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## Plastic additives and microplastics as emerging contaminants: Mechanisms and analytical assessment



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### ABSTRACT

Plastic additives comprise a plethora of substances that serve numerous purposes in the plastic industry. These can be used to assist the molding of plastics and can contribute to providing optimal performance to the material when molded and used, or, simply, to reduce costs. However, these additives, and non-polymerized monomers, far more hazardous than their poly-counterparts, may be released throughout the entire life cycle of plastics, posing risks to the environment and, ultimately, human health. Hence, many studies have delved into the mechanistic uptake/release of these compounds. Herein, we briefly overview the current knowledge on the underlying processes affecting these mechanisms. We also outline the potential ecological consequences of these hazardous substances and explore some of the prevailing analytical methodologies used for their determination in both environment and laboratory studies. Furthermore, we highlight the key limitations of the currently available literature and present a prospective outlook for future research.

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## 1. Introduction

### 1.1. Plastic pollution, use, and life cycle

Plastics encompass a wide variety of synthetic or semi-synthetic materials that, owing to their plasticity, can be molded, extruded or pressed into solid objects of a nearly endless multitude of shapes. Global plastic production reached 367 million tons in 2020, a figure that does not account for the production of recycled plastics [24]. Overall, it is estimated that plastic production reached the 400 million tons mark in that year [45].

These materials may be categorized according to multiple classifications, such as those based on the polymer's backbone and side-chain (e.g., acrylics, polyesters, silicones, polyurethanes), the chemical process used in their manufacture (e.g., condensation, polyaddition, and cross-linking), their physical properties (e.g.,

density, hardness, tensile strength, thermal resistance, and glass transition temperature) or manufacturing characteristics and/or product purpose, as thermoplastics (plastics that do not undergo chemical change in their composition when heated and can thus be molded repeatedly, as polyethylene), thermosets (plastics that can melt and take shape only once and that, if reheated, decompose rather than melt, as is the case with rubber), conductive polymers (e.g., polyaniline), biodegradable plastics (e.g., polylactic acid (PLA)) and engineered plastics [25].

The life cycle of plastics, summarized in Fig. 1, initiates with the extraction of raw materials, and continues through design and production, followed by packaging and distribution, use and maintenance and, lastly, recycling, reuse, recovery or final disposal [52]. After usage and/or consumption, plastic materials may follow several routes. Some, for example, are collected and sorted through formal or informal waste management schemes [45] or by manufacturers. They may be transformed into plastic pellets or flakes, and thus re-enter the production and use phases. However, most of these plastics are incinerated or openly burned, with severe environmental and health impacts [2], disposed of in landfills/dumpsites, or escape into the environment [30]. Indeed, it is estimated

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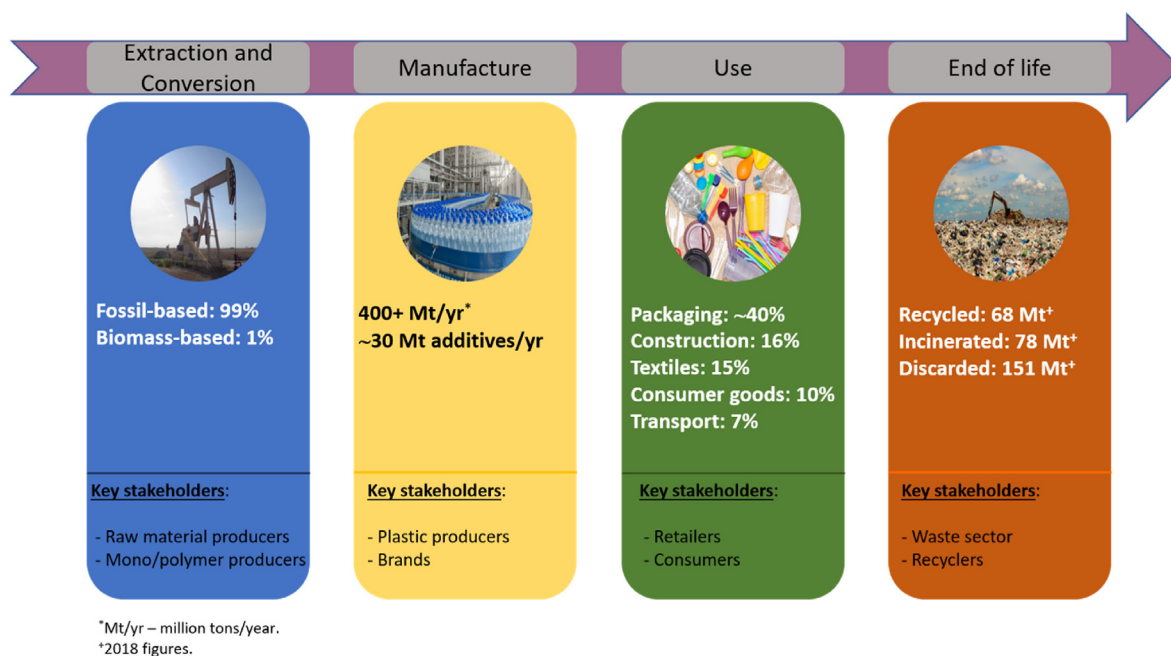


Fig. 1. The plastics life-cycle. Some of the key stakeholders at each stage are identified. Adapted from Ref. [45].

that, of all the plastic produced since the industrial manufacture of plastics initiated in the early 1950s, less than 10% has been recycled or re-used, with concomitant economic and environmental impacts, due to loss of resources and removal from the value chain. Additionally, a significant proportion of recycled plastics are downcycled, meaning these are used within a “recycling path with low requirements and low-quality end-products” [67], reducing or eliminating the possibility of multi-cycles of recycling, though this may be, in developing countries, a key aspect in the future of plastic recycling, given its potential for local economies.

