



Review

Unpacking the complexity of the polyethylene food contact articles value chain: A chemicals perspective

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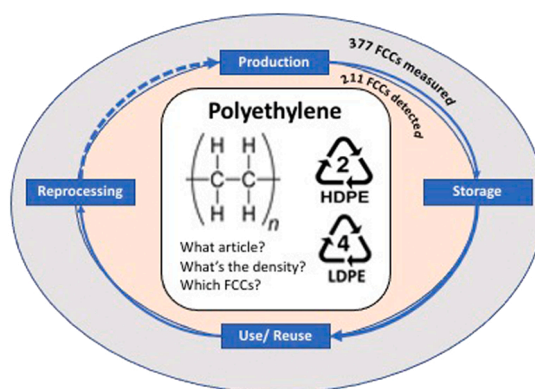
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HIGHLIGHTS

- There is a migration of FCCs across the lifecycle of PE food packaging.
- Out of the 377 FCCs measured, 211 were found to migrate from PE at least once.
- A quarter of 211 FCCs are included in the Union list, of which 25% exceeded the SML.
- One-third (53) of non-authorized FCCs exceeded the 10 µg/kg threshold at least once.
- The reprocessing stage of the PE is under-researched.

GRAPHICAL ABSTRACT



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ABSTRACT

Polyethylene (PE) is the most widely used type of plastic food packaging, in which chemicals can potentially migrate into packaged foods. The implications of using and recycling PE from a chemical perspective remain underexplored. This study is a systematic evidence map of 116 studies looking at the migration of food contact chemicals (FCCs) across the lifecycle of PE food packaging. It identified a total of 377 FCCs, of which 211 were detected to migrate from PE articles into food or food simulants at least once. These 211 FCCs were checked against the inventory FCCs databases and EU regulatory lists. Only 25% of the detected FCCs are authorized by EU regulation for the manufacture of food contact materials. Furthermore, a quarter of authorized FCCs exceeded the specific migration limit (SML) at least once, while one-third (53) of non-authorized FCCs exceeded the

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threshold value of 10 µg/kg. Overall, evidence on FCCs migration across the PE food packaging lifecycle is incomplete, especially at the reprocessing stage. Considering the EU's commitment to increase packaging recycling, a better understanding and monitoring of PE food packaging quality from a chemical perspective across the entire lifecycle will enable the transition towards a sustainable plastics value chain.

1. Introduction

Global plastic production was estimated at 390.7 million metric tonnes (Mt) in 2021, seeing an annual increase of 4% [112] due to the increasing demand for plastic packaging in Europe and North America [91]. Among plastic packaging solutions, polyethylene (PE) is the most widely used due to its good processability and excellent barrier properties [96]. PE is produced by the polymerisation of ethylene monomer with a high variety of crystalline structures, depending on its density and its chain branching. PE is commonly found in two main variations; high-density PE, known as HDPE, and low-density PE, known as LDPE. In 2021, the global production of PE was estimated at 106.6 Mt (i.e., more than one quarter of total global plastic production), and it is expected to reach 124 Mt by 2027 [111]. Considering that the global plastic packaging sector accounted for 44% of the total plastics production in 2021 [103], with an estimated value of 265 billion USD in 2021 [112], PE packaging represents a large market segment with an estimated global value of 110 billion USD [123].

In Europe, most of the plastic packaging waste generated is currently being landfilled or incinerated, and only a limited fraction is reprocessed into secondary material domestically, even though 38% of plastic packaging waste was collected for recycling in 2020 [54]. The export of plastic waste for recycling overseas has contributed to pollution with PE, specifically, being the most common type of waste found in marine and coastal environments; and 79% of total PE waste is estimated to be sent to landfills [128]. This might be attributed to PE's high use in the production of flexible plastic packaging that is considered of low monetary value and thus, less challenging to recycle, as well as to poor waste management and plastic waste mismanagement in societies where copious amounts of flexible PE are being used and imported for recycling [105]. To address plastic pollution, poor waste management, and mismanagement problems, legislative targets have been set to increase the circularity of plastic packaging. Specifically, the Directive on Packaging and Packaging Waste requires that the recycling rate of plastic packaging waste should reach 55% by 2030 [26], while the EU Packaging Levy further supports the increased demand for recycled plastic packaging [47]. This mandate could soon be replaced by a new Regulation on Packaging and Packaging Waste that is proposed to make improvements in the design of plastic packaging, increase the use of recycled content in packaging and promote plastic packaging waste reduction targets a binding commitment for all the EU member states. This proposal is envisaged to contribute to the EU's efforts to create a resource-efficient, clean, and growing economy with zero net-carbon emissions by 2050 [49].

