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An overview of chemical additives present in plastics: Migration, release, fate and environmental impact during their use, disposal and recycling.

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Abstract

Over the last 60 years plastics production has increased manifold, owing to their inexpensive, multipurpose, durable and lightweight nature. These characteristics have raised the demand for plastic materials that will continue to grow over the coming years. However, with increased plastic materials production, comes increased plastic material wastage creating a number of challenges, as well as opportunities to the waste management industry. The present overview highlights the waste management and pollution challenges, emphasising on the various chemical substances (known as “additives”) contained in all plastic products for enhancing polymer properties and prolonging their life. Despite how useful these additives are in the functionality of polymer products, their potential to contaminate soil, air, water and food is widely documented in literature and described herein. These additives can potentially migrate and undesirably lead to human exposure via e.g. food contact materials, such as packaging. They can, also, be released from plastics during the various recycling and recovery processes and from the products produced from recyclates. Thus, sound recycling has to be performed in such a way as to ensure that emission of substances of high concern and contamination of recycled products is avoided, ensuring environmental and human health protection, at all times.

Keywords: plastics; additives; migration; recycling; toxicity; environmental fate

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NOMENCLATURE

ABS	Acrylonitrile-butadiene-styrene
ATBC	Acetyltributyl citrate
BADGE	Bisphenol A diglyceride ether
BAT	Best available technique
BBP	Benzyl butyl phthalate
BBP	Butyl benzyl phthalate
BEP	Best environmental practice
BHA	2- and 3-t-butyl-4-hydroxyanisole
BHT	Butylated hydroxytoluene
BPA	Bisphenol A
DBP	Dibutyl phthalate
DCHP	Dicyclohexyl phthalate
DDE	Dichloro-diphenyl-dichloro-ethylene
DEHA	Di-(2-ethylhexyl) adipate
DEHP	Bis (2-ethylhexyl)phthalate
DEHS	Diethylhexyl succinate
DEP	Diethyl phthalates
DHA	Diheptyl adipate
DHNUP	1,2-Benzenedicarboxylic acid, di-C7-11-branched and linear alkyl esters
DiBP	Diisobutylphthalate
DIDP	Diisodecyl phthalate
DIHP	Diisoheptylphthalate
DINP	Diisononyl phthalate
DL-PCBs	Dioxin-like polychlorinated biphenyls
DMEP	Bis(2-methoxyethyl) phthalate
DOA	Di-octyladipate
DOP	Dioctyl phthalate
DPP	Dipentyl phthalate
EEE	Electrical and electronic equipment
EfW	Energy from Waste
EoL	End-of life
EPDM	Ethylene-propylene diene rubber
ESBO	Epoxidized soybean oil
EU	European Union
FDA	Food and drug administration
FR	Flame retardants
FS	Food simulants
HAD	Heptyl adipate
HALS	Hindered amine light stabilizers
HBCCD	Hexabromocyclohexane
HBCDD	Hexabromocyclododecane
HDA	Hexanediol adipate
HDPE	High-density polyethylene
HOA	Heptyl octyl adipate
LCA	Life cycle analysis
LCCP	Long chain chlorinated paraffins
LDPE	Low-density polyethylene
LLDPE	Linear low-density polyethylene
LOQ	Limit of quantification
MCCP	Medium chain chlorinated paraffins
MDA	4,4'- Methylenedianiline
MOCA	2,2'-dichloro-4,4'-methylenedianiline
MTBE	Methyl tert-butyl ether

MW	Microwave
NCTR	National Center for Toxicological Research
PAEs	Phthalate esters
PAHs	Polycyclic aromatic hydrocarbons
PBDD/Fs	Polybrominated dibenzo-p-dioxins and furans
PBDEs	Polybrominated diphenyl ethers
PC	Polycarbonate
PCBs	Polychlorinated biphenyls
PCDD/Fs	Polychlorinated dibenzo-p-dioxins/furans
PET	Polyethylene terephthalate
PLA	Polylacti(c)de
PMMA	Polymethyl metacrylate
POM	Polyoxymethylene
POPs	Persistent organic pollutants
PoTSs	Potentially toxic substances
PP	Polypropylene
PS	Polystyrene
PSW	Plastic solid waste
PTFE	Polytetrafluoroethylene
PVC	Poly vinyl chloride
PXDD/Fs	Mixed halogenated dibenzo-p-dioxins/dibenzofurans
RfD	Reference dose
RoHS	Restriction of hazardous substances
rPET	Recycled polyethylene terephthalate
SCCP	Short chain chlorinated paraffins
SRF	Solid recovered fuel
TBBPA	Tetrabromobisphenol A
TCEP	Tris(2 chloroethyl)phosphate
TCPP	Tris(2-chlorisopropyl)phosphate
TEs	Trace elements
TGIC	1,3,5-Tris(oxiran-2-ylmethyl)-1,3,5-triazinane-2,4,6-trione
TNPP	Tris(4-nonyl-phenyl) phosphate
TPS	Thermoplastic starch
TVOCs	Total concentration of VOCs
UN	United Nations
UV	Ultraviolet
VOCs	Volatile organic compounds
WEEE	Waste electrical and electronic equipment
β-TGIC	1,3,5-Tris[(2S and 2R)-2,3-epoxypropyl]-1,3,5-triazine-2,4,6-(1H,3H,5H)-trione

1. Introduction

The production of plastic materials started to flourish on an industrial scale in the 1940s and 1950s. During the last 15 years, the global annual production of plastics has doubled, reaching approximately 299 million tonnes in 2013 [1, 2]. Global plastic demand is dominated by thermoplastic types of polypropylene (PP) (21%), low -and linear low- density polyethylene (LDPE and LLDPE) (18%), polyvinyl chloride (PVC) (17%), and high-density polyethylene, (HDPE) (15%). Other plastic types with high demand are polystyrene (PS), and expandable PS (8%), polyethylene terephthalate (PET) (7%, excluding PET fibre) and the thermosetting plastic polyurethane [3].

Plastic polymers are not only used for consumer products but also to make synthetic fibres, foams, coatings, adhesives and sealants, which are used in numerous applications [4]. In Europe, the use of plastics is mostly dominated by packaging (38%), followed by building and construction (21%), automotive (7%), electrical and electronic (6%), and other sectors (28%), such as medical and leisure [2].

Plastics are important in our society providing a range of benefits for human health and the environment. For example [2, 5]:

- Plastic packaging protects food and goods from getting wasted and/or contaminated, thereby saving resources.
- The light weight of plastic packaging compared to other materials saves fuel and decreases emissions during transportation.
- Plastic water supply systems and storage containers/tanks can provide clean water.
- Low-density plastic materials, used as replacements for metals or ceramics in cars and aircraft, save fuel and decrease emissions.
- Plastic protective clothing and safety equipment (e.g. fire proof materials, helmets, air bags) prevent injuries.

- Plastic products for medical applications contribute to improved heath (e.g. blood pouches, tubings, disposable syringes, prosthesis).

However, such diverse consumption leads to a diverse waste stream. Large volumes of plastic wastes are generated, mainly due to the short lifespan of many plastic products (it is estimated that approximately 40% of plastic products have a service life of less than 1 month). This large waste creates serious environmental and management problems [6].

The key problems associated with plastic waste, their disposal and treatment are analysed briefly in the sections below. The main objective of this article is to delineate the challenges and complications – both environmental and technical- encountered during the disposal of plastic products at their end-of-life (EoL) stage. It is, also, in the authors' intention to include any barriers and/or constraints that are posed when trying to "close the loop" in the plastic sector. Specific focus is given on the migration and release potential of various additives present in plastic food contact materials. In addition, an assessment on the emission/leaching of potentially toxic substances (PoTSs) during recycling processes for all kinds of plastics, with an emphasis on developing countries, is also presented. Finally, some recent improvements on the recycling of plastics are briefly discussed.

2. Methodology

The present work is neither a systematic nor a comprehensive review. The authors, here, attempt to give a generic overview on the several implications that are associated with plastics, both as products as well as waste, and on the various PoTSs embedded in them, under a three pillar approach:

a) to briefly delineate the problem of plastic waste and their potential risk when/if entering the marine and terrestrial environment, by accounting also for alternative waste management options (e.g. sound recycling methods);

b) to list the most commonly used additives in plastics and plastic packaging (mostly food packaging) and discuss their uses and applications with specific focus on the potential migration of the various chemical substances embedded in them under various conditions; and

c) to outline the potential risks (emission, release, fate, etc.) that could result from the presence of those additives during inappropriate and/or uncontrolled disposal and recycling processes (e.g. in developing countries). A general description of the potential of these additives to contaminate soil, air and water is, also, provided.

The methodological approach to this review, therefore, took 3 steps:

1) Plastics -as a "waste" problem- was delineated. The authors thoroughly reviewed literature on studies relative to: a) the improper management of plastics (e.g. insufficient sorting), proposing, at the same time, several alternatives for recovering value from waste plastics (through optimising recycling and/or recovery processes) and b) their potential risk when/if entering the marine environment, particularly examining the possibility and consequences of fragmentation and/or degradation processes.
2) The most commonly used additives in plastics and plastic packaging (mostly food packaging) were categorised and described; their uses, applications, properties enhanced and chemical substances involved for each individual category.
3) The potential risks that could result from the presence of those additives and the various mechanisms / phenomena associated with the use, disposal, and recycling of plastics (e.g. migration, release, emission and fate in general) were presented.

The data collection process involved searching both scientific literature (Scopus, Science Direct) and "grey" literature (general commercial, trade body and industrial collections), using theoretical and empirical articles. The keywords, either individually or in combinations, under which the articles were sought and finally selected, were: additives, plastics, plastic waste, microplastics, macroplastics, release, migration, recycling and plastic debris.

3. Plastic waste - A growing challenge

3.1 Plastic waste management options

The life cycle of a plastic material passes, mainly, through the three phases presented in Fig. 1. Despite significant worldwide advances in management, treatment and recycling in the last three decades, the largest fraction of plastic waste still possibly ends up in dumpsites or is openly burned, emitting carbon monoxide (CO) and carbon dioxide (CO_2). At best, they might end up in engineered landfills. In Europe, which arguably with Japan has the most technically advanced and environmentally conscious waste and resource recovery systems deployed on the ground, approximately 50% of plastic waste is still directed to controlled landfill disposal [2]. This is highly debated, as many consider it as an unacceptable wastage of resources and promote at least resource recovery via energy form waste (EfW), and others aspire to it as temporary storage, anointing so greenhouse emissions release during thermal processing, until, if ever, landfill mining becomes viable [7].

