. (2020)
UV-326	UV stabilizer	PE, PP	Small group: n.d.-54,400 Large group: n.d.-680,000	141 plastic fragments collected on a beach on the Hawaiian island of Kauai	Tanaka et al. (2020)
UV-328	UV stabilizer	PE	Small group: n.d. Large group: n.d.-200	141 plastic fragments collected on a beach on the Hawaiian island of Kauai	Tanaka et al. (2020)
UV-327	UV stabilizer	PE, PP	Small group: n.d.-315,000 Large group: n.d.-763,000	141 plastic fragments collected on a beach on the Hawaiian island of Kauai	Tanaka et al. (2020)
BP-12	UV stabilizer	PE, PP	Small group: n.d.-75,500 Large group: n.d.-698,000	141 plastic fragments collected on a beach on the Hawaiian island of Kauai	Tanaka et al. (2020)

beaches and the open ocean (Hirai et al., 2011). Thus, it is difficult to establish a typical concentration of a residual additive or the influence of weathering on these concentrations.

HBCDD was abundantly detected in buoy debris and MP stranded along the Korean coasts, and in debris collected from the Asia-Pacific coastal region, in a study exclusively addressing EPS (Jang et al., 2017), indicating that HBCDD contamination via EPS debris is a common environmental issue worldwide. Particles from Alaskan beaches that were suspected to be debris from the 2011 tsunami in Japan, indicated that EPS debris had the potential for long-range transport in

the ocean, enabling the movement of plastic additives in marine environments over long distances, e.g. from source regions to regions of possible accumulation (Jang et al., 2017).

Aminot et al. (2020) measured concentrations of HBCDD (α-, β-, and γ-isomers), tetrabromobisphenol A (TBBPA), 2,4,6-tribromophenol (TBP) and BPA in foam-like plastic macrodebris (n = 16), identified as PS, from beaches. The outer surface (2–3 mm) of each macro debris was removed and only pristine material within the interior was analyzed for chemical composition and additives. Of the 16 PS debris screened for plastic additives, HBCDD was found in 8 fragments where the highest ΣHBCDD concentration was observed in a sample of black colour, attributed to graphite beads typically used in insulation boards, e.g. in buildings. Phenolic compounds were found in 3 samples. As TBP and BPA are additives and also degradation products of TBBPA, the presence of these phenolic compounds in some of the PS debris could either be because of their use as additives or be derived from the degradation of TBBPA during migration from the plastic debris, or photodegradation that might occur in the PS (Khaled et al., 2018).

Rani et al. (2015) conducted a qualitative screening of plastic debris (n = 19) collected from coastal beaches in South Korea along with similar new plastics (n = 25) and reported the presence of monomers, oligomers, intermediates, antioxidants, compounds for dyes and inks, flame retardants, plasticizers, lubricants, UV stabilizers, and agents for vulcanization. In total 231 chemicals were found in new plastic items and marine debris, of which the UV stabilizers UV-326, UV-327, UV-328, UV-320, UvinualMC80, antioxidants Irganox 1076, DEHP, antioxidant no 33, di-n-octylisophthalate, diisooctyl phthalate, hexanoic acid 2-ethyl-hexadecyl ester, and hydrocarbons were most frequently detected (included in Table 3). The chemical analysis yielded that 63 chemicals were detected in both new and debris plastic, and that 83 were detected in debris plastic only, possibly indicating adsorption of these chemicals from the environment.

The opposite was found in a study involving UV stabilizers and antioxidants (Rani et al., 2017). Except for Irganox 1076 and UV-326, levels of most antioxidants and UV stabilizers were relatively high in new plastics compared to corresponding plastic marine debris. However, additive levels varied widely among plastics, and variations also differed among chemicals, probably reflecting different weathering processes. Varying addition of chemicals within plastic products may also explain these variations, which further complicates the efforts of establishing concentrations of residual additives in weathered plastic particles.

Data on BPA, alkylphenols (and their ethoxylates) and PBDEs are also available for plastic debris from the Atlantic Ocean (Rochman et al., 2014b). All chemical groups were detected, some at concentrations up to 6 orders of magnitude above those found in the water column. Also studying BPA, NP and OP, in a broad study on various EDCs, Chen et al. (2019) reported the highest detection frequencies for BPA (75%), bisphenol S (BPS, 68%), OP (63%) and NP (49%) in MP and mesoplastic sampled from the North Pacific Subtropical Gyre.

New analysis techniques based on high resolution chemical characterization have been applied to identify additives in a variety of polymer types (Zhang et al., 2020). Additives found included plasticizers, antioxidants and cross-linking agents, however, these were not quantified. Tanaka et al. (2019) used a non-target screening analysis of additives and by-products for a total of 194 pieces of plastics ingested by three different species of seabirds, namely northern fulmars (*Fulmarus glacialis*) from the Faroe Islands, laysan albatross (*Diomedea immutabilis*) chick and 5 black-footed albatross (*Phoebastria nigripes*) chicks, both at Mukojima Island, Japan. Three benzotriazole UV stabilizers (UV-326, UV-327, UV-328), one benzophenone UV stabilizer (BP-12), two brominated flame retardants (HBCDD, deca-BDE), and styrene oligomers were identified and quantified. The concentrations were comparable to the concentrations of UV-326, UV-327 and UV-328 in PE, PP and PET pieces from beaches and new plastic products by Rani et al. (2017). Log Kow values for these compounds are all above 5.6, and the results indicate that such hydrophobic compounds can be transported within plastic fragments such as PE, polyethylene propylene diene (PEPD), PP and PS, and be taken up by seabirds. Styrene oligomers are retained in PS resin as impurities during the manufacture, but were also detected in PE and PEPD fragments, possibly due to contamination during production. In addition, Tanaka et al. (2019) also discuss the possibility of adsorption of leachate from PS fragments in the birds' stomach or the environment before ingestion.