Increasing the recycled content in food contact materials (FCMs) such as plastic packaging may pose a safety challenge that needs urgent attention because recycling processes may introduce unknown and/or hazardous chemicals that can potentially migrate from recycled FCMs into food, creating safety concerns [32,45,64,67]. FCMs represent a relevant pathway of chronic human exposure to substances of high concern arising from chemical migration into packaged and processed foodstuffs [66,95], such as phthalates from pastry [13] and plastic food packaging [63] and Bisphenol-A (BPA) from polyethylene terephthalate (PET) bottles [43]. Although thousands of chemicals are intentionally used in the manufacturing of FCMs [69] and thousands of chemicals have been shown to migrate from FCMs into food [68] (see details in the respective references), there are limited insights into the safety of FCMs due to hazard data gaps [69]. This skews our understanding of the enormous number of the often-unknown chemicals found in finished

FCMs and their fate across FCMs' entire lifecycle, which in turn leads to insufficient policy attention and intervention.

Existing research on the migration of chemicals from finished plastic FCMs has focused on specific, well-known chemicals, such as bisphenols, phthalates, mineral oils, and heavy metals (a list of studies is provided by [68]). While there are several overviews on chemicals that can potentially migrate from plastic FCMs [59,68,70,73,100], polymer-specific studies that offer insights into FCCs migration from the polymer to food samples or simulants are limited. Considering this knowledge gap, this study offers an in-depth analysis of FCCs migration from PE to inform on PE-FCMs safety.

The study builds on ongoing work that looks into unpacking the complexities in plastic value chains, exploring the challenges of plastic packaging used in food contact applications through their entire lifecycle and focusing on chemical safety and sustainability [64]. Specifically, it provides a deep dive into the available evidence on chemical migration across the lifecycle of PE food packaging, focusing on three key objectives, as follows: i) listing all chemicals that have been measured and detected in migration experiments conducted with PE food packaging, ii) assessing the relation of these chemicals with the characteristics of different types of PE, and iii) evaluating the conditions and factors that may affect the migration of chemicals across the entire lifecycle of PE food packaging. Finally, considering the evidence compiled from a system perspective, recommendations for the future are provided to improve the safety of recycled PE for food contact.

2. Legislative framework

Food contact materials (FCM) employ a large variety of polymers, including PE, thanks to their diverse functional properties, that cater to different food types, storage conditions, shelf life and the supply chain involved [118]. At the EU level, all FCMs need to comply with the framework Regulation (EC) 1935/2004, which requires all packaging materials, including bio-based, bio-degradable, or compostable FCM, to be inert and to follow and comply with good manufacturing practices according to [25] (Article 3, EC 1935/2004).

At present, the EU regulatory framework on recycled plastic FCM is covered by [28] and repealing Regulation (EC) No.282/2008. This regulation came into force in October 2022 [48] and aims to ensure the safety of recycled plastic FCMs by setting rules applicable to recycling processes and particularly to decontamination processes, as well as to quality control of recycled FCMs [48]. As a rule, recycling processes with a positive European Food Safety Authority (EFSA) opinion can be implemented to produce recycled food-grade plastics [48]. In addition, [28] allows for novel recycling technologies to operate prior to a safety assessment by the authorities, and to place recycled plastics on the market for use in FCMs [48]. A declaration of compliance is necessary from the moment a substance, mixture or (bio)plastic material is intended to be recycled. Each manufacturer has the responsibility to declare compliance with the manufacturing steps [27,46]. However, various sources of contamination might lead to unknown and unpredictable chemicals that can potentially migrate from recycled FCMs across the several steps of the recycling process, indicating the need to find a means for their identification and better monitoring [118].

So far, a high number of recycling processes have been evaluated favourably by EFSA for PET (ca. 200), while for polyolefin (i.e., PE and polypropylene, PP) FCMs there are only two – one for HDPE bottles [2] and one for HDPE and polypropylene (PP) crates [12]. The approaches used to assess the decontamination efficiency of a recycling process for

PET cannot be used for polyolefins due to the higher diffusion coefficient of a given substance in polyolefins than in PET [102]. Also, the US Food and Drug Administration (FDA) has developed criteria to guide the use of recycled plastics in the food packaging sector [120], along with specific guidance for the industry together with chemical considerations [121]. However, the criteria for polyolefins are vague since important aspects of recycled plastics such as microbial contamination and structural integrity were not discussed, and also outdated as they are based on the original FDA document that was issued in 2006 [14].