But plastics may escape into the environment not only during their end of life, but at every stage of their life cycle [76]. During production, transport and/or conversion, plastic pellets (“nurdles”), as well as fibers, may be lost. However, the main leakage of plastics into the environment takes place following their use and during disposal, most of which because of littering and poor or insufficient waste management practices. It has been suggested that between 19 and 23 million tons of plastic waste entered aquatic ecosystems from land-based sources alone in 2016 [12]. Assuming the projected proportion of land-based and marine-sourced plastic waste (70–80% to 20–30% by weight, respectively [61]), stemming from plastic nets and fishing lines, for example, then, in 2016, conservatively, 24 to 27 million tons of plastic waste entered the marine environment in that year. For these reasons, plastic contamination is ubiquitous, having been described from the arctic deep seafloor [11] to remote high-altitude atmosphere [3].

### 1.2. A matter of size – microplastics

In the marine environment, over three-quarters of the mass of plastic waste consists of debris larger than 5 cm, with at least 46% being comprised of fishing nets [38]. However, although pieces smaller than 0.5 cm only account for 8% of the total mass, they correspond to over 94% of the estimated 1.8 (1.1–3.6) trillion pieces floating in the area [21]. These smaller particles, frequently considered as particles <5 mm, are known as microplastics [20]. Also colloquially referred to as “mermaid’s tears”, due to their size and a vast array of colors, microplastics may be defined as primary

or secondary, depending on their source, and they exist in many forms, including fibers (or microfibers), spheres, pellets (“nurdles”), foams, films and fragments [20].

Primary microplastics are deliberately manufactured within the millimetric or sub-millimetric size and can be found in various household items, ranging from exfoliators to toothpaste and cleaning products. Pre-production plastic pellets, used as feedstock in the manufacture of plastic products, are an example of primary microplastics. These small pellets are frequently lost during handling or due to spillages during transportation or at production facilities [47]. Microplastics may also be directly discharged in domestic and drainage wastewaters, reaching the (marine) environment [6]. It may be possible to identify the sources of primary microplastics and, therefore, mitigation actions to reduce their input into the environment may be, theoretically, implemented. Nonetheless, the vast majority of microplastics in the environment – up to 2/3 – are secondary microplastics. These result from the breakdown of larger plastic particles [37]. When exposed to the elements, these larger plastic particles are subjected to physical, chemical and biological processes that lead to the reduction or loss of the structural integrity of these plastics, resulting in their fragmentation [22]. Ubiquitous in the environment, microplastics have been found in all spheres of the environment, namely, soil, water and air, as well as in food and water [57]. This degradation/fragmentation process can result in multiple-sized particles – meso, micro and nano – and they can further release dissolve organic matter and the additives they contain [40].

Researchers have thus recently begun to use a new classification of these, plasticides [21]. These additives, which are usually of low molecular weight and not bound to the polymers, may elicit biochemical responses and effects. It should also be noted that, to a lower extent, residual monomers, that may be present due to incomplete polymerization reactions, can also migrate, similarly to other adsorbed organic pollutants, and these frequently exhibit a considerable degree of toxicity [36]. As most of these substances are lipophilic, they have an inherent affinity for cell membranes, rendering them potentially hazardous, as they may transverse membranes and actively participate or interfere in biochemical

reactions [56].

The risks of these substances become of even greater concern when associated with increasingly smaller particles, such as microplastics, owing to their high surface area-to-volume ratios. As such, microplastics containing additives and/or residual non-polymerized monomers may constitute new sources of exposure to chemical compounds, particularly when ingested. However, this will depend on the physical-chemical properties of the matrix and the ecosystem compartments they will be present. Some studies have highlighted that the observed ecotoxicity of microplastics in water was mainly due to the release of additives, rather than to the ingestion of the particles themselves (e.g. Ref. [10]), and additive release might have a bigger impact on plastic ecotoxicity than initially thought [82]. Contrarily, nonetheless, it has also been stated that the characteristics of both plastics and additives may render the former *de facto* sinks for not only additives but also other organic pollutants present in the environment [42]. These issues highlight the intrinsic difficulties in assessing the risk linked to the presence of additives in environmentally-contaminating polymers, also because a large part of the used additives have unknown hazardous effects. Further, many additives are not solely used for polymeric function enhancement, but in other engineering and industrial processes as well, and the source of origin of a given pollutant in ecosystems remains difficult to assess. Finally, additives released in various environmental/biological compartments will be dictated by their migration and desorption from the polymers, which would be dependent on the additive, polymer, and the matrix physical-chemical conditions [79], as described in Section 3.