Kühn et al. (2020) investigated the transfer of plastic additives from a marine litter derived MP reference mixture (PTX001) and a marine litter derived PS foam sample to stomach oil of northern fulmars. PTX001 was made of 351 macroplastic litter items, collected from a Dutch beach and cryomilled to a mix of variable sizes less than 3.0 mm in diameter. One gram of the PTX001 material contained around 400,000 plastic particles. The sample comprised a mixture of rigid (37%) and flexible (63%) items. The polymer composition comprised mainly PE (60.9%) and PP (27.7%), but with many other polymers present in small amounts. A sample of weathered expanded PS foam collected at the same time from the same beach was also used. Chemical analyses of the PTX001 sample showed the presence of various heavy metals, e.g. Cd, Cr, Cu, Ni and Pb, and light metals, e.g. Al. Non-target screening with a 90% confidence match to library spectra permitted identification of 15 different organic chemical additives, with 14 identified in PTX001 and 4 identified in PS. These substances include common additives such as plasticizers (TCEP, DBP, TPhP, DEHP, di(2-ethylhexyl) terephthalate), antioxidants (dibutylphenol, degradation product: 7,9-di-tert-butyl-1-oxaspiro(4,5) deca-6,9-diene-2,8-dione), UV stabilizers (bumetrizole), flame retardants (TCPP), preservatives (phenyl benzoate) and others (acetophenone, p-benzoquinone, propanediylbisbenzene, BCPP, triphenylbenzene). For some identified chemicals, however, the use or origin is unclear.

Tanaka et al. (2020) analyzed 141 plastic fragments collected on a beach on the Hawaiian island of Kauai for additives, i.e., a flame retardant (deca-BDE), ten benzotriazole UV stabilizers (UV-P, UV-PS, UV-9, UV-320, UV-350, UV-326, UV-329, UV-328, UV-327 and UV-234), and a benzophenone UV stabilizer (BP-12). The fragments were mostly PE with the remainder dominated by PP. They analyzed and compared two different size fractions, a small group (range: 4–7 mm) (PE, 90%; PP, 10%) and a large group (range: 1.5–8 cm) (PE, 89%; PP, 10%; polyethylene vinyl acetate copolymer, 1%). Deca-BDE was not detected in any sample. UV stabilizers were found in 13% of small fragments and in 33% of large fragments. UV stabilizers are generally used in plastic products at concentrations between 0.05%–2% (500–20,000 μg/g) of the plastic weight (Chanda, 2017). The upper limit, around 2% of addition is only exceptionally used, and may be found in low-density PE film for outdoor use (Chanda, 2017). The levels of additives in “small” fragments were lower than those in “large” fragments, which illustrates that the loss of additives had progressed further in “small” fragments compared to “large” fragments, indicating leaching or degradation of additives during fragmentation. However, notable concentrations of additives still remained in the “small” fragments, with a concentration in the same order of magnitude as the range used in production, indicating their importance as a vector of chemical additive exposure. Tanaka et al. (2020) furthermore analyzed five layers of three pieces and found that all of the chemicals in each of the three pieces showed lower concentrations in the surface layers than the other layers, and no clear difference in distribution trend among chemicals was observed. The observation of lower concentrations in the surface layers indicated that the loss of chemicals from the surface was faster than diffusion inside the polymer and/or leaching related to the degradation of the polymer in the surface layer.

The studies on chemical analysis of weathered plastic particles, compiled in Table 3, include samples mostly from beaches and shorelines, predominantly of PE, PP and PS, which reflects that they are produced in high volumes, and they have the ability to float. Thus, most of these measurements represent macroplastic rather than MP, often described as plastic debris. These studies do not distinguish between