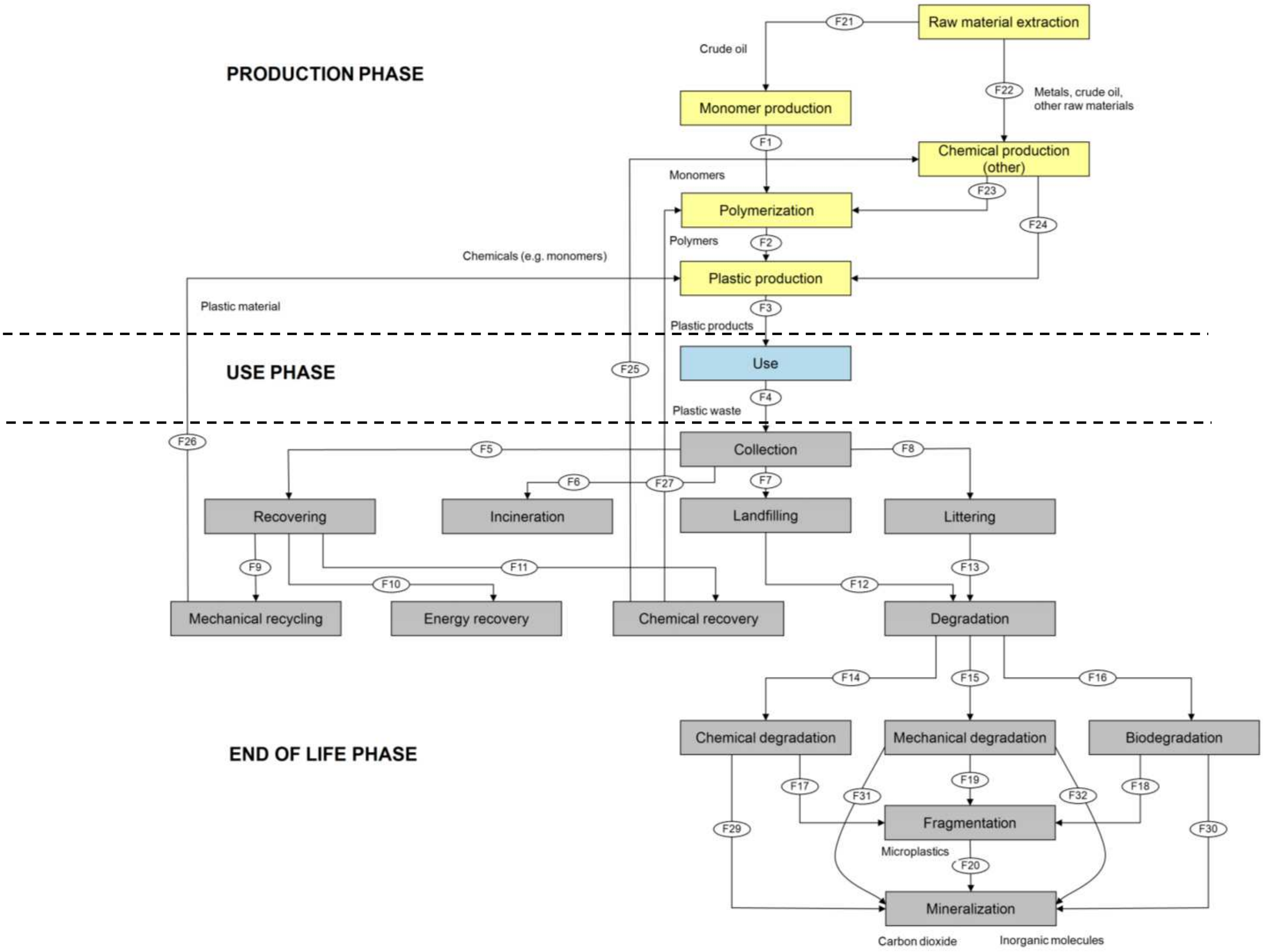

Fig. 1. The life cycle of plastic products (excluding energy input and emissions) created in STAN (sub**ST**ance flow **An**alysis) Software (redrawn from source: [8]). "F" stands for Flow and the number shown next to it corresponds to the particular flow series (an automatic procedure incorporated in STAN).

Resource recovery alternatives to landfill are mechanical recycling (primary recycling substituting virgin materials and secondary recycling), chemical recovery (tertiary recycling) or energy recovery (quaternary recycling) [9, 10]. Primary recycling substituting virgin polymers in the same application is possible for some plastic types and fractions (e.g. for PET plastic bottles or car bumpers) [10]. However, among else, the great variability in plastics polymers and post-use contamination obstructs closed-loop recycling or makes it difficult [10, 11].

For the vast majority of plastic waste fractions (e.g. most packaging, plastic from electronics, plastic and polymers from the transport and construction sectors) labour or technology intensive sorting is needed in order to get a high quality recyclate which can be used for substituting virgin materials [12]. Often, plastics are in most cases secondary recycling applies, in which used plastics are cascaded into material applications different than the original, and often of less demanding material specifications (e.g. PET bottles into fleece). Plastic waste for recycling could be transported over long distances, for instance exported from the Global North to the developing countries, particularly to Asia [13]. It was shown that 46% wt. of the plastics collected for recycling in Europe were eventually exported, with 90% wt. of it directly or indirectly ending up in China [14, 15].

Chemical recovery (tertiary recycling), involves chemicals, e.g. raw materials such as monomers, being recovered and/or converted from the plastic material [9]. This can potentially be done by catalytic depolymerisation or by controlled thermal degradation, such as thermolysis, which is a non-catalytic cracking process [16]. Pyrolysis is also considered to be a sustainable and efficient treatment that can produce a range of useful hydrocarbons, potentially used as a chemical feedstock or as energy, thereby minimizing the dependency on non-renewable fossil fuels, while solving the landfilling problem [17, 18].

Whereas in the Global North plastics production, use and recycling is regulated to varying degrees, in many developing countries plastic recycling is often not controlled by an appropriate regulatory framework, and environmental protection is poorly enforced, resulting in significant contamination of the ambient environment in areas where plastic is recycled [19]. Uncontrolled recycling can also result in the transfer of potentially harmful substances or PoTSs into plastics for sensitive uses, such as children’s toys and food contact materials [20-

23]. Some processes for cascading involve innovative uses for plastic waste, with untested potential for dispersion into the environment and associated unknown pollution. For instance in India, used plastic is recycled into asphalt, as an alternative road material [24]. The asphalt is made from churned plastic waste (mainly composed of plastic bags, PET bottles and thin plastic film) which is blended with bitumen [25, 26]. Such roads are only expected to last for 4-6 years, and the EoL management of the material recovered from road maintenance activities is unclear [25, 26].

In industrial countries a large share of plastic waste is used for energy recovery. In Europe, more plastics waste is destined for energy recovery (39.5%) (in EfW or via solid recovered fuels (SRF) recovered in cement kilns) than for recycling (29.7%) [27]. However, uncontrolled combustion of plastic waste and, in particular of those containing halogens such as, PVC, polytetrafluorethylene/teflon, plastic containing brominated flame retardants, etc. can cause emissions of hazardous substances, e.g. acid gases and unintentional persistent organic pollutants (POPs) such as dioxins [28, 29]. Halogens emitted from the combustion of plastic waste can also cause corrosion in incinerators and other thermal facilities. Chlorine and bromine may accumulate in cement kiln system limiting their capacity for thermal recovery of plastic [28, 30, 31]. Furthermore since most plastics are fossil-fuel based, incineration may also contribute to global warming and depletion of petrochemical resources. On the other hand, controlled combustion in EfW plants and cement kilns equipped with state of the art air pollution control technologies may be the best way available to limit the dispersion of POPs.

3.2 Plastic waste leaking into the marine and terrestrial environment

A non-negligible fraction of plastic waste, unfortunately, ends up as litter in the marine and terrestrial environment and ecosystem, creating various environmental, economic and social impacts [32, 33].

Although the precise amount of plastics entering the marine environment is yet unknown, by linking worldwide data on solid waste, and using population density, a rough estimate within the range of 4.8-12.7 Mt per year on the mass of land-based plastic waste entering the ocean has been calculated [34, 35]. According to Jambeck et al. [34] the quantities of plastics entering the oceans from land are expected to increase by approx. an order of magnitude by 2025 (calculations involving several uncertainties and assumptions).

The marine environment and its living organisms are particularly exposed to plastic waste contamination and various studies have documented its negative consequences [36-42]. Large and continuously increasing amounts of plastic products and debris are found in the open ocean, either on the surface or in the benthos of the deep seas, as well as in shorelines and living organisms [34, 43-47]. Evaluating the external cost of the plastic life cycle, a United Nations (UN) report notes that marine pollution is the largest downstream cost, and that that the calculated first approximation cost of US$13 billion is likely a significant underestimate [40]. The sources of marine plastic litter vary from land based releases, e.g. land littering (especially coastal areas), plastics that could have been blown from open dumpsites or leached sewage effluents, as well as spillage during transport and/or accidents with only a minor share from dumping at the sea [43]. This plastic waste can then be transported far away by currents, e.g. the great oceanic gyres [48], and/or accumulated in the centre of gyres [49, 50].

Plastics can be divided mainly into three groups, according to their particle size. All plastic materials >5 mm are considered to be macroplastics. When entering the marine environment, plastic products and debris can potentially be fragmented into smaller pieces, <5 mm, called microplastics [43, 51]. Microplastics are usually defined within the approx. particle size range of 50 μm - 5 mm [52]. In fact, the pollution caused by this group of plastics is considered to be more prevalent owing to their larger quantities and small particle sizes. Finally, several implications of nanometre-sized plastic particles (mostly known as ‘nanoplastics’) and defined in <100 nm of size, constitute a very recent area of the environmental science, probably the least known area of marine litter but potentially equally (if not more) hazardous [53].

Yamashita and Tanimura [48], performed studies in surface trawls (net mesh size 330 μm) and found plastics at 55 of 76 locations in the Kuroshio Current area (North Pacific), in quantities ranging from $0–3.52\times10^6$ pieces per km^2, with a mean abundance of 1.74×10^5 pieces km^{-2} [48]. Particle sizes between 1-3 mm constituted 62% of all marine plastic pieces. However, even smaller sizes of plastic pieces ≤20 μm (close to the range of nanoplastics) in sediment have been reported [44].

Moore [49] not only underpinned the negative effects of synthetic polymers in the marine environment and their abundance in the marine litter, but also calculated a mean abundance of 3.34×10^5 plastic pieces km^{-2}, near the central pressure cell of the North Pacific subtropical high [49, 54]. Microplastics were even found in Antarctic waters but relevant studies report on available data being scarce [55].

Various consequences from ingestion of macro-, micro- and nano- plastics or entanglement of macroplastics have also been reported and well documented for various species i.e. birds, turtles, fish larvae and marine mammals [35, 56]. Results of this ingestion include suffocation or blocking of digestive tract causing death [35]. Several studies performed in amphipods, lugworms, sea cucumbers, and mussels that have been exposed to microplastics, have shown that these organisms tend to ingest the microplastics [44, 45, 56]. Further studies on the mussels demonstrated that the microplastics were translocated from the gut to the circulatory system and were found in the hemocytes [45]. Several consequences of exposure to microplastic in filter feeding organisms and deposit feeders are yet to be discovered.