## 2. Plastic additives

Plastics are not solely comprised of polymers, they also contain additives that enhance plastic functions by modulating their physical and chemical properties. Indeed, every plastic item contains additives that determine the properties of the material and influence its cost of production [45]. In 2020, the global plastic additives market was USD 48.41 billion and is expected to grow from \$51.04 billion in 2021 to \$75.20 billion in 2028, at an average annual growth rate of 5.7% during 2021–2028 [27]. Estimating absolute quantities, levels and types of additives used in different plastic products is inherently difficult, owing to the limited disclosure by manufacturers, often driven by proprietary concerns, with subsequent increasing challenges in the identifying and quantifying additives present and potential consequences for the environment.

These plastic additives are numerous in chemistry and nature and serve many functions. They can be found as traces (<1% w/w) or as major components (>50% w/w) of these materials and are usually embedded into the polymer structure. Table S1 lists some of the most used additives, as well as their function and the polymers to which they are typically added. Multiple compounds may be added to plastics, depending on their purpose. Van Oers and colleagues, for example, stated that any randomly chosen plastic product contains about 20 additives [53], which highlights the widespread use of these chemicals.

Owing to the fact that these additives are weakly bound to the plastics, they may leach during use or when disposed of. Additionally, additives themselves, still associated with the plastic products or following their leaching, could degrade and form other toxic compounds, potentially persisting in the environment and (bio)accumulating in biota (e.g. Ref. [55]). The potential hazards associated with additives continue throughout the life-cycle of plastics. Because they are difficult to remove, when plastics are recycled it is highly likely that these compounds will be integrated into the newly manufactured product [77]. And, because many

industrial manufacturers are not fully transparent regarding the used additives and their concentrations (Table S1), a detailed view across the value chain of the chemical profile of the final products is not available, as well as concomitant health risks [45]. For example, brominated flame retardants, recognized endocrine disruptors, have been incorporated into recycled products, including household items [58] and toys [48], posing a significant health risk.

These health risks are not minimized when plastic waste is burned, which is particularly problematic when using low-technology incinerators or under uncontrolled conditions. When combustion is incomplete, the process causes the emission of hazardous compounds, including persistent organic pollutants (POPs), namely, dioxins, as well as acid gases and contaminated ashes [21]. Hence, the risk for ecosystems and biota linked to the presence of additives in microplastics is associated with their inherent hazards and exposure to and within the ecosystem. Additives identified as hazardous to humans, the environment and organisms are regulated internationally through initiatives like the Stockholm Convention. This legally binding agreement adopted in 2001 and entered into force in 2004 is a global treaty that requires parties to take active measures to reduce or eliminate the release of POPs into the environment [21]. The text focuses specifically on compounds that persist in the environment for long periods, that are widely geographically distributed and that are susceptible of accumulating in the fatty tissues of organisms with deleterious effects on health and the environment. Despite its generally broad encompassing provisions, the Convention only regulates a small fraction of the hazardous chemical compounds contained in plastics and plastic waste and some of these chemical compounds remain in use owing to current exemptions [45], though the vast majority of the ever-increasing list of additives still does not fall under the existing regulations. Yet, while the toxicity of the additives contained in (micro)plastics towards biota has been reviewed (see section 4 below), their exposure remains poorly understood. To assess biota exposure, in-depth comprehension of the mechanisms happening at the plastic-biota-environment interface driving additives released is needed, and, as such, more research is needed.

Herein, we provide an overview of the underlying migration mechanisms of these contaminants, with a special focus on microplastics, and how these materials could act as *de facto* vectors for these additives and organic pollutants. The potential ecological consequences of these classes of hazardous substances are also addressed, as are the analytical methodologies used in the quantification of such contaminants in environmental matrices. However, in spite of the increasing concern regarding this class of environmental contaminants, much remains unknown and, as such, current knowledge gaps are identified and a prospective research outlook for the future is presented.

## 3. The mechanistic leaching of additives

### 3.1. Sources of contamination

The types of additives occurring in the environment, whether in isolation or associated with (micro)plastics, are highly dependent on their source [69]. Although most of these contaminants have been found away from their sources (e. g., in marine ecosystems [8]), assessing these same sources is extremely difficult, as most of them are not used solely as additives, but also in a wide range of applications (see Table S1). For example, bisphenol A is not only used in the production of plastic bottles, but is also a key ingredient in the manufacture of thermal paper and food can lining, among others [66]. Classified as a xenoestrogen, exhibiting hormone-like properties that mimic the effects of estrogen in the body, bisphenol A has been found in environmental samples from seawater,