chemicals added to the polymer and those sorbed from the environment. Brominated flame retardants, phthalate softeners and some metals, that have often been associated with marine plastic debris, can also originate from other sources, such as wastewater, sewage sludge or atmospheric deposition (Hermabessiere et al., 2017; Al-Odaini et al., 2015). However, concentrations in plastic particles are likely to be initially much higher than those that could be acquired through sorption from seawater. For example, the concentrations close to 10,000 ng/g of the flame retardant deca-BDE (BDE-209) in plastic fragments collected from the open ocean and from remote and urban beaches, are most likely the result of its use as an additive flame retardant in the plastics rather than as a result of uptake from seawater (Hirai et al., 2011). On the other hand, many potential additives have been detected in polymers where they are not expected to be used, e.g. according to Tables 1a and 1b, raising questions about the extent of sorption, or alternatively a more widespread use of high concentrations of additives than assumed: Polybrominated diphenyl ethers (PBDEs) detected in PE and PP (Hirai et al., 2011), are not normally used in products made of these polymers, but are more typical of PUR, PVC and EPS. Similarly, nonylphenols and octylphenols incidentally found in PE and PP particles (Mato et al., 2001, 2002; Hirai et al., 2011), typically occur as unreacted constituents in PVC and PC. Surprisingly, Fries et al. (2013) and Rani et al. (2015) identified several phthalates in PE, PP, PS, PET, PC, and polyamide particles, although they are mainly used as plasticizers in PVC. Hirai et al. (2011) found sporadically high concentrations of BPA in PE and PP fragments sampled at remote coasts and in the open ocean, which suggests that BPA most likely is a constituent of the plastic products, even though BPA is mainly considered a constituent monomer of PC and epoxy resin and not a common additive in PE or PP products. Due to its relatively low hydrophobicity (log Kow = 3.40), sorption of significant concentrations of BPA dissolved in surrounding seawater to marine plastics, appears unlikely (Hirai et al., 2011). In conclusion, the use of large amounts of leachable additives in plastic products, with or without obvious targeted purposes, seems more common and widespread than previously assumed.

3.4. Biouptake of residual additives from ingested plastics

Bioavailability in the aquatic environment can be defined as the external availability of a chemical to an organism. With respect to plastic additives, their bioavailability is influenced by the properties of the particle, the physical-chemical properties of the additive, environmental factors, such as temperature and pH, and the exposed organism (Fu et al., 2021; Schrap, 1991). Considering direct exposure, i.e. after ingestion of a plastic particles, the desorption from the particle depends on the conditions in the gut and stomach of an organism, including a concentration gradient of the chemical in question. In order to understand and describe better the bioavailability of additives in ingested plastic particles to marine organisms, studies on biodynamic modelling and experimental studies on leaching and uptake in different marine species have the focus in this chapter.

In a summary of studies on the adsorption of organic pollutants on MP, Fu et al. (2021) state that the main adsorption mechanisms of organic pollutants to MPs include hydrophobic interactions, van der Waals forces, partitioning, electrostatic interactions, and other non-covalent actions, and that multiple mechanisms are often involved. Weaker association, such as van der Waals forces, gives the additives the ability to leach out of the polymer matrix into the surrounding environment including the stomach and gut of organisms (Schrank et al., 2019; Zhang and Chen, 2014). The bioavailability of additives in the plastic may therefore differ from that of chemicals adsorbed from the environment, which can have stronger chemical bonds to the plastic particles (Lassen et al., 2015).

Bioaccumulation of plastic associated additives, i.e. the gradual accumulation of additives in an organism, and biomagnification, i.e. accumulation of additives through consumption of lower organisms containing the additives, are often inferred to occur in marine food webs (Miller et al., 2020). However, Miller et al. (2020) conclude that MP biomagnification across a general marine food web is not supported by field or laboratory studies, and that a lack of data precludes an assessment of potential trophic transfer and biomagnification of plastic additives. These findings indicate that, although bioaccumulation of MPs occurs within trophic levels, no clear evidence of biomagnification of MP and thus additives at higher trophic levels is available. Some additives, e.g. NP and BPA that are potentially metabolized, would less likely biomagnify up the food web (Takeuchi et al., 2009). However, a connection between the lack of biomagnification of e.g. NP and BPA and exposure of high trophic level animals remains obscure. More data are needed to fully understand the relative importance of direct exposure through ingestion of additives on plastic particles compared to their biomagnification potential (Ziccardi et al., 2016; Kwon et al., 2017).

Despite concentrations of HOCs associated with MP that can be orders of magnitude greater than the chemical concentrations in the surrounding seawater, fractional intake of nonpolar organic chemicals via the ingestion of plastic particles, estimated with a food-web model and a one-compartment model for a benthic invertebrate, a fish and a seabird, would be much smaller than via exposure from water, sediment and dietary uptake (Gouin et al., 2011; Bakir et al., 2016). The hypothesis that MP is not a significant contributor to the exposure of hazardous substances is substantiated by the argument that MP plays a minor role in the environment compared to natural organic particulates and natural prey because of the relatively lower abundance (Koelmans et al., 2016). Herzke et al. (2016) found from measurements of POP concentrations in fulmar liver and muscle tissue that plastic is more likely to act as a passive sampler than as a vector of POPs. Moreover, it has been indicated that the likelihood of additives leaching out into the gut lumen of organisms exposed to MP is low (Schrank et al., 2019). However, residual additives in plastic materials are typically not ubiquitous in the environment. The plastic particles are thus a primary source and migration/leaching would result in an increase in environmental concentrations potentially leading to indirect exposure, at least at a local scale (Kwon et al., 2017).