The propensity (or ability) of plastics to sorb POPs is, also, known to potentially cause additional problems. There are various articles in literature that have attempted to investigate this tendency, either by examining the different conditions of microplastics present in marine and estuarine systems (e.g. salinity) [57, 58] or by determining the effect of the different characteristics of the polymers that constitutes them [52, 59]. However, it is not clear yet as to what extent this bidirectional interaction (sorption and release of POPs in plastics) can take place. Marine PP pieces were found to have 100.000 to 1 million times higher concentrations of PCBs (polychlorinated biphenyls) and DDE (dichloro-diphenyl-dichloro-ethylene) than in the surrounding seawater [60]. To what extent these pollutants are released when ingested by an organism, is an issue that still remains to be investigated and clarified. In recent model analyses it was reported that the effects of plastic waste on bioaccumulation of POPs may be rather small, due to a lack of a gradient between POPs in plastic and the biota lipids, and that it is possible for a cleaning mechanism to dominate at higher Log K_{OW} values [38].

The various additives present in almost all plastic-derived materials can also contribute to marine pollution. Some plastics contain POPs as additives (e.g. hexabromocyclododecane (HBCDD or HBCD) and/or polybrominated diphenyl ether (PBDE)) at a concentration of 0.7-25% wt. [12, 61]. For additives therefore, plastic ingestion by marine organisms may be more relevant than the accumulated diffusely spread POPs, since the levels are 7-10 orders of magnitude higher [62-64]. Even if not ingested the additives containing polymers, still constitute exposure sources, e.g. increased HBCDD content has been found in oysters in a farm where PS buoys containing HBCDD were used [65]. On the other hand, the leaching of additives may be more relevant for species with longer gut retention times, such as fish [66]. While microplastic ingestion lead to increased bioaccumulation of plastic additives, there is speculation that they might lead to a decrease of bioaccumulation of traditional POPs [46, 62] which has however not been demonstrated.

Specific insight on the release and fate of plastic additives to the environment, owing to their use, disposal and uncontrolled recycling, as well as the mechanism of their migration to food and, consequently, humans through the various packaging materials are given in Sections 5.2 and 5.3.

3.3 Plastic materials persistence and degradation

The state in which a plastic material remains over time could be affecting how it interacts with its environment and therefore affect the potential release of substances (additives) contained in it [41]. Most of engineered polymers are manufactured with long-term stability of their properties in mind: they are not meant to break down easily, which in many cases is part of the core functionality delivered by the material in the intended use [41].

Plastics are considered to be persistent pollutants. The majority of plastic polymer types are non-biodegradable, i.e. degradation by microorganisms, where PET and PP being the most abundant ones, are practically non-degradable [67]. In a PET polymer, only a mere 0.1% of the carbon will be transformed into CO_2, per year, via biodegradation and that will only occur under ideal laboratory conditions. Biodegradable plastics today have a minor, but continuously growing, share in the plastics market. However, not all of them are entirely biodegradable in the natural environment [68, 69], plus specific types (e.g. biodegradable plastic bags) may alter marine sediment geochemistry and influence species coexistence [70].

However, non-biodegradable polymers can be degraded/fragmented by various mechanisms: physical, such as heat and light, and chemical, such as oxidation, ionic radiation, and hydrolysis. Certain air pollutants such as CO, sulphur dioxide (SO_2), nitrous oxide (NO) and ozone (O_3) can also play a major role in the degradation of polymers [71]. The effect of the aforementioned degradation mechanisms to the polymer is to embrittle and fragment it into smaller pieces.

At molecular level, various degradation mechanisms exist and the domination of one mechanism over the others often depends on the polymer type. Chain scission, for example, involves breaking the chemical bonds of the polymer molecule, and although it is often random, for some polymers it takes place at the polymer end chains and the initial monomers are broken off, a process known as depolymerisation [72]. Another degradation mechanism is chain stripping according to which the side atoms/groups attached to polymer chain are released; usually these are volatile molecules. In crosslinking bonds are created between the polymer chains. However, a complete conversion of aplastic material to its main constituents (CO_2, water and inorganic molecules) via photo- and bio- degradation is rather unlike to happen [73, 74]. Under marine conditions, any degradation that might occur would be quite slow, given the main involving mechanisms e.g. solar radiation and slow thermal oxidation [73]. Hence, the time frame for a complete degradation could be extensively prolonged, reaching, in some cases, even hundreds of years.

4. Additives in plastic products

In plastic materials used in most products the basic polymer is incorporated into a formulary (plastic compound) with different ‘additives’, which are chemical compounds added to improve the performance (e.g. during shaping of the polymer, through injection moulding, extrusion, blow moulding, vacuum moulding, etc.), functionality and ageing properties of the polymer. The most commonly used additives in different types of polymeric packaging materials are: plasticizers, flame retardants, antioxidants, acid scavengers, light and heat stabilizers, lubricants, pigments, antistatic agents, slip compounds and thermal stabilizers. Each of them plays a distinct role in delivering/enhancing the (final) functional properties of a plastic product.

For instance, catalyst deactivators neutralize any remaining catalyst residues, nucleators increase resin clarity and reduce processing time, and pigments provide a variety of colours. Antistatic agents permit the discharge of static electricity from the film or part, and the addition of flame retardants allows the use of PP in electronics, construction, and transportation applications. Antiblock and slip agents are commonly used in films, to prevent the latter from sticking together, or to metal surfaces. All the aforementioned additives can be mainly divided into the following 4 categories [75]:

- Functional additives (stabilisers, antistatic agents, flame retardants, plasticizers, lubricants, slip agents, curing agents, foaming agents, biocides, etc.)
- Colorants (pigments, soluble azocolorants, etc.)
- Fillers (mica, talc, kaolin, clay, calcium carbonate, barium sulphate)
- Reinforcements (e.g. glass fibres, carbon fibres).

Table 1 presents a more detailed, but still brief, description of the most common functional types of additives used in plastics. It needs to be stressed that additives, in nearly all cases, are not chemically bound to the plastic polymer. Only the reactive organic additives, e.g. some flame retardants, are polymerised with the plastic molecules and are becoming part of the polymer chain. It should, also, be noted that substances used as monomers, intermediates or catalysts in plastic manufacturing are not considered to be additives and therefore have not been included in Table 1.

Table 1. Short description of the most commonly used additives in plastic materials (recreated from source: [75])

Category/Type of additive	**Typical amount range (% w/w)**	**Substances**	**Additional comments-explanations**
Functional additives			
Plasticisers	10-70	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).	About 80% is used in PVC while the remaining 20% in cellulose plastic.
Flame retardants	3–25 (for brominated)	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)); Phosphorous flame retardant (e.g. Tris(2-chloroethyl)phosphate (TCEP) Tris(2-chlorisopropyl)phosphate (TCPP)).	Three groups: • organic non- reactive (e.g. phosphate esters, halogenated phosphate esters, halogenated hydrocarbons) • inorganic nonreactive (e.g. antimony oxide, aluminum oxide trihydrate, zinc borate, ammonium orthophosphate, ammonium sulfamate) and • reactive (e.g. bromine and/or phosphorus containing polyols, halogenated phenols, tetrachlorophthalic anhydride, phosphonate esters, dibromoneopentyl alcohol)
	0.7-3	hexabromocyclohexane (HBCDD)	
Stabilisers, Antioxidants and UV stabilizers	0.05-3	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-	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.

		phenyl phosphate (TNPP), tris (2, 4-di-tert-butylphenyl) phosphite, (Irgafos 168).	
Heat stabilisers	0.5-3	Cadmium and Lead compounds; Nonylphenol (barium and calcium salts).	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.
Slip agents	0.1–3	Fatty acid amides (primary erucamide and oleamide), fatty acid esters, metallic stearates (for example, zinc stearate), and waxes.	The amounts are dependant on the chemical structure of the slip agent and the plastic polymer type.
Lubricants (internal and external)	0.1–3	-	-
Anti-statics	0.1-1	-	Most types are hydrophilic with the potential to migrate to water.
Curing agents	0.1-2	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-epoxypropyl]-1,3,5-triazine-2,4,6-(1H,3H,5H)-trione (β-TGIC).	Peroxides and other crosslinkers, catalysts, accelerators.
Blowing agents	Depends on the density of the foam and the potential gas production of the agent	-	Azodicarbonamide, benzene disulphonyl hydrazide (BSH), pentane, CO_2.
Biocides	0.001-1	Arsenic compounds; Organic tin compounds; triclosan.	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.
Colorants			
Soluble (eg. 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.

Organic pigments	0.001-2.5	Cobalt(II) diacetate.	They are insoluble with low migration tendency.
Inorganic pigments	0.01-10	Cadmium compounds; Chromium compounds; Lead compounds.	E.g. zinc sulphide, zinc oxide, iron oxide, cadmium-manganese based, chromium based, ultramarine and titanium dioxide.
Special effect	Varies with the effect and substance in question	-	Al and Cu powder, lead carbonate or bismuthoxichloride and substances with fluorescence. Substances with fluorescence might migrate, the former not.
Fillers			
	Up to 50	-	Calcium carbonate, talk, clay, zinc oxide, glimmer, metal powder, wood powder, asbest, barium sulphate, glass microspheres, silicious earth.
Reinforcements			
	15 -30	-	Glass fibers, carbon fibers, aramide fibers. 15-30% is for glass only due to is high density.

NOTE: “-” there is not information reported

For most of them, their uses, applications and potential toxicity are reported in the sections below.

4.1. The role of compatibilizers in the miscibility of polymers

Polymer blending has been extensively used over the last few decades to produce new polymeric materials that combine the individual attributes of the component polymers [76]. Compatibilizers are substances that are commonly used to enable the creation of such special resin blends (co-polymer), with the desired performance, starting from component resins that would otherwise be incompatible. Compatibilization is often a necessary procedure in blending polymers mainly due to the immiscibility and incompatibility of most polymer pairs. Specifically, use of compatibilizers improve the overall performance of the blend thorough: improved blend morphology and enhanced interfacial adhesion [77-79].

Furthermore, a number of countries have adopted legislation to promote the use of biodegradable additives in polyolefins and PET [80]. The targeting goal behind the use of biodegradable plastics is the assimilation of these materials back into the environment [81].

Grigsby et al. [82] reported that tannin esters exhibited potential to be used as functional additives in biodegradable polymers enhancing the plastic's UV stability [82]. A key factor in such assimilation is the time period required. In order to obtain substantive environmental benefit, such assimilation must occur within a reasonable time frame [83].