sediments and atmosphere, where its presence has been associated to plastic burning [29]. Similarly, perfluorinated compounds (PFCs), which display water and oil resistant properties and that are used in food packaging and textiles, for example, and that are persistent in the environment, have also been described in water, sediment, soil and biota [51], as well as in the atmosphere [41]. As such, particularly when associated with microplastics, it is not difficult to envision a continuous “cycle” of these compounds in the environment, especially when considering that the transfer of plastic particles at the water-air interface has been experimentally demonstrated by Ref. [46], namely, through bubble bursting. Bringer and colleagues [14], for instance, characterized (micro) plastics and associated additives along the French Atlantic coast, at the beach, surface water and coastal (semi-depth) sediments levels, and found that around 70% of all identified materials and compounds (e.g., polycyclic aromatic hydrocarbons (PAHs) and naphthalene, among others) stemmed from aquaculture activities. In Fig. 2, the major sources and sinks of microplastics, as well as associated chemicals, are summarized. These do not constitute an exhaustive list, but do underscore the varied nature of these sources and how such materials and compounds may accumulate and distribute in the environment.

### 3.2. Migration and sorption

Chemical substances added to plastics hold the potential to migrate from the plastic material to the surrounding matrix (food, water, soil, or others), but also within the plastic itself, to its surface [31]. In some cases, this migration is engineered and desirable, according to the function of said material. One such example is the gradual and continuous release of mold release agents to the surface that results in improved antistatic, mechanical or optical properties (e.g. Refs. [16,19]) or preventing the oxidation of silver, extending food preservation [71]. In most cases, however, this release of additives stems from the uncontrolled and unplanned migration of these chemical compounds that may have profound health and environmental consequences.

Generally, the migration can be divided into four steps: 1) diffusion through the polymer; 2) desorption (from the polymer surface); 3) sorption at the plastic-matrix interface and 4) absorption/dispersion in the matrix [31].

Overall, the migration process is ruled by Fick's Law, which, simply put, states that the “rate of diffusion of a substance across [an] unit area (such as a surface) is proportional to the concentration gradient” [23]. In other words, it postulates that a flux of a given compound occurs from regions of high concentration to regions of low concentration and that the magnitude of said migration is proportional to the concentration gradient (spatial derivative) [17]. It can be written under various forms, but the most common version is based on molarity (Equation (1)):

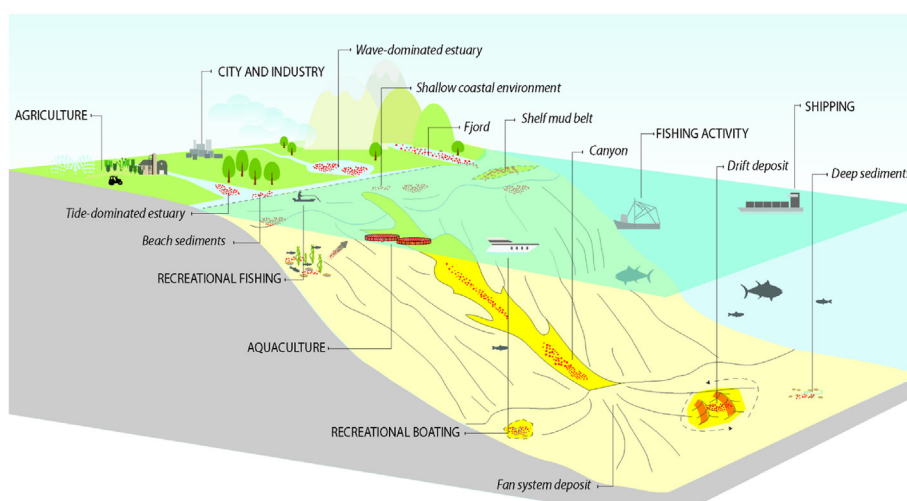
$$J = -D \frac{d\varphi}{dx} \quad (\text{Equation 1})$$

where.

- $J$  is the diffusion flux. It refers to the amount of substance per unit area per unit time. This parameter measures the amount of substance that flows through a unit area during a unit time interval.
- $D$  is the diffusion coefficient. It is expressed in area per unit time.
- $\varphi$  is concentration, expressed as the amount of substance per unit volume.
- $x$  is position, the dimension of which is length.

The rate of migration of organic chemical substances, such as plastic additives, is size-dependent. Usually, smaller molecules, as monomers, with lower boiling points, will show a tendency to migrate faster [31]. Monomers such as formaldehyde, ethylene, butadiene or vinyl chloride will migrate faster even at ambient temperatures.

Sorption, in turn, refers to the process of transfer of chemical compounds between phases: a fluid phase, as air or water, and a solid phase, as plastics. The term encompasses both absorption and adsorption. In the case of the former, the molecules of the chemical compound penetrate and become embedded in the matrix of the solid phase; in adsorption, the molecules of the chemical compound remain on the interface between the fluid and the solid phases [64]. The process of adsorption commonly involves van der Waals, ionic, steric or covalent bonding interactions, while absorption occurs by the partitioning of the sorbate molecules into the sorbent matrix and is held by weak van der Waals forces [28].



**Fig. 2.** Major sources and sinks of microplastics. Most sources of (micro)plastic pollution are land-based and include agriculture, construction, transportation and a wide variety of personal care, pharmaceutical, healthcare and hygiene products. Maritime equipment, as well as fisheries and aquaculture, shipping, offshore operations and ship-based tourism are also important sources. Sources are indicated in capital letters, whilst sinks are denoted in italic lettering. Image credit: GRID-Arendal, available at <https://www.grida.no/resources/15042>.