The exposure to chemicals in plastic particles in the aquatic environment depends on the ingestion by organisms (Besseling et al., 2019). When organisms ingest plastics with adsorbed chemicals, it is debated whether an increased bioavailability to organisms from bidirectional interaction, i.e. sorption and release of chemicals in plastics, can take place (Hartmann et al., 2017). Laboratory experiments and modelling studies linked concentrations of HOCs from MPs to concentrations in marine species, such as fin whale (*Balaenoptera physalus*), basking shark (*Cetorhinus maximus*) and juvenile yellowtail (*Seriola lalandi*). Combined with adsorption and desorption studies of HOCs on MPs the results indicated that chemicals can partition from MPs to organisms or from organisms to MPs, depending on experimental conditions (Ziccardi et al., 2016; Kwon et al., 2017; Nor and Koelmans, 2019). Model analyses showed that the contribution of POPs from plastic waste to their overall bioaccumulation may be rather small, due to a lack of a gradient from chemicals in ingested plastic to the biota lipids over time as the concentration in biota lipids increases. In fact, plastic particles can act as a cleaning mechanism for lipophilic chemicals with log Kow values higher than 5 or 6, due to a gradient from the lipids to the plastic particles (Koelmans et al., 2013, 2014). Thus, MP ingestion may either clean or contaminate the organism, depending on the chemical fugacity gradient between ingested plastic and organism tissue (Koelmans, 2015). These models showed that plastic ingestion by the lugworm (*Arenicola marina*) yielded NP and BPA concentrations in the tissue that stayed below the lower ends of global NP and BPA concentration ranges in lugworms, and therefore were not likely to constitute a relevant exposure pathway (Koelmans et al., 2014). For Atlantic cod (*Gadus morhua*), plastic ingestion also appeared to be a negligible pathway for exposure to NP and BPA. Koelmans et al. (2014) note that the results do not imply that leaching from marine plastics is irrelevant for aquatic

species, as additives can cause indirect exposure via leaching into the water.

Transfer of plastic additives to marine organisms upon plastic ingestion has been demonstrated in laboratory experiments and in field studies (Hermabessiere et al., 2017; Andrady, 2017, 1990). PBDE detected in tissues of marine organisms such as bivalves, fish and sperm whale blubber suggested that transfer from e.g. plastics to organisms occurs. These studies have focused on plasticizers and flame retardants, which may be found at relatively high concentrations (10–15 wt-%) in the plastic material. Other findings, e.g. from Aminot et al. (2020) were that HBCDD leachate from PS foam macrodebris was readily bioaccumulated and well-retained by corals. In order to understand the processes behind bioavailability and bioaccumulation of leached additives, these findings need to be interpreted with model work. Biodynamic modelling to determine bioaccumulation of hydrophobic contaminants through ingestion of plastic particles has been performed for humpback whale (*Megaptera novaeangliae*) (Besseling et al., 2015a) and birds (Besseling et al., 2015b; Herzke et al., 2016). Herzke et al. (2016) found that modeled lipid normalized PCB concentrations in fulmar agreed well with the measured values. By combining information on daily filtered water volume, gut passage time and plastic particle numbers in humpback whale, the calculated plastic concentration in the foraging area compares with data on measured concentrations (Besseling et al., 2015a). This calculation of steady-state MP concentration may also apply to other organisms and can be used to model MP transfer for entire food webs (Diepens and Koelmans, 2018). Tissue or organ concentrations of nanoplastic particles that are possibly transferred beyond the gut system might be modeled in a similar way (Besseling et al., 2019).

Experimental desorption studies, addressing sorbed rather than added chemicals or residuals, indicated a higher transfer of chemicals into biota than into seawater: Bakir et al. (2014) examined the potential for PVC and PE to sorb and desorb four different chemicals, i.e. DDT, phenanthrene, perfluorooctanoic acid (PFOA) and DEHP, in both seawater and under simulated gut conditions, in cold and warmed blooded biota respectively. They found desorption rates to be faster under simulated gut conditions and highest when simulating a warm blooded organism. Compared to desorption rates in seawater, these conditions resulted in rates being 30 times greater, suggesting that there is a possibility of transfer of chemicals into biota. Teuten et al. (2007) examined the uptake and subsequent release of phenanthrene from three types of plastics in comparison to natural sediments. In all cases, sorption to plastics greatly exceeded sorption to two natural sediments. Desorption rates of phenanthrene from the plastics or sediments back into solution spanned several orders of magnitude and were higher for sediments than for plastics. Teuten et al. (2007) estimated that the addition of as little as 1 μg of contaminated polyethylene to a gram of sediment would give a significant increase in phenanthrene accumulation by *A. marina*. The authors conclude that plastics may be important agents in the transport of hydrophobic contaminants to sediment-dwelling organisms, based on the sorption of these contaminants from seawater. However, the studies by Teuten et al. (2007) and Bakir et al. (2014) focused on contaminants sorbed from the seawater, not on residual additives.