Peres et al. [84] evaluated the effect of reprocessing on the structure and properties of a low density polyethylene/thermoplastic starch (LDPE/TPS) blend compared to LDPE. The results indicated that multiple extrusion steps led to a reduction in the average size of the starch-rich phases of LDPE/TPS blends and to minor changes in the mechanical and rheological properties of the materials. Hence , LDPE/TPS blend presented similar reprocess behaviour to LDPE [84].

Xanthos et al. [85] reported the use of ethylene-propylene diene rubber (EPDM) for PP and PE, typical materials for bottles. It is available for example as Keltan 5170P from Lanxess Gmbh [85].

Another worth mentioned nuance is that compatibilization of two or more polymer sources in waste plastics can lead to advantageous combinations of properties and/or new properties, not present in either of the initial materials. For example, contamination of polylactide (PLA) by linear LDPE makes it significantly more impact resistant when these two polymers are compatibilized [86]. It may even lead to a possible price premium for recycled materials.

4.2 Use and application of most common additives

4.2.1 Plasticizers

Plasticizers are most commonly used for improving the flexibility, durability and stretchability of polymeric films, reducing, at the same time, melt flow [87, 88]. Plasticizers reduce shear during the mixing steps of polymer production and improve impact resistance in the final plastic film. They, also, provide the material with limp and tacky properties [88-90]. Some important plasticizers include: phthalic esters (PAEs), such as DEHP used in PVC formulations, and constitute about 80% of the plasticizer volume for PVC production; plasticizers for PET may include DPP, DEHA, DOA, DEP, diisobutylphthalate, and DBP; acetyltributyl citrate (ATBC) is a plasticizer for PVDC-based cling-films; finally, other commonly reported plasticizers are DBP, DEHP, DHA, DCHP, BBP, HAD, and HOA [90].

4.2.2 Antioxidants

Antioxidants are embedded in various polymer resins to delay the overall oxidative degradation of plastics if/when exposed to ultraviolet (UV) light [88, 90, 91]. The highly reactive free radicals that are generated by heat, radiation, and mechanical shear (often exacerbated by the presence of metallic impurities), cause the polymer to degrade. In food packaging, the potential for oxidation increases in the case of exposure to high temperatures, including contact with hot foods, exposure to infrared heating, retort processing and microwave (MW) heating [88].

Arylamines are the most commonly used antioxidants in plastic food packaging. Phenolics and organophosphites (used to reduce hydroperoxides formed during oxidation to alcohols) are also used as antioxidants. The first group includes BHT, BHA, Irganox 1010, BPA and Cyanox 2246 and 425 [92], while the latter group includes TNPP and Irgafos 168 [88].

4.2.3 Heat stabilizers

Heat stabilizers are responsible for preventing thermal degradation of polymers when exposed to elevated temperatures i.e. during the thermal processing of foods. Certain types of polymers i.e. PVC, PVDC, vinyl chloride copolymers (for example, vinyl chloride/vinyl acetate) and PVC blends require the addition of heat stabilizers in order to maintain their functionality. Nonetheless, other types of polymers, such as LDPE and polyamides, can retain their stability even under severe heat conditions without the presence of heat stabilizers [88, 93]. There are three types of primary heat stabilizers (i.e. mixed metal salt blends, organotin compounds, and lead compounds) and three types of secondary heat stabilizers (i.e. alkyl organophosphites, epoxy compounds, and beta diketones) [92]. In general, epoxy stabilizers are derivatives of epoxidized soybean oil (ESBO), linseed oil, and sunflower oil, most commonly found in food packaging plastics [88, 91, 92, 94]. There are other heat stabilizers, some even more effective; nonetheless, they are not recommended or considered appropriate for use in food packaging plastics due to their potential toxicity [88].

4.2.4 Slip agents

Slip compounds are responsible for significantly reducing the surface coefficient of friction of a polymer. In addition to providing lubrication to the film surface, they are also used to enhance the polymer with antistatic properties, enable better mould release, reduced melt viscosity, and anti-sticking properties [88, 90]. Some of the most commonly used slip compounds are fatty acid amides (primary erucamide and oleamide), fatty acid esters, metallic stearates (for example, zinc stearate), and waxes [88].

4.2.5 Residual or unreacted monomers and oligomers

The macromolecules that form plastic materials are created via the chemical reaction of the respective monomers. Both monomers and oligomers tend to migrate from packaging materials into foods [95]. Consequently, health risks could potentially arise when/if the concentrations of unreacted monomers or low-molecular-weight substances in food reaches a certain limit, which could potentially be absorbed by the human body [96, 97]. For instance, residual styrene from PS food packaging can migrate and may result in health issues [91]. Epoxy resins of BPA, also known as bisphenol A diglyceride ether (BADGE), have been reported to create cytotoxic effects in living tissues and may increase the rate of cell division [95]. However, recent FDA (food and drug administration) studies, in collaboration with the National Center for Toxicological Research (NCTR), have shown that the use of BPA in containers and other food-packaging materials is safe [98].

The concentration of unreacted epoxy groups determines the degree of toxicity in the respective compounds. As it is reported in the Commission Regulation (EU) No 10/2011, the vinyl chloride monomers in PVC may pose acute toxicity to the human body [97, 99] and is considered carcinogenic to humans (Group 1) [100]. Therefore, the materials contacting food must not contain vinyl chloride monomers exceeding 1 mg kg^{-1}.

BADGE is an example of a major monomer of epoxy resins used for internal can linings and added to polymers to serve as antioxidant [88]. Nonetheless, it can potentially migrate into foods during heating and storage, plus it may contain unreacted BPA. Since BPA is an endocrine disrupting chemical, low levels of exposure are of concern, too [101].

On the other hand, PET contains miniscule amounts of oligomers ranging from dimers to pentamers. For instance, cyclic compounds have been reported to be between 0.06%-1% depending on the type of PET [95].

5. Migration, release and fate of POTSs contained in plastics

Emissions of PoTSs stemming from plastic products into the air water and soil may occur in all phases of the product life cycle, as outlined in Fig. 1. Release of PoTSs from plastic products to air, extraction fluids, water, food, food simulants (FS), saliva and sweat have been identified by chemical (laboratory) analysis. Identifying the magnitude and type of such emissions is a complex task, because it depends on many factors. In general, the fate of the polymer product, any substances released, any degradation process products and their persistence in various environment and bioaccumulation potential will affect the exposure to humans and the environment, both in the short and in the long term.

Simplifying, the composition of non-polymeric substances defines what can be released from the plastic in the first place. However, other factors may also be responsible for controlling the actual potential release of such substances in a surrounding medium, i.e. the migration potential (e.g. availability vs. solubility behaviours during leaching). Moreover, there are additional aspects involved in assessing the risk posed to various types of receptors (e.g., animals, humans, habitats), because presence in the plastics or release doesn't automatically constitute hazard. Here we focus upon two prominent cases that are critical for a circular economy of cleaner material cycles [102], so that longer-term dispersion of PTEs to the environment is sufficiently mitigated and effectively controlled: (a) the migration mechanism(s) and release potential of most substances and additives existing in plastic food-contact materials; and (b) to the emission/ leaching of PoTSs during the recycling process of all types of plastics (Sections 5.2 and 5.3, respectively).

Examples of PoTSs studied for potential release from various plastic products include: phthalates [103, 104], brominated flame retardants (BFRs) [105], BPA [106-109], bisphenol-A dimethacrylate [108], lead, tin and cadmium [110], formaldehyde and acetaldehyde [111], 4-nonylphenol [112, 113], MTBE (methyl tert-butyl ether), benzene [114] and many other volatile organic compounds [115]. In several of these studies the concentrations released are low compared to the guideline/legal limit values, but there are also occasions where they are considerably higher. Notably, the guideline values do not consider the low levels at which endocrine disrupting chemicals may be in effect [101] and also do not consider the toxicity of mixtures [116].

The degradation products formed are polymer type dependant [71]. The type and quantity of degradation products may be influenced by degradation mechanisms and the presence of polymerisation impurities and/or surrounding factors, e.g. temperature and oxygen [71, 117]. During thermal degradation, nitrogen-containing plastics (e.g. nylons, polyacrylonitrile, and polyurethanes) release hydrogen cyanide; chlorine-containing materials (e.g. PVC) release hydrogen chloride and dioxins; and fluorine-containing polymers (e.g. polytetrafluoroethylene (PTFE) and polyvinylidene fluoride) release hydrogen fluoride and perfluoroisobutene by a chain stripping mechanism and other degradation pathways [71, 118]. Polymers capable of depolymerisation by chain scission include polymethyl metacrylate (PMMA), PTFE, and polyoxymethylene (POM), which can depolymerise completely into their initial monomers. Also PS, polyesters (e.g. PET and polycarbonate), nylons and polyurethanes can depolymerise to some extent into their monomers [71, 117].

5.1 Migration of chemical substances present in plastics

Chemicals present in plastics can potentially migrate from the plastic product to the medium in contact with the product and can, also, slowly migrate within the plastic to the surface.

Bhunia et al. [88] has comprehensively reviewed the migration of various chemical substances from plastic packaging materials during MW and conventional heating, under various storage conditions. Some of these studies have also been identified, presented in Table 2, and briefly discussed in the next section (Section 5.2) of the present work, too.

In some cases, migration can actually be an engineered and controlled/required process; nonetheless, in most cases it is not. An example of a desirable migration is that of mould release agents to the surface, to give a better slip to the mould or to provide antistatic properties [93]. Additionally, a controlled release of some drugs from their plastic matrix, to provide the precise desirable dosage to the patients, could also be considered a desirable controlled migration. In most cases, however, there is unwanted migration and release of additives, such as plasticizers from plastic products (e.g. from a PVC toy or shower curtain) or the migration and release of flame retardants (e.g. from plastic casings of televisions or computers). Migration of chemical substances in food or medicine plastic packaging are other examples of undesirable migration, as some of the migrating substances may be toxic or give an unpleasant taste to the food or affect the medicine or enhance the degradation of the active substances in the medicine.

The migration process can be divided into four major steps exemplified for a food contact material: 1) diffusion of chemical compounds through the polymers, 2) desorption of the molecules from the polymer surface, 3) sorption of the compounds at the plastic–food interface, and 4) absorption of the compounds in the food [119]. The mass diffusion process is usually governed by Fick's law. The steady state diffusion process indicates no change in concentration over time; however, most of the interactions between the packaging and food are determined and/or influenced by non-steady state conditions. In practice, though, the migration of substances from plastics is measured in contact experiments under "worst case" scenarios. Some methods for sensitive uses such as food contact materials and for pharmaceuticals are standardised while others set up according to the use of the plastic.