This process is highly dependent on the (hydrophobic) properties of the sorbate chemical compound, properties of the solid phase and surface area-to-volume ratio, which increases exponentially with decreasing particle size, as is the case for microplastics [21]. For these materials, their surface polarity determines the extent to which an organic contaminant interacts. For example, hydrophobic organic contaminants preferably adhere to non-polar surfaces [50]. Indeed, the environmental hazards of these additives when associated with plastic particles, namely, microplastics, stem from the fact that many, such as PAHs, are apolar planar molecules that exhibit adsorption coefficients commonly higher than those of non-planar molecules of identical hydrophobicity [28]. When the concentration of the sorbate (chemical compound) is low, there is a higher degree of partitioning of the organic compound between both fluid and solid phases, owing to stronger forces of interaction at the surface. Hence, generally, low concentrations of sorbate favor adsorption and high concentrations favor absorption [32]. The sorption process can be further classified as physisorption (physical adsorption) or chemisorption (chemical adsorption). Under physisorption, the molecular interactions between the adsorbate molecules and the adsorbent (solid phase) are primarily governed by van der Waals forces [1], whilst chemisorption involves a chemical reaction between the surface and the adsorbate, during which new chemical bonds are generated at the surface of the adsorbent [54]. Chemisorption may therefore be irreversible, except when the covalent bonds are broken, making desorption of the chemical compounds from the solid phase an inherently difficult process. Physisorption, on the other hand, is non-specific in nature and is generally regarded as a weak and reversible process. For example, as mentioned by Ref. [1], Atugoda and colleagues, when studying the interaction between the antibiotic ciprofloxacin and PE microplastics, observed that the adsorbate molecules accumulated and adsorbed onto the heterogeneous polymeric surface the PE microparticles through weak van der Waal's forces and electrostatic interactions [7]. Thus, in this specific case, the adsorption mechanism was primarily governed by physisorption accompanied by partitioning into the porous surface layer of the microplastics.

Multiple factors and parameters relative to the particles (microplastics) and contaminants affect the sorption mechanistic processes. Regarding the former, features such as morphological characteristics – size, shape and color, for example – have been found to play a role in the sorption of contaminants by plastics. Darker plastics typically have higher contents of additives, and, subsequently, their sorption capacity may be enhanced when compared to lighter plastics [50]; similarly, smaller and more irregularly shaped particles display higher surface area-to-volume ratios, resulting in higher rates of sorption [20]. Other particle parameters described as having an effect in the overall rate of sorption of contaminants include crystallinity, glass transition temperature, surface chemistry, polymer functionalization and degree of weathering, all explored in detail elsewhere (e.g. Refs. [28,50]).

At the sorbate level, hydrophobicity and planarity of the contaminant's molecules, as well as its functionalization and ionization, have been demonstrated to have an impact in the overall sorption process, factors that are most closely associated to the interactions, spatial and electrostatic, between sorbate and particle [32].

Matrix characteristics also play a role in sorption mechanisms. Variables such as salinity, pH and ionic strength, temperature and presence of organic matter and/or other contaminants have all been shown to, to different extents, affect the rate or extent of sorption between (micro)plastics and sorbates. Again, these interplays have mostly been explained based on competitive and electrostatic interactions, and detailed reviews on the topic are readily available ([28,32,50]).

#### 4. Limitations and environmental implications

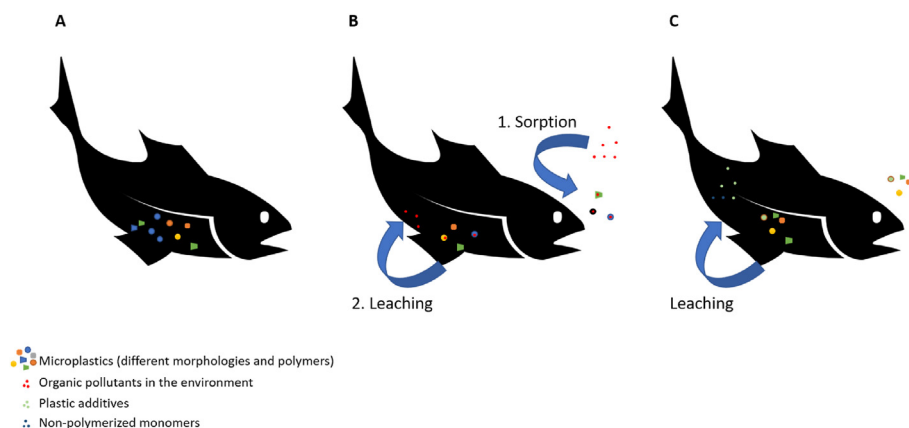
Numerous organisms have been found to ingest (micro)plastics, ranging from zooplankton (e.g. Ref. [72]) to whales (e.g. Ref. [49]). Moreover, these particles are also routinely described in commercially important species. In soils, ingestion of microplastics has also been described (e.g. Ref. [63]) and the *de facto* trophic transfer of these polymeric contaminants has also been demonstrated, for example, from blue mussels (*Mytilus edulis*) to green crab (*Carcinus maenas*) [26]. However, the putative effects of this co-contamination remain, largely, hypothetical, as it is inherently difficult to clarify how frequently and to what extent these phenomena occur in the natural environment. Nevertheless, with an estimated 25 trillion macro- and 51 trillion microplastics – i.e., 500 times the estimated number of stars in the Milky Way – presently littering the oceans [21], as well as with the ever-increasing number and quantities of plastic additives used in the plastic industry and the accumulation of organic pollutants in the environment, this remains an issue of emerging and urgent concern that warrants extensive research. But, as highlighted by Barrick and colleagues, one essential challenge is assessing the risk of chemical additives and discerning their impacts from those of ambient contaminants subsequently accumulated in the polymeric materials [9] and how to best study these impacts. Broadly, such impacts may be divided into three tiers, as summarized in Fig. 3.