Rochman et al. (2014b) found a positive relationship between the abundance of plastic debris and the chemical body burden of higher brominated PBDE congeners (BDE-183 to BDE-209) in fish. This was consistent with previous research examining PBDEs in seabirds from the North Pacific Gyre (Tanaka et al., 2013). Further experimental evidence confirmed that PBDEs could transfer from plastic to organisms upon ingestion (Browne et al., 2013; Rochman et al., 2013). These lines of evidence suggest that brominated PBDEs may be an indicator for the exposure of plastic debris to marine animals. On the other hand, investigations under the EU ECsafeSEAFOOD (2015) programme concluded that the levels of additives such as brominated flame retardants and BPA in tissue were not directly related to the MP body burdens of these animals. This is in agreement with Burns and Boxall (2018) who performed an evaluation of evidence for ingestion and subsequent desorption of contaminants from MP as a significant exposure pathway. The authors concluded that available evidence did not support that uptake of HOCs (e.g. PCBs, PBDE, PAHs) could be attributed to transport into the organisms by MP. Likewise, no measureable PCB amounts were transferred to larvae of Atlantic herring (*Clupea harengus*) upon active tube-feeding of individual larvae with MP particles spiked with PCB-153 (Norland et al., 2021). Within the study period of 24 h, some larvae had excreted all MP particles. In this case, levels of PCBs in the fish were not significantly different from controls.

In the study by Kühn et al. (2020) the transfer of plastic additives, from a marine litter derived MP reference mixture (PTX001) and a marine litter derived PS foam sample, to stomach oils of northern fulmars, was investigated. As discussed in Section 3.3, a total of 15 additives, including plasticizers, antioxidants, UV stabilizers, flame retardants, and preservatives, were identified in the original plastic mixtures. Of these, five substances showed strong leaching from the MP reference mixture into the stomach oil during the experiment, i.e. acetophenone, dibutylphenol, TCEP, DEHP, and bumetrizole (also known as Tinuvin 326). Of the four substances identified in a marine litter-derived PS foam two leached into stomach oil, i.e. acetophenone and phenyl benzoate. Observations by Tanaka et al. (2019) indicated that compounds with log Kow $>$ 5.6 could be retained in plastics during weathering in the ocean until ingestion of the plastics by seabirds, while chemicals with low hydrophobicity have the potential to leach out rapidly into the marine environment. In previous studies, transfer and accumulation of BDE-209, a highly hydrophobic compound with a log Kow of 10, from ingested plastics into seabird tissue was observed for field-caught seabirds (Tanaka et al., 2013; Tanaka et al., 2015). In addition, oily components in a bird's stomach facilitated the leaching of BDE-209 (Tanaka et al., 2015). Similar oil-facilitated leaching could also occur for other hydrophobic additives. Therefore, hydrophobic (log Kow $>$ 5.6) additives and by-products identified in the present study seem to pose a high risk of exposure to seabirds, because they were confirmed to be retained in plastics until ingestion by seabirds, and can be leached out in the stomach.

Beiras et al. (2019) tested the hypothesis of PE as vectors of HOCs to marine organisms (marine plankton (copepod *Acartia clausi*), larva of the *Paracentrotus lividus* sea-urchin), for two model chemicals with similar hydrophobic properties, the 4-n-NP and the 4-methylbenzylidene-camphor used as plastic additive and UV filter in cosmetics, respectively. Both test species actively ingested the MP particles. However, the presence of MP never increased the bioaccumulation of the model chemicals, nor their toxicity to the exposed organisms. Bioaccumulation was a linear function of waterborne chemical disregarding the level of MP. These results are in line with those obtained by Besseling et al. (2017), who performed a similar analysis for lugworm and also found that bioconcentration and bioaccumulation were only driven by aqueous phase concentration and not ingestion of MP. These consistent results challenge the assumption that MP act as vectors of hydrophobic chemicals to planktonic marine organisms.

In summary, modelling studies predict a generally small contribution of plastic-associated chemicals to bioaccumulation. Experimental studies have shown the leaching of hydrophobic compounds in particular into animal stomachs. While the transfer of e.g. plastic-related PBDEs has been demonstrated, other studies have not found a transport of BPA and HOCs (e.g. PCBs, PBDE, PAHs) from MP into organisms. HOCs are the most studied compounds although less bioaccumulating and biomagnifying compounds, such as phenols and benzotriazoles, may also be relevant to study in relation to exposure of many high volume plastic types, such as PE, PP and PET. Overall, the studies on uptake, bioavailability and bioaccumulation of plastic additives do not give a conclusive link between MP body burdens and additive body burdens in marine organisms.

4. Hazard and risk assessments

Risk assessments of plastic debris should separate the risk component of the plastic associated chemicals from that of the plastic material itself (Koelmans et al., 2017), therefore only studies considering hazard and risk assessment of residual chemical additives are considered in this section. Risk assessment of additives can be performed using the chemical risk assessment procedure outlined in ECHA's Technical Guidance Document (EC, 2003). This document provides guidance to a hazard identification, chemical exposure assessment and a hazard assessment, leading to Predicted Environmental Concentrations (PEC) and Predicted No-Effect Concentrations (PNEC), respectively. The risk characterization is based on Risk Quotients (RQ), dividing PEC by PNEC.

A high number of different chemicals are used in plastic production and the fact that producers want to keep the exact composition of their products confidential (industrial secret) prevents a comprehensive overview of plastic additives (Groh et al., 2019). Existing studies therefore focus on monomers and/or a limited number of additives used in high concentrations, such as phthalate plasticizers and various flame retardants. The case of 6PPD and its transformation product 6PPD-quinone, however, demonstrates that substances used in relatively low concentrations can be highly toxic for certain species (Tian et al., 2021).