The migration rate of organic chemical substances is size dependant. Small molecules, (e.g. monomers and residual solvents), with low boiling points, will migrate fast. In fact, some monomers e.g. formaldehyde, vinyl chloride, ethylene and butadiene have a tendency to migrate quickly even at ambient temperatures [75]. The molecular weight of substances used as additives in plastics is estimated to be in the range of 200–2000 g mol^{-1}. A high molecular weight corresponds to a large molecule and, thus, a slow migration rate and visa-versa. This rule of thumb is to some extent used for the design of several antioxidants, flame retardants and plasticizers. However, the design and use of some plasticizers and flame retardants may not be based on this principal and this largely because of their historic development and use or due to the higher cost of producing and using high molecular additives. Another rule is that the solubility of the additives in the plastic should be kept at high levels, but low in the liquid (or food) in contact with the plastic. The initial concentration of the chemical substance present in the plastic, the thickness, crystallinity and the surface structure of the plastic are all factors that complicate and influence the migration rate [75].

Furthermore, the Regulation sets out 'Specific Migration Limits' (SML). These are established by the European food safety authority (EFSA) on the basis of toxicity data of each specific substance. To ensure the overall quality of the plastic, the overall migration to a food of all substances together should not exceed the limit of 60 mg kg^{-1} food, or 10 mg dm^{-2} of the contact material. The Regulation sets out, also, detailed migration testing rules using 'simulants', representative for relevant food categories, for the tests performed [99].

Table 2 presents a number of studies that have assessed the migration of various chemical substances from food packaging materials at different conditions of temperature and contact time. Some of these studies are discussed in Section 5.2. However, apart from the works reported in Table 2, more relevant research and their findings on migration of additives from treating food packaging materials either via MW heating, or under the use of various simulants, or even studying their kinetics mechanisms, are also reported and discussed in the following section.

Table 2. Migration of various chemical compounds from plastic food packaging materials under different conditions of temperature and contact time

Packaging material/ type	Migrated substance studied	Food/FS	Storage/Heating conditions		Comments - Findings	References
			Temp (˚C)	Contact time		
PS cups	Styrene	Distilled water	60, 40, 20, 4	3 d	Styrene migration was influenced by fat content and storage temperature of food, exhibiting higher migration levels in hot beverages than in the cold ones.	[120]
		Distilled water	100	1, 2 h		
		Milk	100,60,40,20	2 h		
		Milk	40, 20, 4	24 h		
		Milk	4	3 d		
		Juice	20	16 h		
		Jelly, pudding	4	1, 3, 7 d		
		Hot beverage	100	1 h		
		Drinking chocolate	20	16 h		
		Cola, beer	20	16 h		
		Ice-cream	–10	30, 60 d		
		Drinking yogurt	4	3, 7, 14 d		
PS	DEHA Styrene, Overall migration	Iso-octane	40	2 h	For overall migration isooctane is an alternative FS. For DEHA each of the FSs should be considered separately. Styrene migration was in all cases higher than ethylbenzene. In addition, longer contact time and higher fat content favored migration.	[121]
		Yoghurt, dessert	25	8-28 d		
PS, PP, PET	Relative migration	Vegetable pure oil, 3% (v/v) aqueous acetic acid, 15% (v/v) ethanol, and olive oil	5	10	PS caused the fastest migration in olive oil while PET had the highest migration in the FS 15% ethanol.	[122]
PVC	DHA,HOA, HAD, DEHA and overall migration	Sliced ham	25	0, 1, 3, 5, 10, 30, 45 min	HDA demonstrated higher migration in ham after 45 min, while the migration was found to be proportional to fat content of material and contact time.	[123]
		Olive oil	40	10 d		
PVC	Ethylbenzene DEHA	Yoghurt, dessert	25	8-28 d		[124]
		Kefalotyri, Edam and Feta cheese	5	1-240 h	Kefalotyri exhibited the highest level of migration, followed by Edam and Feta.	
PVC	DEHA	Cheese	40	2h, 1d	DEHA migration was highest at 21°C after 5d. Lowest migration was observed at 5°C after 2 h.	[125]
			21	2h, 1, 5 d		
			5	2h, 1, 5, 10 d		
PVC	DHA,HOA, HDA	Cheese	25	5 min		[95]
LDPE	Irganox-1076	Ethanol	28-60	-	No influence of the FS type on the transport properties into the plastic films were observed; thus, no absorption of the FS into the plastic tested films occurred in this work.	[126]
rPET	Toxic metals	5% aqueous citric acid or deionized water	1700 W or 7.2–22.2	5 min or 1, 7, 14 d	Neither the storage nor the MW treatments had significant effect on metal migration. Exposure to 5% citric acid resulted in a higher rate of leached metals compared to deionized water.	[127]
Melamine Formaldehyde	Overall migration	3% (w/v) acetic acid	25, 800W	1, 2, 3 or 5 min (repeated heating, cycles)	MW heating for 1–2 min over long-term use creates concern. Service terms in a MW oven were drastically reduced, by more than 10-fold, compared to conventional heating.	[128]

Packaging material/ type	Migrated substance studied	Food/ FS	Storage/Heating conditions		Comments - Findings	References
			Temp (°C)	Contact time		
Retail Packaging material	HA, DBP, BHT, Cyanox 2246, Chimassorb 81,Irganox 1035, 1010, 1330, 1076, Irgafos 168, Tinuvin 326, 328	FS-A, B, C, D	40	10 d	Low-molecular weight compounds were detected in aqueous simulants. Irganox 1010 and 1330 were found in oil stimulants.	[129]
PVC gasket	ESBO, DEHP, DINP, DIDP, DEHA, DEHS, ATBC	Oily food (Olive, mussels in oil, tuna in oil, and so on)	120, 150, 40	1, 4, 10 d 10 d	Migration of DEHA, ATBC, and DEHS was higher than ESBO.	[130]
Plastic container	Phthalates	Cooking oil and mineral water	20, 40, 60	60 d	Cooking oil proved to be a more suitable medium for phthalate migration than mineral water. Higher temperatures and longer contact time favoured migration.	[131]
Plastic vs non-plastic packaging material	BPA	Olive oil	25	1 y	Higher BPA levels were measured in oil samples stored in plastic vs. non-plastic packaging materials. Estimated exposure was 1.38% of the EFSA tolerable daily intake, thus no concerns arose of potential health risks from olive oil consumption	[132]
LDPE	DPBD	Chicken, pork	5, 25	10 d	High storage time and temperature favoured migration. No significant difference was observed between the two temperatures tested.	[133]
PVC	DEHA, ATBC	Sesame paste	25	0.5–240 h	ATBC at equilibrium was found to be approx. 2.5 times lower than DEHA which can be attributed to lower initial concentration of ATBC (1.8 mg dm^{-2}) in the film, compared to that of DEHA (3.2 mg dm^{-2}).	[124]
Cup, plate, container meat	Styrene	Oil	70	10 d	Cup has exhibited the highest migration levels of all other materials at 150°C (1.39 μg cm^{-2}) after 10 d of exposure.	[134]
LDPE, PVC LDPE, PS LDPE, PP	Oleamide, Erucamide, Stearamide	FS-A, B, C, D	40	10 d	Polyolefin exhibited the highest amount of migration. Slip compounds were almost totally migrated from 65 μm LDPE film, whereas PVC or PS exhibited miniscule migration (<1% of total).	[135]
LDPE	BHA, DBP, BHT, Irganox 1010, 1076, Irgafos 168, Ethanox 330	Distilled water	60 40±1	20 d 10 d	Of all migrated substances studied, only Irgafos 168 and Ethanox 330 were detected in FSs.	[136]
PA, PE/PA, PP	Overall migration	Olive oil, ethanol 95%	40	10 d	For PA/PE, 95% ethanol appeared to be the best alternative fatty FS. For PP, isopropanol and n-heptane yielded almost the same amount of migration.	[137]

NOTE: “-” not reported in the specific study

5.2 Migration of the most common additives in plastics

5.2.1 Migration of plasticizers

Plasticizers, being of comparatively low molecular weight (300-600 g mol^{-1}), could potentially migrate from packaging materials into food, thereby becoming indirect “food additives”. The most commonly used plasticizers in PVC, PVA, and PE, are phthalate and adipate; their migration to food, under various conditions, has been widely reported and documented in literature [124, 130, 138-140]. Migration tests are commonly performed using FSs under a uniformly contact of the packaging material with the food.

Simoneau et al. [141] investigated the phthalate migration from baby bottles (n = 277) under hot-fill conditions of 70°C, for an approx. contact time of 2 h and found that migration levels of DiBP and DBP were in the range of 50-150 μg kg^{-1}, with DEHP also having been detected but in lower migration levels (ranging from 25-50 μg kg^{-1}) [141].

Fankhauser–Noti and Grob [130] noticed that phthalates exhibited an extremely high-transfer (migration) rate (350%), when used in gasket material for closures, in a study using olive oil [130]. This indicates that transfer was not only stemmed from the gasket, but also from underneath the seal or rim.

The same team of authors, as well as Ežerskis et al. [142] studied, the migration of seven plasticizers (ESBO, DEHP, DINP, DIDP, DEHA, DEHS, and ATBC) from PVC gaskets in the closures of glass jars, when in contact with oily foods [130, 142]. The average migration rate was calculated by comparing the amount of plasticizers in direct food contact with gasket material and the plasticizers found in food. The average transfer was found to be 46%, with 90% being the highest percentage observed for ESBO.

Li et al. [143] studied the migration of 5 phthalates (BBP, DBP, DEHP from disposable tableware (simulating the normal use of plastic cups -among others- as drinking utensils) to drinking water using hexafluoroisopropanol-induced cationic surfactant coacervate extraction.

Concentrations of DBP and DEHP in the drinking water samples (10.13 ng mL^{-1} and 5.83 ng mL^{-1}) exceeded the limit levels for drinking water (8 ng mL^{-1} and 3 ng mL^{-1}, respectively) regulated by some of the relevant known organizations [144-146].

Fasano et al. [139] studied the migration of phthalates, BPA, DEHA and alkylphenols, from PE and PS food-packaging materials to various FSs (3% acetic acid, distilled water, and 15% ethanol) after 10 d of storage at 40°C. PE bread-bag exhibited the higher amounts of released plasticizers, compared to PE film, whereas low levels of PAEs and DEHA migrated from tetra pack packaging materials. The PS packaging for yogurt demonstrated very low DMP migration, but higher amounts of DEHA [139].

Xu et al. [131] evaluated the migration of 8 PAE compounds (DMP, DEP, BBP, DBP, DEHP, DINP, DOP, and DIDP) from plastics to a) cooking oil and b) mineral water, under various storage conditions. Storage times tested were up to 2 months, under several static conditions (20°C, 40°C, and 60°C) and under a "dynamic" state (20°C). For the dynamic state, the packaged FS was treated at a frequency of 50 times per minute, for 5 minutes per day, for a total period of 2 months. The PAE content was always measured in higher levels in cooking oil than in mineral water. DBP and DINP demonstrated the highest migration into the mineral water. DEHP and DBP displayed the highest level of migration into cooking oil at 20°C after 2 months. It was, thus, concluded that the dynamic process favours the migration of the compounds more than it does in the static state. PAE migration into cooking oil (fatty food 1% to 14%) was found to be higher than into mineral water (aqueous food < 0.35%) [131].