- 1) Physical effects of the plastic particles (includes mechanical and nutrient displacement);
- 2) Sorption of ambient organic and metal contaminants;
- 3) Sorption of monomers and chemical additives incorporated into plastic.

Despite these more recent environmentally relevant studies, field and laboratory studies continue to differ drastically. In the laboratory, experimental conditions are determined and tightly controlled, while in the environment, conditions can vary considerably, including sorbate concentration – frequently much lower than those tested in laboratory conditions – temperature and media composition [75]. As noted by Rochman and colleagues, one key difference in field and laboratory settings is the time for reaching equilibrium [62]. Whilst sorbate equilibrium may be reached in laboratory experiments in minutes or hours, and, sometimes, days, which may be attributed to the usual use of constant agitation in the laboratory, as well on the use of significantly higher concentrations of sorbate, and on the uniform composition of the test media, the equilibrium time of sorbate in the environment may take months or even longer.

Furthermore, laboratory tests are typically designed to assess the behavior of one compound, but, in the environment, multiple compounds co-exist, and the ensuing potential competition between sorbates, as well as biofilm formation, microbial activity and polymer degradation, are additional confounding factors that are not accounted for in the laboratory. Yet, despite these limitations, some authors have successfully fitted sorption isotherms and kinetics equations to samples collected from the field [78], suggesting that, although not all variables may not be accounted for, the currently developed methods to predict sorbate-microplastic interactions may still be valid under field conditions.

The physical effects of microplastics and their ecotoxicological consequences are continuously being explored and their impacts on various faunal species have been extensively researched. Nonetheless, very limited knowledge on the leaching of plastic additives from these polymeric materials exists. Hence, data on the leachability and ecotoxicity of the additives associated with microplastics occurring in the environment remains limited and



**Fig. 3.** Overview of the most prominent pathways of microplastic toxicity. In A), obstruction due to physical uptake; false satiety is a common effect. In B), sorption of chemical compounds (and metals) and potential subsequent leaching; in C), ingestion followed by monomers and plastic additives leaching.

subsequent ecological impacts of different types of microplastics associated with their additives, as well as the basis for said toxicity, are yet to be fully explored [69]. Jang and colleagues initially explored the role of Styrofoam as a source of hazardous additives for different marine organisms and found that this material could, indeed, constitute a potential source of hexabromocyclododecane (HBCD) intake for mussels [34]. The authors later not only confirmed this supposition but also observed significant ingestion of these materials and concomitant bioaccumulation of HBCD [33]. The toxicity of microplastics and their lixivates on planktonic organisms was assessed by Ref. [10] and the authors noted that the different microplastic particles – PA, PE, PVC – were not toxic to these organisms *per se*, but chemical compounding with their additives, in the form of lixivates, rendered them toxic, affecting reproduction, survival and population growth. The complexity of the interaction between environmentally occurring chemical compounds and microplastics on fauna was further evidenced by Ref. [4], whom noted that ingested polyethylene microplastics could result in an inhibition in eliminating perfluoroalkyl substances (PFASs) from *M. galloprovincialis*' system. Conversely, the authors also found that, under similar conditions, a reduction in the accumulation of pesticides and personal care products was observed. This complexity can, nonetheless, be further compounded by considering additional factors, such as weathering. Indeed, concurring reports by Refs. [44,80] described an increased rate of adsorption of weathered microplastics for different additives when compared to that of pristine microplastics. Oppositely, however, Lee and co-workers estimated the microplastic-bound intake of hydrophobic organic chemical compounds by fish using measured desorption rates to artificial gut fluid and concluded that the overall uptake of these HOCs, present in equilibrium with seawater would be limited, even for highly hydrophobic chemical compounds. Moreover, they determined that the steady-state bioaccumulation factor was predicted to decrease with increasing ingestion of microplastics, owing to dilution effects [39]. By increasing the complexity of the system, namely, by considering the contribution of the often transient nature of exposure, as well as the different compartments of said system – e.g., sediments, animal activity, currents, among others – the effects of exposure to microplastics could result in a decreased toxicity for some chemical compounds, as demonstrated for media of different complexities [81]. In fact, in spite of the prevailing paradigm that microplastics are effective vectors for the uptake of hazardous organic contaminants by marine biota, HOC microplastic-water partitioning can generally be assumed to be at equilibrium for most microplastics

and chemical compounds occurring in the oceans. Consequently, as modeled by Koelmans and colleagues, the fraction of total HOC sorbed by plastics is comparatively small when compared to that sorbed by other media in the ocean and, therefore, the overall flux of bioaccumulated HOCs stems, overwhelmingly, from natural prey and not from ingested microplastics [35].