4.1. Hazard assessments

Lithner et al. (2011) established a hazard ranking of polymers based on the toxicity of their respective monomers (Table 1a). Of 55 examined polymers, 56% consisted of carcinogenic or mutagenic monomers, including PUR, polyacrylonitriles, PVC, epoxy resins, and styrenic copolymers (ABS, SAN and HIPS). They all have a large global annual production, ranging from 1 to 37 million tonnes (Lithner et al., 2011). The most hazardous monomers were 1,3-butadiene, acrylonitrile, epichlorohydrin and vinylchloride. Polymers that were composed of monomers that potentially exert germ cell mutagenicity and acute toxicity and that have a large global annual production of 1–5 million tonnes, are phenol formaldehyde resins, unsaturated polyesters, polycarbonate, polymethyl methacrylate, and urea-formaldehyde resins. PVC should receive extra attention because of its carcinogenic monomer, its high production volume and because it requires the highest amount of additives, which in turn are often hazardous themselves (Lithner et al., 2011).

Hazards from a total of 906 chemicals associated with plastic packaging have recently been classified (Groh et al., 2019), where 63 (7%) were hazardous to human health and 68 (8%) hazardous to the environment according to the EU classification and labelling (CLP/GHS) system of hazardous chemicals (ECHA, 2022). Furthermore, 7 (1%) had been classified as persistent, bioaccumulative, and toxic (PBT), or very persistent, very bioaccumulative (vPvB), and 15 (2%) as endocrine disrupting compounds (EDCs) (Groh et al., 2019). Thirty-four (4%) have also been recognized as EDCs or potential EDCs (UNEP, 2018). The identified hazardous chemicals are used in plastics as monomers, intermediates, solvents, surfactants, plasticizers, stabilizers, biocides, flame retardants, accelerators, colorants and others. Based on the hazard characterization there is clearly a need to perform a more comprehensive risk assessment and use this to investigate the need for alternative chemicals. A risk assessment is hindered by data gaps, e.g. on how the specific chemicals are used and in which quantities. With respect to a hazard assessment many of the substances lack any publicly available hazard data. This missing information should, however, not prevent manufacturers from providing information on the use of chemicals in plastics, including chemicals identity and quantity in plastics articles.

4.2. Risk assessments

Hazard assessments of monomers and plastic additives need to be complemented with effect thresholds to characterize risks arising from plastics containing these compounds in the marine environment. Eventually, these can be combined with an exposure concentration in a risk characterization (Besseling et al., 2019). Beiras et al. (2019) developed a 2-tier standardized protocol that combined the sensitivity of leachate tests with the environmental relevance of particulate phase tests to discriminate between toxic and non-toxic plastic materials. The protocol was used to test the toxicity of MP to marine plankton organisms, i.e. *Paracentrotus lividus* and *Acartia clausi* planktonic larvae, and they applied it to MP obtained both from commercial plastic products (PE and PVC) and from plastics retrieved from the marine environment (PE). They found that additives were responsible for the toxicity found in certain plastic materials, but did not identify specific compounds.

Table 4
Risk assessment studies for the marine environment. Low (RQ < 0.1), moderate (0.1 ≤ RQ < 1) and high (RQ ≥ 1) risks are stated.

Additive	Risk quotient (RQ)	Environmental organism/ compartment	Model/ laboratory/ field study	Reference
Nonylphenol (NP)	Moderate	Water (coastal area)	Field	Salgueiro-Gonzalez et al. (2019)
NP	High	Sediment (coastal area)	Field	Salgueiro-Gonzalez et al. (2019)
4-tOP	Moderate	Sediment (coastal area)	Field	Salgueiro-Gonzalez et al. (2019)
4-NP	Low to high	Coastal ecosystems	Field	Tato et al. (2018)
BPA	Very low	Coastal ecosystems	Field	Tato et al. (2018)
TCS	Very low	Coastal ecosystems	Field	Tato et al. (2018)
OD-PABA	Very low	Three model organisms in a coastal marine ecosystem	Bioassay	Giraldo et al. (2017)
OC	Moderate	Three model organisms in a coastal marine ecosystem	Bioassay	Giraldo et al. (2017)
PeBDE	High	Copepods and cod	Model	Fauser et al. (2020)
TBT	High	Copepods and cod	Model	Fauser et al. (2020)
HBCDD	High	Copepods and cod	Model	Fauser et al. (2020)
PeBDE	High	Fulmar	Model	Fauser et al. (2020)
DEHP	Moderate	Copepods and cod	Model	Fauser et al. (2020)
Cu	Moderate	Copepods and cod	Model	Fauser et al. (2020)
Metals, org. Compounds, monomers	Very low to low	Copepods and cod	Model	Fauser et al. (2020)
6PPD-quinone	High	Coho salmon	Field	Tian et al. (2021)
DEHP	Low	Freshwater	Field	Bolívar-Subirats et al. (2021)
NP	Low	Freshwater	Field	Bolívar-Subirats et al. (2021)
OP	Low	Freshwater	Field	Bolívar-Subirats et al. (2021)