Lau and Wong [95] assessed the migration of 3 plasticizers under different fat content and contact time after MW heating. DHA, HOA, and HDA from "cling" film (0.02 mm thick PVC film plasticized, respectively, with DHA, 2240 μg dm^{-2}; HOA, 2680 μg dm^{-2}; and HAD, 2550 μg dm^{-2}) into cheese and ham. Cheese with different fat contents (8.2%, 12.5%, 21.3%, and 32.8%) and ham slices were analysed in this study. Migration of all plasticizers increased with increasing fat content and contact time. Adipate plasticizers migrated from the packaging film into these foods (within 22% migration percentages) with migration rates observed in foods increasing proportionally to the increase of fat content [95, 124, 147, 148].

Badeka and Kontominas [149] studied the effect of MW heating on the migration of DOA, ATBC from food-grade PVC, and PVDC/PVC (Saran) films into olive oil and distilled water. Results showed that migration of DOA into olive oil reached at a steady state (equilibrium) (604.6 mg DOA/L) after heating for 10 min at 700 W [149]. Migration of DOA and ATBC during MW heating was found always higher for olive oil compared to water, under similar conditions. Migration was also observed at room temperature after 20 min of contact without MW treatment (145.7 mg DOA L^{-1} or 15.3 mg dm^{-2}) for olive oil [149], which is above acceptable levels for global migration (60 mg L^{-1}) set by the EU [99].

It needs to be taken into consideration that several migration limits have been set from European Commission (EC) for different plasticizers, e.g. 1.5 mg kg^{-1} for DEHP, 18 mg kg^{-1} for DEHA, 0.3 mg kg^{-1} for DBP, 30 mg kg^{-1} for BBP [97]. Divinyl esters of adipic acid should not exceed 5 mg kg^{-1} of the final product and can only be used as co-monomers.

In general, it can be stated that migration of plasticizers is dependent on food composition, contacting phase, time and temperature exposure of the food to the packaging film; the initial concentration of the migrant components in the film plays also a major role [149]. PVC is not suitable for food-contact applications in a MW oven due to high migration of DOA, but Saran may be used given that direct contact with high-fat foodstuffs is avoided.

5.2.2 Migration of antioxidants

Several studies have reported quantification of migrated antioxidants and their degraded products from different polymers under various conditions [129, 136, 150-154].

Antioxidant migration has been widely studied, however, mostly in the cases of PP and LDPE [153, 155].

Garde et al. [150] evaluated the migration of antioxidants from PP films of several thicknesses into n-heptane and 95% ethanol at 20°C, 37°C and 60°C. They found that the thicker the polymer in n-heptane the higher the migration rates, compared to ethanol [150].

Alin and Hakkarainen [151, 152, 156] noticed that continuous heating for up to 1 h in MW favours degradation of antioxidant in FSs, when compared to conventional heating using oil bath. The experimental temperature was kept at 80°C for both MW and conventional heating. However, this 1 h of MW heating in contact with food is not relevant for industrial or domestic applications. High temperature caused more swelling of PP in isooctane during MW heating and increased the diffusion coefficient by factors of 100 to 1000. They also observed that aqueous soluble antioxidants tend to migrate into aqueous FSs [151, 152, 156].

Gao et al. [129] studied the migration of 8 antioxidants: BHA, BHT, Cyanox 2246, Irganox 1035, Irganox 1010, Irganox 1330, Irganox 1076, Irgafos 168 and its degradation product DBP, at 40°C in a 10 d storage experiment, under various simulants. BHA, DBP, BHT, Cyanox 2246, Irganox 1035 migrated into aqueous simulants in respective concentrations <LOQ, ≤14.43 μg g^{-1}, ≤706.3 μg g^{-1}, ≤20.68 μg g^{-1}, ≤2.03 μg g^{-1}; Irganox 1010 and Irganox 1330 were detected in oil simulants in concentrations within the following respective ranges: 20.28-330.44 μg g^{-1}, 3.08-47.31 μg g^{-1}, whereas BHT was not detected at all [129].

Beldí et al. [153] studied the effect of fat content and storage temperature on the migration of Irganox 1076 from LDPE to several foods (cheese sauce, chicken, chocolate, margarine, mayonnaise, milk, orange juice, soft cheese, pork, salmon, and wheat flour) and FSs (distilled water, 3% acetic acid, ethanol 10%, rectified olive oil, isooctane, and 95% ethanol). They concluded that migration tend to increase, with increasing fat content of the food and storage temperature. The highest level of migration (1413 µg dm^{-2}) was observed in chocolate (32.1% fat) at 40°C after 30 d of storage [153].

Reinas et al. [154] compared migration kinetics of antioxidants (Irganox 1076 and Irgafos 168) into precooked white rice and TenaxR at 23°C, 40°C, and 70°C, and found that migration into rice is slower than into Tenax due to the lower porosity and adsorption capacity of rice [154].

Linssen et al [157] found that migration of antioxidants Irganox 1076 and Irgafos 168 increased with increasing concentration (40% to 100%) of ethanol in FSs, with the highest percentage of migration to 100% in ethanolic simulants [157].

Noguerol-Cal et al. [158] concluded that migration concentration values of antioxidants BHT and Irganox 1010, measured in the analysis of commercial toys, are lower than their specific migration limits regulated in the Directive 2002/72/CE for food packaging [158, 159].

5.2.3 Migration of monomers and oligomers

Several studies have reported the migration of styrene into food [120, 160, 161] and have estimated the daily styrene exposure at 18.2-55.2 µg for individuals, with an annual exposure of 6.7-20.2 mg. This level of exposure causes irritation to the human organs and skin, as well as neurological disorders [91].

Lickly et al. [134] studied the migration of styrene from several food-contact PS foam materials (meat trays, egg cartons, cups, plates, and hinged carry-out containers) to oil (mixture of canola, sunflower, and other vegetable oil) and 8% ethanol, under 21°C for 10 d, 49°C for 4 d, and 65.5°C for 1 d. Migration followed the Fickian diffusion model, with an increasing tendency, and was found to be proportional to the square root of the increase in time at a specific temperature, for all materials except for drink cups. [134].

Other researchers have reported increasing styrene migration with increasing fat content [120].

Paraskevopoulou et al. [162] noticed higher migration levels of styrene in ethanolic solutions, compared to isooctane, with no styrene at all found in aqueous FSs [162].

BPA can potentially migrate from plastics resins that are commonly used as can linings and polycarbonate (PC) bottles into food, acting as an endocrine disruptor [163] causing, in turn, developmental and neurological impacts. Several studies have investigated BPA release from can linings and PC bottles [164-166].

Goodson et al. [164] conducted a study to examine how storage conditions and can-denting influence BPA migration into 4 different food-media products: minced beef in gravy (20% fat), spring vegetable soup (0.3% fat), evaporated milk (8% fat), carrots in brine (0% fat), and a FS (10% ethanol) [164]. Filled cans were processed at 121°C for 90 min prior to storage. The cans were stored for up to 9 months, at 5°C and 20°C to represent chilled and ambient storage conditions, respectively. In addition, to simulate 3 y of storage, cans were stored at 40°C for 10 d to 3 months. The amount of migrated BPA from the can coating into the food (during processing for 90 min at 121°C) was found to be quite high (80% to 100% of the total BPA present in the can coating). The migrated amount of BPA into 10% ethanol (68.3 ± 9.0 $\mu g\ kg^{-1}$) was significantly higher than in the other foods (minced beef: 53.8 ± 7.6 $\mu g\ kg^{-1}$, milk: 49.8 ± 10.9 $\mu g\ kg^{-1}$, carrots: 47.2 ± 5.1 $\mu g\ kg^{-1}$, soup: 45.7 ± 5.0 $\mu g\ kg^{-1}$) [164]. This may be attributed to solubilisation of ethanol with the can coating during processing. Finally, results indicated that can damage did not play any role in the migration of BPA.

Kubwabo et al. [165] studied the migration of BPA into water, 10% and 50% ethanol using a PC and other plastic containers (PC baby bottles, non-PC baby bottles, baby bottle liners, and reusable PC drinking bottles). They reported that higher temperatures and longer treatment periods resulted in higher BPA migration from PC bottles. The average concentration of residual BPA in 50% ethanol was higher (2.39 μg L^{-1}) compared to water (1.88 μg L^{-1}) at 40°C after 240 h [165].

In another study on PC baby bottles, Nam et al. [166] demonstrated that experimental extraction at 40-100°C up to 100 times resulted in concentrations of BPA migrating from brand-new PC baby bottles, ranging from 0.03 ppb and 0.13 ppb, at 40°C and 95°C, respectively [166].

Begley et al. [167] investigated the migration of caprolactam (residual monomer) and oligomers from nylon 6 and nylon 6/66 polymer into oil, under a 30 min treatment at 176°C (representing almost oven cooking conditions). The total amount of nylon 6/66 oligomers migrating after oven heating (176°C for 30 min) was 15.5 μg g^{-1}. This was equivalent to almost 43% of the total oligomers present in the packaging polymers [167].

Bomfim et al. [168] studied the migration of ε-caprolactam from nylon 6 packaging to 95% ethanol. Packages were kept at 72-100°C for 1-4 h. A total of 40 samples were analysed, including poultry breast (n=2), ham (n=9), pâté (n=3), turkey blanquettes (n=3), and bologna sausages (n=23). The results indicated that migration of ε-caprolactam exceeded the EU limit of 15 mg kg^{-1} [97] in 35% of the bologna sausage packaging, 33% of the turkey blanquettes packaging, 100% of the pâté packaging and 100% of the poultry breast packaging [168].

Munguía-López and Soto-Valdez [169] investigated the potential migration of BPA and BADGE from 2 types of cans; one made for tuna fish and the other from jalapeňo peppers, into distilled water. The results indicated that migration of BPA from tuna cans is storage time independent. However, an increase in BPA migration from jalapeňo pepper cans was observed during the storage period. BADGE migration during the storage was found to decrease over time due to its instability and the fact that it hydrolyzes in the aqueous medium. The level of migration for BPA and BADGE were within 0.6 to 83.4 and <0.25 to 4.3 μg kg^{-1}, respectively, which is below the level set by EU 10/2011 [97, 169].

PET is known to contain small amounts of low-molecular weight oligomers of cyclic compounds ranging from dimer to pentamer. PET oligomers showed less migration (29% of the available unreacted/unpolymerized oligomer in PET) compared to nylon (43% of total oligomers) [167].