## 5. Analytical determination of plastic additives

In order to ascertain the toxicity of these additives, and how their leaching from plastics may cause harm (Fig. 3), chemical identity and exposure to additives must be established. However, experimentally obtained data is scarce, although there is some ample knowledge on additive release from plastics in the food, pharmaceutical and medical, and even construction industries [13]. Such analyses are conducted through extraction and leaching studies. Whilst the former allows for the identification and quantification of additives within a sample, thus identifying the possible hazards associated with the material, the latter enables an assessment of the associated risk, through the estimation of its physical availability, and, consequently, exposure and dose. Hence, extraction studies typically involve isolating the additives by resorting to harsh conditions, namely, strong organic solvents and elevated temperatures for extended periods, to ensure the exhaustive recovery of the broadest range of chemical compounds (e.g. Ref. [73]); on the other hand, leaching analyses are performed under milder conditions that attempt to mimic realistic conditions of exposure (e.g. Ref. [82]). Extractables and leachables, therefore, constitute unique sets of chemical compounds whose presence and concentration may vary greatly, even within identical samples, and that represent distinct risks and hazards for the environment. These studies vary widely, depending on the origin of the sample, environmental matrix, exposure pathways considered and purpose of the study. Detailed works focused on the methodologies used in for the extraction and leaching of plastic additives are available elsewhere (e.g. Ref. [13]), and, thus, a summary of some of the most commonly used methodologies for the identification and quantification of additives in plastic samples in environmental studies is found in Table 1.

From Table 1, it also emerges that multiple analytical approaches and methodologies exist to characterize and quantify plastic additives derived in different products and matrices, such as pharmaceuticals, food and food contact materials and medical devices, among others. Nonetheless, the variety and analytes, their disparate concentrations and often complex matrices render these

**Table 1**

Commonly used methodologies for the extraction/leaching and analysis of plastic additives. Plastic material samples, including their origin, as well as experimental conditions are noted (examples are merely illustrative and do not purport to constitute an exhaustive list of methodologies and/or analyses performed).

Type of study	Polymer sample(s)	Sample origin	Extraction/Leaching	Analytical technique	Analytes	Ref.
Extraction	Microsized PE, PP, PS	Beached marine debris	Hexane-sonication accelerated extraction	GC-MS	Phthalates; benzotriazoles; brominated flame retardants	[18]
	Microsized PE, PP, PS, polybutadiene	Floating marine debris	Toluene-sonication accelerated extraction	LC-MS	Phthalates, BPA derivatives, benzotriazoles, phenolic antioxidants	[43]
	Microsized PET, PA	Commercial	Dichloromethane -sonication assisted extraction	GC-MS, LC-MS	Phthalates, BPA derivatives, benzophenones, benzothiazoles, organophosphates	[68]
	Microsized PS	Beached debris	Dichloromethane -microwave accelerated extraction	Pyr-GC-MS	Phthalates, NIAS	[10]
Leaching	Microsized PET, PA, PAN	Commercial	Static; fresh water, seawater; 5 °C; 20 °C; stirred	GC-MS, LC-MS	BPA derivatives, benzophenone, NIAS	[65]
	Mesosized ePS	Commercial	Static; seawater; 10 mg/500 mL; 25 °C; stirred	GC-MS	Styrene oligomers	[74]
	Microsized PVC	Commercial	Static; seawater; 2 g/500 mL; 25 °C; stirred	GC-MS, ICP-MS	Organotin compounds	[15]
	Microsized PC, car tyre rubber	Commercial	Static; seawater; 20 °C; stirred twice daily	HPLC-FLD	BPA, alkylphenols	[70]

PE – polyethylene; PP – polypropylene; (e)PS – (expanded) polystyrene; PET – polyethylene terephthalate; PA – Polyamide; PAN – polyacrylonitrile; PVC – polyvinyl chloride; PC – Polycarbonate; GC-MS – Gas chromatography-mass spectrometry; LC-MS – Liquid chromatography-mass spectrometry; HPLC-FLD – High-performance liquid chromatography with fluorescence detector; Pyr-GC-MS – Pyrolysis gas chromatography; ICP-MS – Inductively coupled plasma mass spectrometry; BPA – Bisphenol A; NIAS – Non-intentionally added substances.

analyses highly complex. Analytical methods may, therefore, be targeted, non-targeted and non-specific. The latter provide general information about the extract as a whole, whilst targeted analyses may provide qualitative and quantitative information for specific and known analytes; on the other hand, non-targeted analyses aim at providing a broad and comprehensive screening of as many analytes as possible, as well as providing their partial or complete identification [13]. Non-specific methodologies may include UV–Vis absorbance, Total Organic Carbon, pH or gravimetric-based methods, whilst targeted and non-targeted analyses of organic compounds typically resort to chromatographic separation techniques coupled with selective (e.g., MS) or non-selective (e.g., UV) detectors, as underscored in Table 1.