Several studies have determined risk quotients in marine compartments and ecosystems for plastic additives such as NP, with different results. In a study of selected EDCs, i.e. NP, BPA, 4-*tert*-octylphenol (4-tOP), in seawaters and sediments from two coastal areas in Spain, risk quotients were calculated in a worst-case scenario where maximum detected concentrations were applied (Salgueiro-Gonzalez et al., 2019). Risk quotients for seawater showed a moderate risk posed by NP, and for sediments moderate risks related to 4-tOP and a high risk of NP were estimated. Also studying 4-NP and BPA, in addition to triclosan (TCS), Tato et al. (2018) calculated risk quotients indicating very low or low risks for BPA and TCS, and low to high risks for 4-NP in coastal ecosystems. Thus, the risk estimations for 4-NP reached different conclusions where the slightly higher risk detected in the study of Salgueiro-Gonzalez et al. (2019) could originate from the presence of wastewater treatment effluents, and nautical, agricultural and industrial activities in the area combined with the worst-case approach in the selection of measured concentrations.

There has been a significant increase in the use of UV filters in cosmetics and as plastic additives during the last few decades. Sunscreens may contain up to 10 wt-% of organic UV filters such as ethylhexyl dimethyl p-aminobenzoic acid (OD-PABA) and octocrylene (OC). Giraldo et al. (2017) investigated their toxicity by performing ecotoxicological bioassays with three model organisms from different trophic levels in a coastal marine ecosystem, i.e. microalga *Isochrysis galbana*, the sea urchin *Paracentrotus lividus*, and the mussel *Mytilus galloprovincialis*. The calculated RQs were 0.27 for OC and 0.007 for OD-PABA. OC presents a moderate environmental risk ($0.1 < RQ < 1$) leaving a safety margin of approx. a factor four, which is expected to increase during the bathing season in the summer. Derivation of environmental quality standard values should be provided in order to refine the risk estimates and use them in a regulatory context.

In a theoretical study the risk of 30 additives in eight polymer types, i.e. PUR, PVC, PE, PP, acrylics, SBR, EPS and cellulose acetate (cigarette butts), was investigated for three marine organisms, representing three trophic levels, i.e. pelagic/planktonic zooplankton: copepod, benthopelagic fish: Atlantic cod and seabird: northern fulmar (Fauser et al., 2020). The investigated additives were e.g. flame retardants, softeners, biocides and metals used as pigments, which all have Environmental Quality Standards (EQS) (European Directive 39/2013/EU) as toxicity threshold values. RQs > 1 were found for copepods and cod and the flame-retardant PentaBDE used in PUR, the biocide tributyltin (TBT) present as impurity in PVC and PUR, and HBCDD used in EPS. A $RQ > 1$ was found for fulmar and PentaBDE used in PUR. Approx. 70 additives were identified as potentially present in the investigated polymers, which suggests that EQS values should be derived for the compounds that have largest uses and hazards, in order to obtain a more complete estimate of risk.

Tian et al. (2021) investigated the incidences of unexplained acute mortality when adult coho salmon (*Oncorhynchus kisutch*) migrated to urban creeks to reproduce in the U.S. Pacific Northwest. Retrospective analysis of representative roadway runoff and stormwater-affected creeks of the U.S. West Coast indicated widespread occurrence of 6PPD-quinone, a highly toxic quinone transformation product of the globally ubiquitous tire rubber antioxidant N-(1,3-dimethylbutyl)-*N*′-phenyl-*p*-phenylenediamine (6PPD). Concentrations of 6PPD-quinone were at toxic levels, i.e. $RQ > 1$. The study concludes that 6PPD-quinone is the primary causal toxicant of stormwater-linked coho salmon acute mortality observations. This is an example of the formation of a more toxic and mobile compound (6PPD-quinone) from the transformation of a rubber additive (6PPD), which increases environmental risks. On the basis of the ubiquitous use and substantial mass fraction (0.4 to 2%) of 6PPD in tire rubbers, 6PPD-quinone may be present broadly in stormwater and roadway run-off at toxicologically relevant concentrations for sensitive species, such as coho salmon.

For the freshwater environment, Bolívar-Subirats et al. (2021) identified low risks of DEHP, NP and OP in downstream waters in the Besos river basin in Spain. The study included five phthalates, their substitutes acetyl tributyl citrate (ATBC) and DEHA, 12 long and short chain alkylphenols, BPA and benzophenone. Their RQs in river water were calculated by relating their measured environmental concentrations to EU-set EQS values (European Directive 39/2013/EU) as well as to EC50 values based on tests with the planktonic crustacean *Daphnia magna*, which is commonly used for environmental monitoring of pollutants. Although this study did not address the marine environment, it can indicate potential risks as the studied polymers and high volume plastic additives can also be expected in the marine environment.

In summary, only few studies exist that have conducted a risk assessment of chemical residuals in MP in the marine environment (Table 4). The existing results vary between low ($RQ < 0.1$), moderate ($0.1 \leq RQ < 1$) and high ($RQ \geq 1$) risks of specific MP-associated compounds and are not necessarily consistent for the same compound. For example both low, moderate and high risks have been identified for NP. These variations in RQs could be caused by nearby sources, such as wastewater treatment effluents and industrial activities, and the choice of minimum, mean or maximum concentration levels in the calculation of RQs. The sources of additives resulting in the risk estimates in Table 4 are diverse, including cosmetic products, various plastic polymers in different applications, and car tires.