Mountfort et al. [170] studied the migration of PET cyclic trimers from impregnated susceptors used for pizza, pasty, popcorn, and French fries after conventional oven and MW heating. Trimmers were detected only in pizza (after the application of both heating methods), with a higher migration (7.4%) than other marker substances. It was then concluded that oil is not considered a top FS choice for MW-treated foods, since absorption into packaging material may be too high to provide reliable results [170].

Castle et al. [171] studied the possibility of PET migrated oligomers from plastics to various foods and beverages, under several MW and oven conditions. The abundant factors of the overall migration procedure were temperature and exposure times. MW heating exhibited lower migration compared to oven heating due to shorter exposure time (maximum 15 min for MW, maximum 80 min for oven) [171].

5.2.4 Migration of light stabilizers

Light stabilizers are used for the protection of plastics from sun and weather exposure. Polyolefins are susceptible to UV light, O_2, moisture, and heat, resulting in polymer brittleness, surface crazing, colour change and product failure. Polyolefins usually contain hindered amine light stabilizers (HALS) such as Tinuvin 622, Tinuvin 765 and Chimasorb 944 [95, 172].

Monteiro et al. [173] investigated the migration of Tinuvin P from PET bottles into fatty-FSs (olive oil, soybean oil, n-heptane, and isooctane) at 40°C, for a period of 2-10 d. The stability performance of different UV stabilizers (Cyasorb UV 5411, Tinuvin P, Tinuvin 326, and Tinuvin 327) in n-heptane and isooctane was also assessed. Migration demonstrated a quick increasing tendency up to 10 d of storage. Olive and soybean oils favoured the migration of Tinuvin P compared to n-heptane. Isooctane was found to be a more suitable fatty-FS than n-heptane with similar migration levels as olive and soybean oil [173].

Begley et al. [174] studied the migration of Tinuvin 234 (T234) from PET into Miglyol, water-ethanol solutions, and isooctane. They concluded that the migration of Tinuvin 234 from PET is very slow. The obtained migration data (2 µg dm^{-2}, at 40°C in 95% ethanol) were almost comparable to the amount of migration (3 µg dm^{-2}) in olive oil and soybean oil found by Monteiro et al. [173]. They also reported that similarly natured polymer and foods (i.e., polar solvents in contact with polar polymer) may result in faster migration, which is evident from the obtained value of diffusion coefficients. The diffusion coefficient of T234 from PET with isooctane ($D = 3\times10^{-16}$ $cm^2\ s^{-1}$) was found to be less than the values obtained with ethanol at 60 °C ($D = 1\times10^{-14}$ $cm^2\ s^{-1}$) [174]. Both Ethanol and PET are considered as polar substances in this case, while isooctane can be a substitute to the fatty FS Miglyol.

5.2.5 Migration of slip additives

The most commonly used slip additives in plastics are fatty acid amides such as oleamides, stearyl erucamide, stearamide, erucamide and oleyl palmitamide. They usually act as lubricants, thus preventing films from sticking together [95].

Cooper and Tice [135] studied the migration of 5 fatty acid amides: oleamide, erucamide, stearamide, stearyl amide, and oleyl palmitamide from 4 polymer materials (LDPE, PP, PS, and PVC) was determined at 40°C after 10 d of storage [135]. The highest migration occurred from LDPE to olive oil. Additive migrations from LDPE were: 88% for oleamide, 98% for erucamide, and 95% for stearamide, respectively. The highest slip additive migration was measured in 65 µm LDPE film, compared to PVC or PS films (<1% of the total compounds). The different migration levels observed can be attributed to the low solubility of fatty acid amides in LDPE and the high penetration rates into LDPE polymer, which increases diffusion rates [135].

5.3 Recycling of plastics: emission, release and fate of additives/other PoTSs

All four levels of plastic recycling processes - primary and secondary mechanical recycling, chemical depolymerisation and thermal recovery - are currently implemented worldwide to a different degree in order to recycle and recover plastic waste. Each of the technologies can have different impact on the environment and on human (occupational/public) health which may also depend on factors such as, geo-spatial characteristics, socio-political aspects and regulatory framework. In particular the use of material that has been recycled from plastic waste by non-environmentally sound technologies and open burning in developing and transition economies, especially in Asia and Africa, result in environmental and human pollution. Furthermore technical and economic difficulties in recycling plastics may also involve, lack of fiscal incentives, poor waste separation, high energy cost, contamination by other materials, difficulty in cleaning process, unstable economic market, etc. [175].

Luijsterburg and Goossens [176] reported that the collection method for the plastic packaging waste has hardly any influence on the final quality of the recyclate; however, the sorting and reprocessing steps influence the final quality of the recyclate. Although the mechanical properties of recyclate are often considerably different from those of the virgin polymers, improvement to the sorting and reprocessing steps can improve the quality [176]. A particular challenge is the transfer of certain groups of additives, which contain PoTSs, into new recycled products often with more sensitive use areas. For instance, brominated flame retardants including POPs, phosphorous flame retardants and phthalates have been found in children toys from recycling [20, 177, 178]. BFRs have also been detected in food contact materials and household products [22, 179]. Within the Stockholm Convention process ratified by 180 countries a BAT/BEP (best available technique/best environmental practice) guidance has been developed addressing the recycling, separation and management of POP-BFR containing plastics [12] which might improve this situation.

However, the major challenge with attempting to save resources and maintain the value of used materials during recycling of plastics is the high heterogeneity of the polymers present in many plastic waste products or in the mixed way they are collected [28]. The first compilation of separation technologies and approaches described in the Stockholm Convention PBDE BAT/BEP guidance and the related implementation might lead to an improvement also on this problem [28].

Some additives have direct impact on the recyclability of plastics [180] or even might support the degradation of plastic. One concern is e.g. the potential of several metal-containing additives to form pro-oxidants and photo-oxidation catalysts, which promote the degradation of plastics during reprocessing (melting/extruding) or even during their use-life phase [181]. In particular, metal salts or oxides such as Fe_2O_3, Cu_xO and ZnO have been found to act as pro-oxidants [181].

Moulding and extrusion are key stages in the mechanical material recycling process of plastic waste that usually is operated at 200-300°C. In this temperature area a range of hazardous substances (e.g. toxic metals, volatile organic compounds (VOCs), phthalates, polycyclic aromatic hydrocarbons (PAHs), PBDEs, PAEs, polybrominated dibenzo-p-dioxins and furans (PBDD/F) may be released from this process with associated exposure [180, 182-185].

Pivnenko et al. [180] studied a number of selected phthalates in samples of virgin, waste and recycled plastics, and concluded that DBP, DiBP and DEHP had the highest frequency of detection in the samples analysed, with 360 $\mu g\ g^{-1}$, 460 $\mu g\ g^{-1}$ and 2700 $\mu g\ g^{-1}$ being the maximum concentrations measured, respectively [180].

He et al. [182] analysed the VOC emission characteristics, health risks, and indoor microenvironment exposure during the melting/extrusion stages of the recycling processes at seven different types of plastic solid waste (PSW). The first group, consisted of both ABS and PS, contained the same monomer; styrene. The total concentration of VOCs (TVOC) with a mean value of $1.0 \pm 0.4 \times 10^3$ mg m^{-3} in the ABS recycling workshop was much higher than that in the PS workshop ($4.7 \pm 1.0 \times 10^2$ mg m^{-3}). Nevertheless, mono-aromatics was the predominant group in both workshops (≥84.7%). The second group of PSW included PE and PP, whose monomers were aliphatic olefins. Results indicated that alkanes are the most abundant VOCs for polyolefins, contributing 50.8% and 37.5% to the PE and PP recycling VOC emissions, respectively. The third group of PSW included PVC, PA and PC, whose monomers contained heteroatoms. During the extrusion of these three types of PSW, the TVOC emissions were also much lower than those of the ABS and PS recycling processes, but not so much different from the PP and PE recycling processes.

In general, VOCs could be emitted from polymers and additive pyrolysis at operating temperatures, and the types and concentrations of VOCs emitted mainly depended on the plastic composition during the extrusion process [182]. A health risk assessment that was also performed to evaluate the results of the aforementioned study indicated that for the non-cancer risk, benzene, toluene, ethylbenzene, styrene, methylene chloride and trichloroethylene were the major contributors to the chronic health effects in these workshops; while acrylonitrile, styrene, ethylbenzene and 1,2-dichloromethane were the major contributors to cancer risks, like tumour of the lungs, liver, kidneys, and brain via inhalation exposure [186, 187].

Huang et al. [183] demonstrated that the exhaust gases emitted from plastic waste recycling granulation have an effect on the ambient environment in Xingtan, Guangdong, China [183]. Also PAHs were detected inside and outside of the recycling granulation plants in the area. In the same study, PAEs were largely distributed in the particle-phase. High levels of DBP and DEHP could be detected inside the plants. The detected DiBP, DnBP and DEHP inside the Huachang plant were 30, 20 and 5 times greater than background concentrations of the area, respectively. Despite there is no standard for PAEs emitted from plastic waste recycling plants, the occupational health effects on workers should be further considered and evaluated because of their long term exposures [183].

Many metals such as Cd, Pb, Sb and Sn (as organotin) that have been used as plastic additives, have now been found to be toxic. According to the present restriction of hazardous substances (RoHS) directive, plastics containing Cd, Pb, Hg and Cr^{6+} may not be recycled if the content is higher than 1000 ppm of Pb, Hg or Cr^{6+} or 100 ppm of [188]. Such regulation control the levels of toxic metals in electrical and electronic equipment (EEE) in the EU and some other industrial countries where, also, the treatment and recycling of e-waste including plastic is controlled. However, assessments on the release of these toxic metals into the environment in areas where plastic waste recycling is carried out by non-environmentally sound methods, and the potential ecological and human risks of such releases may be high, are still under scrutiny [189, 190].

Tang et al. [191] demonstrated that the surface soils and sediments have suffered from moderate to high Cd and Hg pollution. The mean concentrations of Cd and Hg were 0.355 and 0.408 mg kg^{-1}, respectively, in the soils and 1.53 and 2.10 mg kg^{-1}, respectively, in the sediments [191].

Tang et al. [192] reported that in road dust samples collected from an area where intense mechanical recycling of plastic wastes occurs (Wen’an, north China), PBDE concentrations were found to be 1–2 orders of magnitude higher than concentrations in outdoor or road dusts from other areas. This indicated that plastic waste processing is a major source of toxic pollutants in road dusts in that area.

Song and Li [189] reviewed the reported in literature effects from recycling activities of “e-waste” (mostly known as Waste Electric and Electronic Equipment, WEEE) in China in air [193], soil [194, 195], sediments [196] and plants [194, 197]. Compared with the levels of the toxic metals in the outdoors, pollution from toxic metals indoors, specifically in WEEE workshops (formal and informal e-waste recycling enterprises) was more critical [189]. More specifically, the mean Pb concentrations in workshop dust were much higher than those from other studies the share of Pb from plastic or other source was not clarified in the studies [193, 198, 199].