As surmised from Table 1, the most used analytical identification and quantification methods are GC-MS or LC-MS techniques. These enable a wide range of additives to be accurately identified and quantified, owing to the methods' specificity, sensitivity and reproducibility. Nonetheless, the current sheer number of chemicals added to plastic products, as well as the often-limited disclosure on both the type and level of additives present render the identification and quantification of leachables/extractables challenging. This is particularly difficult for environmental samples, such as plastic debris, given that the composition is frequently entirely unknown and non-targeted analyses are needed [13]. It is therefore necessary not to rely on a single analytical method, as the chosen technique may not be suited for identifying all of the chemicals present. For example, GC-MS analyses performed on beached plastic debris showed no evidence of the additive Organo-1010 on any of the tested samples [59], but a subsequent study by the same research group demonstrated that same additive was present on all samples when resorting to GC-MS [60].

The extent of leaching has been quantified by measuring the decrease in concentration within the tested material, but, most frequently, it is determined by measuring the variations in the concentration of the chemicals of interest in the liquid media. These concentrations are often considered to be bioavailable, and subsequent ecotoxicity assessments are based on these measurements. However, bioavailability is a rather dynamic concept that considers chemical, physical and biological processes related to both exposure and dose. Bioavailable exposure doses depend on the mass transfer and uptake of contaminants into organisms, which in turn

are determined by the properties of the chemical(s), matrix characteristics, the biology of organisms, and climatic influences [5]. As such, to predict the impacts of plastic additives on ecosystems, it will be necessary to understand the individual bioaccessible availability and associated uptake mechanisms, as the total amount of chemicals released to the environment does not equal the potentially toxic dose.

A wide range of different resins of distinct and varied physical and chemical properties have been studied in order to better understand the potential exposure to leachables originating from plastic materials found in the environment, of which PE, PS and PVC are the most frequent examples. However, other materials, as polyester and polyurethane, remain severely understudied. Most studies also focus on plastics of considerable size, and, given that the rates of additive release have been shown to increase with decreasing plastic particle size [15], as well as the environmental prevalence of smaller plastic particles, the current lack of studies that examine the sorption and release by minute plastic materials (<100 µm), currently constitutes a research discontinuity that is of environmental relevance.

## 6. Outlook

Both microplastics and plastic additives constitute contaminants of emerging concern, an umbrella term that includes naturally occurring or synthetic chemicals, as well microorganisms, that are not commonly monitored in the environment, although having the potential to cause known or suspected adverse ecological and/or human health effects. With limited or no applicable regulatory standards, these contaminants have been described in an increasing number of samples from distinct environmental matrices. Notwithstanding the extensive literature review and the currently available data on the mechanistic behavior between microplastics and organic contaminants, much remains unknown, particularly regarding their overall impact in the environment. Most of the currently available data stems from simplified systems, and the complex environmental interactions and their consequences on the sorbate-sorbent interactions in real-world settings are still very much undetermined. These oversimplified approaches to the equilibrium isotherms and kinetics should be replaced with integrated strategies that consider all the mass transfer steps and



related limiting effects on the rate of sorption – uptake, and release. Moreover, the vast majority of studies have focused on aquatic systems, mostly, marine environments; hence, it becomes necessary to extend these fundamental experimental works to other environmental matrices, namely, soil. Critical areas, such as landfills, should be prioritized, as the commonly observed leaching processes in these facilities could enhance the role of microplastics as vectors for organic contaminants.

On an ecological level, the accurate bioavailability of organic compounds and the role of microplastics – of different types, morphologies, and degrees of weathering – on this bioavailability should be determined. Additionally, such assessments should be carried out under environmentally relevant conditions, including concentrations of both microplastics and organic pollutants, which, in most cases, far exceed the values found in the environment; similarly, factors such as equilibrium time and the (often) transient nature of exposure should be considered when determining the putative ecotoxicological effects of the sorption/leaching of organic pollutants to/from microplastics.

Analytically, methodologies already in place appear to be suited for assessing the presence of contaminants associated to microplastics; yet, special attention should be paid regarding the overall goal of conducted studies, i.e., determination of leachables and/or extractables, and, whenever possible, complementary assessments should be conducted, in order to ascertain the presence of chemical substances that may not be detectable when using a given technique. Most importantly however, it is necessary, from a regulatory point-of-view, to enforce a full disclosure of the chemicals added to plastic products, so that a more complete and detailed evaluation on their presence in environmental matrices is possible. Finally, studies focusing on the *de facto* bioavailability of these contaminants is necessary, so that their realistic ecotoxicological impacts may be determined.

### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

### Data availability

No data was used for the research described in the article.

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### Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.trac.2022.116898>.

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