5. Conclusions and outlook

The presence of large amounts of plastics in the marine environment, which are eventually broken down to MP, goes along with the introduction of a variety of chemicals into the marine environment, including chemical additives, unreacted monomers, and degradation products. Some of these residual additives have been identified as (potentially) hazardous, but only a few have been evaluated for their risk to marine organisms, e.g. using conventional risk assessment approaches (EC, 2003).

The determination of a PEC is particularly complex in the context of MP-associated chemicals. The PEC is a result of the amount of MP particles in the sea, the initial concentration of a specific chemical in the plastic material and its change during weathering, and potential leaching processes. Furthermore, the situation of direct exposure of an organism after uptake of a plastic particle is different from the indirect exposure via seawater. The latter case obviously coincides with the exposure to the same chemical from other emission sources than MP particles, as well as with uptake of the same chemicals with food. Distinguishing between different exposure pathways is difficult in field studies and will likely require defined laboratory conditions. Attempts to link levels of chemicals used as plastic additives in animal tissues to the presence of plastic particles in the animal's stomach have often been inconclusive, so the significance of ingested plastic particles as a source of exposure to chemicals has not been fully understood.

Monitoring initiatives are in place or being developed for litter, including plastics, in the marine environment, but MP monitoring is still hampered by technical challenges and the large fluctuation of MP concentrations in the marine environment. Consequently, the real exposure to MP particles remains difficult to estimate for marine organisms. Data quality and quantity regarding MP exposure (and effects) were identified as the main shortcoming in developments of MP risk assessment frameworks, even without considering additional complexity due to chemicals (Gouin et al., 2019). Data quality in particular was regarded as insufficient to evaluate impacts of MPs. Data quality can be improved by harmonization and standardization of methods used for the sampling and analysis of plastic particles and associated additive residuals, as well as developing standardized protocols for leaching and toxicity testing, reporting and quality assurance measures.

Knowledge about additives in MP is increasing, via chemical analyses and compilation of data available from manufacturers. In chemical analyses of MP, non-target and suspect screening approaches will be a central element in identifying the multitude of additives. However, as

their purpose mainly is the identification of chemicals, quantification, as needed for PEC determinations, currently still requires optimized target analyses, especially as PECs should be determined with high precision and accuracy in order to obtain accurate risk estimates. Although tedious, this is technically possible. Furthermore, if data quality is poor for MP, this will cascade down to relatively high uncertainty for PECs of MP additives and questionable applicability in risk assessments.

Quantification of leaching of chemicals to the surrounding environment and/or the gastrointestinal tract of an organism is a field of ongoing research, both with and without consideration of weathering processes affecting the structure and composition of plastic particles. The available results are inconsistent, with some studies showing increased bioaccumulation of plastic-associated chemicals in marine organisms after uptake of MPs, while other studies did not show bioaccumulation, possibly related to several reasons, among these the ability of plastic particles to act as a passive sampler (Herzke et al., 2016). A better understanding of leaching and uptake processes and the variability in e.g. bioaccumulation between species and compounds is needed to improve the risk assessment of plastic additives.

Given the large number of plastic additives that marine organisms can potentially be exposed to, a discussion of PNECs and effects on marine organisms go beyond the scope of this paper, not least because it will be vital to consider mixture effects, due to the commercial use of mixtures as additives and the co-occurrence of chemicals in the environment from different sources. Effect-based methods, i.e. starting from an identification of an effect rather than from an identification of a chemical, may be a promising way forward, combined with non-target and suspect screening approaches to identify groups of specific chemicals causing or contributing to an effect. However, the quantitative aspect of a traditional risk assessment will remain a challenge.

Considering these challenges and uncertainties, the question may arise whether or not a classical risk assessment approach is useful and feasible in evaluating risk from chemicals associated with plastics. This question was also discussed in the debate between Backhaus and Wagner (2020), in the broader context of the risk of MP particles. The precautionary principle may call for actions even though scientific evidence of risks is sparse. The authors discuss that the classical risk assessment approach, as described by risk quotients resulting from PEC and PNEC values, might be too narrow to address an issue as complex and widespread as global plastic pollution. It might even prevent us from developing more environmentally realistic risk paradigms, which could be achieved by considering the omnipresence, persistence and uptake potential of MP and their associated additives (Backhaus and Wagner, 2020).

While toxicity evidence of MPs and associated chemicals in marine organisms is sparse, deteriorating effects of entanglement and ingestion of meso- and macroplastic and other litter items are well-documented (Kühn and van Franeker, 2020).Together with impacts on e.g. tourism, consensus has been created on a need to address the overall problem of plastic pollution, as reflected in regional action plans (e.g. OSPAR, 2014). Research and monitoring have to work toward higher quality data in both occurrence and toxicity of MPs and their associated chemicals as well as a better qualitative and quantitative understanding of the processes involved, in order to address questions of risks. Finally, the complex situation of MPs and MP additives should also encourage scientists and regulators to explore new approaches in risk assessment, for example with a focus on occurrence and fate of plastic together with the environmental persistence of chemicals, possibly in combination with their bioactive potential.

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Declaration of competing interest

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Appendix A. Supplementary data

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