Furthermore, Bi et al. [200] investigated Sb (used as synergist of BFR retarded plastic) distributions in indoor dust from 13 “e-waste” recycling villages in Guiyu, Guangdong area, in China. Results revealed significantly elevated concentrations of Sb (6.1–232 mg kg^{-1}) in dust within all the villages. There were villages where the levels appeared to be 3.9–147 times higher than those from the non-WEEE sites, indicating that WEEE recycling was an important source of Sb pollution [200].

Asante et al. [201] reported human contamination by multi-trace elements (TEs) in “e-waste” recycling site at Agbogbloshie, Accra in Ghana. Levels of Sb in workers were significantly higher compared to reference sites in urine and most likely stem from “e-waste” plastic and related releases from open burning. Also levels of As, Fe, and Pb in urine of the workers were found significantly higher than those of reference sites indicating that the recycling workers are exposed to these TEs through the recycling activity [201].

In the life cycle of flame retarded plastic, in particular the end of life treatment brominated and chlorinated flame retardants, brominated, chlorinated, and mixed halogenated dibenzo-p-dioxins/dibenzofurans, PAHs and other organic pollutants are released with associated human exposure [23].

Feldt et al. [202] studied PAH contamination in urine of e-waste recycling workers in Agbogbloshie where plastic from cables and other e-waste plastic are frequently burned. Results indicated that urinary PAH metabolite concentrations were significantly higher in individuals who were exposed to e-waste recycling, compared to controls who were not exposed to e-waste recycling activities [202]. PBDE exposure above reference dose (RfD) values have been found at e-waste sites in China [185]. High levels of PBDE were also found in human milk in Chinese e-waste sites [203].

The open burning of cables and other e-waste plastic result also in the formation and release of a complex mixtures of unintentional POPs including PCDD/Fs, PBDD/Fs, PXDD/Fs and dioxin-like polychlorinated biphenyls (DL-PCBs) documented for e-waste sites in Asia and Africa with often particular high levels of brominated and mixed halogenated dibenzofurans suggesting combustion and PBDE-containing plastics as principal sources [204-207]. The human milk from woman working in e-waste recycling had higher levels of brominated and chlorinated dioxins compared to reference sites confirming human exposure [208].

It also appears that neonates, due to the mothers' exposure to e-waste and related recycling in developing countries, are facing potential health effects, e.g. the neonates from the e-waste exposure areas have been influenced by toxic organic pollutants and toxic metals, including mental health outcomes, growth, changes in cellular expression, and DNA effects [194, 209].

The recycling of brominated flame retarded plastic also faces challenges due to restriction of some brominated flame retardants by national or international regulations such as the Stockholm Convention or the EU waste electrical and electronic equipment (WEEE) directive [28, 99, 210]. Since there is no online method to assess the type of BFR contained in a polymer the separation of only restricted BFR is currently not possible and generally challenges the recycling of BFR containing plastic. The Stockholm Convention has therefore listed an exemption for the recycling of PBDE containing plastic, should that take place under controlled conditions [12].

The industry is improving their policy and commitment towards the management of flame retardants and flame retarded products [211].

Peeters et al. [212] reported the challenges in recycling plastics containing flame retardants (FR) from WEEE. After the implementation of various tests it was demonstrated that after disassembly and plastic identification, the co-polymer poly-carbonate (PC)/acrylonitrile-butadiene-styrene (ABS) containing PFR can be recycled in a closed loop system [212]. Based on the separation efficiency of optical sorters as well as on the plastic density distributions a purity of 82% was calculated for PFR poly-carbonate (PC)/acrylonitrile-butadiene-styrene (ABS) separated from EoL LCD TVs after size-reduction (shredding). It was determined that higher waste volumes are required for a size-reduction based treatment to become economically viable [212]. Although there are some positive national and global efforts for the recycling and management of flame retarded plastic, the overall management of this large material and substance flow need significant improvements in the developing countries (Global South) [213, 214] but also in industrial countries [215].

Another trend and material flow which need also to be considered and examined is the growing production and use of biodegradable plastics (also known as bio-plastics). Whenever innovative products are developed as an alternative to conventional oil-based products, questions arise about the effective reasonableness of the proposed shift. In fact, there are various processes involved in the after-use treatment of such materials (some require specific conditions to be degraded, the majority of them cannot be recycled including e.g. starch-based plastics, etc.) that may actually increase their overall environmental impact [216]. While biodegradable plastics provide a reduction from the oil-dependent businesses and are safer in terms of non-pollutant sources, such as marine litter, nonetheless, renewability and biodegradability are not necessarily a proof of a lower environmental impact [216].

One of the most promising representative of biodegradable plastics used in packaging, characterized by high manufacturing and feedstock cost, is PLA. Hopmann et al. [217] analysed the recycling process of PLA within the context of necessary process adaptions and the effects upon ecological efficiency. The analysis of the recycling behaviour revealed that internal PLA production waste is well suitable for recycling. The influence of the recycling on the molecular weight was considered negligible [217]. Of course, like other polyesters, it can degrade at elevated temperatures under the presence of moisture by hydrolysis, whereby it loses its physical and chemical properties.

Rossi et al. [218] investigated the life cycle environmental impacts of six EoL options of two biodegradable materials, PLA and thermoplastic starch (TPS), used for dry packaging, while accounting for the dynamic pattern of greenhouse gas releases for each combination of material and EoL treatment. The results indicated that mechanical recycling is the most interesting option, followed by direct fuel substitution. Intermediate performances were obtained via anaerobic digestion and municipal incineration, while landfill and industrial composting of dry packaging generated the highest environmental impacts of the studied EoL options [218].

Gu et al. [219] investigated the environmental impacts from the mechanical plastic recycling system implemented in the eastern coast of China and run a sensitivity analysis to evaluate any potential environmental changes occurring by variations in operational parameters. They concluded that: a) specific focus should be given to the extrusion process, b) more centralised plastic recycling practices are desirable, and c) material substitution achieved considerable environmental benefit and d) the studied system should be focusing on materials with higher environmental impacts associated with initial production.

6. Conclusions

The present overview on the implications and potential environmental impact of several additives and various other PoTSs, during the use, disposal and recycling phase of plastic products highlighted the following:

- With regards to plastics entering the marine environment:

The extent of this kind of pollution, especially by microplastics, needs to be further investigated especially since it is considered to be much more prevalent than previously thought, both in terms of larger quantities, as well as of smaller particles. Various effects from the entanglement or ingestion of plastic particles, including suffocation causing death, have been reported, whereas the ability of plastics to sorb POPs may, also, cause additional problems.

- With regards to migration from food packaging products:

It is not possible to conclude unequivocally whether a particular PoTS has a higher migration potential from another, since the amount of PoTSs migrating into food depends upon its initial concentration in the packaging product. In addition, the nature of food, the food-additive interactions and time–temperature-storage conditions may significantly influence the overall migration mechanism of the additive. It is important to quantify the migration of the compound that is under investigation, depending on the toxicity level and packaging material type. Foods with a higher fat content have typically been reported to stimulate a higher level of migration. In addition, MW heating was found to be a reliable technique in food processing, causing lower migration of additives than conventional oven heating under similar processing conditions. PVC was found unsuitable for MW heating.

Given the current situation on what can be placed on the market, while in compliance with all legal requirements that ensure the safety of plastic food contact materials, it can be concluded that only a limited part of the plastic waste stream might not be suitable for recycling purposes. Such plastic materials could be either those with a not well enough defined origin or others coming from long-life applications, thereby failing to meet specific requirements.

- With regards to release and emission of additives during recycling processes:

Several PoTSs (e.g. toxic metals, BFRs, POPs and PAHs) could potentially be released by the application of various recycling techniques, especially in underdeveloped countries where the sorting-reprocessing-recycling conditions are most of the time uncontrolled; it is in fact these stages-steps that influence the final quality of the recyclate. In addition, part of the plastic waste generated in Europe comes from products that have been produced outside of Europe, as in the case of electronic and electrical devices. The possible lack of full enforcement of the applicable European regulation to these products and their constituting materials may lead to the uncontrolled presence of PoTSs in imported products and therefore waste at the end of their life. Additionally, the status of the European regulation itself, not necessarily implying the same level of requirements to products manufactured in Europe and to those produced outside of Europe, could also be responsible for the undesirable presence of PoTSs. Finally, some additives could have direct impact on the recyclability of plastics or even might support the degradation of plastic.

Recycling rates of plastic waste are likely to increase with increased regional circular economy and 3 R efforts. However, as demonstrated in this overview, there are still various environmental and technological challenges. These challenges need to be addressed so that design, use, disposal, recycling and recovering of plastic resources become environmentally sound with an aim to finally substituting a large share of virgin materials.

The presence of various PoTSs contained in plastic products and their potential negative impact on the environment and human health, imposed at all phases of the life cycle of plastic products (use, disposal, recovery and recycling) demonstrates that a proportion of these additives needs to be substituted with more green and sustainable chemicals. Material recycling and other EoL scenarios should be considered in the substitution process and should be integrated in the eco-design of products. On that, better regulatory frameworks and specifications on the use of additives during plastics production, and improved recycling approaches during plastics waste reprocessing in both developed and developing countries could result in the better and more sustainable management of this resource and its associated impacts on the environment and human health, especially when plastic material finds its way into the environment. More controlled and efficient recycling and recovery would give rise to new job opportunities and opportunities for reintegration of the currently discarded materials into the economic cycle. This would then increase the added value of products made out of recycled materials, create a sustainable solution to the polymer waste problem, and decrease dependence of businesses on oil-based raw materials and energy.

Efforts should not be limited to the optimization of recycling and recovery of materials and energy. To become truly sustainable, a substantial reduction in the use of non-renewable materials and energy in products and processes, as well as durable optimization of consumption of energy sources and fuel, still remain important challenges. These two principles of sustainable development are very general and relevant, in particular, for recycling and material isolation from waste recovery processes. Recycling technologies that consume no or small amounts of energy and do not create secondary environmental issues are regarded as sustainable recycling technologies and will be selected after performing LCA for the different treatment options [219]. Even better, if weighing up the benefits and impacts of these options, through a multi-dimensional perspective as suggested by the CVORR approach [220].

Summing up, it should be noted that the use, sorting, recovery and recycling of plastic waste still remains largely unresolved, since many fundamental issues are often overlooked, or lack solutions. If combined efforts are concentrated towards the increased use of recycled plastics as substitutes of virgin plastic material, designing, recovery, sorting and recycling of waste plastics could then gain the required attention in order to become an effective way to improve throughput and redistribution back to the supply chain, close material loops, and ensure optimal environmental performance.

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