Finally, it must be noted that the Council Directive (EU) 2019/904 on the reduction of the impact of certain plastic products on the environment (single-use plastics) has released rules and implemented goals of the European Green Deal and EU plastics strategy, requiring that all plastic packaging placed on the EU market are reusable or easily recycled by 2030 [11].

3. Methodology

3.1. Systematic evidence map: literature search

Evidence from the scientific literature on PE food packaging was selected from within a larger systematic evidence map that led to the development of a database on migrating and extractable food contact chemicals (FCCs), known as FCCmigex [68]. Specifically, FCCmigex collated information from 1353 studies [68] according to a published protocol developed by [92]. The synthesis of FCCmigex was based on a set of eligibility criteria. These are [68]: (i) including studies, in which the design of the experiment was properly performed in order to clearly identify the FCM as the source of the chemical; ii) including studies, in which the analysis enabled the identification of the chemical with appropriate confidence; and iii) excluding studies, in which the experimental evidence on the measured FCCs in food did not indicate the FCM as the source.

Definitions of some key terms widely used in this systematic evidence map are provided below:

- **PE articles:** PE products or items, which intentionally come into contact with food, such as bottles, storage containers, films, bags, packaging, tableware, and cooking utensils.
- **Food contact chemicals (FCCs):** chemicals which are either intentionally added substances (IAS) or non-intentionally added substances (NIAS) that can be present in FCMs and potentially migrate into food or food simulant [64].
- **Migration:** transfer of an FCC from an FCM or food contact article into food or food simulant under realistic, intended use and foreseeable conditions [92].
- **Intentionally added substances (IAS):** FCCs that are intentionally used at the stage of FCMs production including main constituents of the polymer chain (i.e., monomers), catalysts, and/or additives [92].
- **Non-intentionally added substances (NIAS):** FCCs emerging from impurities, reaction products during FCMs manufacturing, and/or polymer and additive degradation [64].
- **Inventory lists [79]:** the Food Contact Chemicals database (FCCdb) includes 12,285 IAS used for FCMs manufacturing [59,69] and Chemicals associated with Plastic Packaging database (CPPdb) that included 4255 FCCs associated with plastic packaging [70].
- **Union list, also known as positive list:** a list of 1072 authorised FCCs that can be used in the EU in plastic FCMs manufacturing under Specific Migration Limits (SML) set by (Commission Regulation (EU) No 10/2011). For FCCs included in the Union list without a dedicated SML migration should not exceed 60 mg/kg food or 10 mg/dm² (expressed on a contact area basis) (Commission Regulation (EU) No 10/2011).

3.2. Systematic evidence map: eligibility criteria

This work was based on the results of the latest update of FCCmigex [68], using two additional eligibility criteria to select evidence related to the chemical migration from PE food packaging. These are:

- i) Focus on migrating FCCs from PE food contact articles, excluding extraction experiments. This is because extracted chemicals have an uncertain potential to migrate while migrating directly [114].
- ii) Excluding studies that assessed only the multi-material or multilayer FCMs containing PE. This is due to challenges of correctly identifying the source of migration.

It should be clarified that using the above criteria, has led to the inclusion of some eligible studies that also determined the migration of chemicals from multi-material/multilayer PE-FCMs (e.g., plastic laminates, coated films, or PE cap of a PET plastic bottle), among other PE articles. Multi-material/multilayer-related data from these studies were thus included for comparison purposes (Section 4.1.2 in Fig. 1). Aside this comparison, data on multi-material/multilayer FCMs containing PE were excluded from the quantitative analysis.

3.3. Data processing and analysis

Data were extracted from the FCCmigex database to compile a new database of all FCCs migrating from PE articles into food simulants or food samples. This dataset has been constructed in an analogous way to our previous work on the migration of FCCs from PET bottles [64], i.e., recording detailed information on the type of FCCs, migration conditions and properties of PE articles across their lifecycle (see [Supplementary Material \(excel file\)](#)). In particular, the new database provides the following information:

- Name of FCC along with its Chemical Abstracts Service (CAS) number, if available.
- Relation of FCCs to inventory lists, i.e., whether they are included in the lists and related to PE according to the lists.
- Relation of FCCs to the Union list, i.e., whether they are authorised, including the reference number of the substance as given in the Union list (FCM No) and its SML if available (Commission Regulation (EU) No 10/2011). In addition to the Union list, the list of metals as an extension of the Union list has set SMLs for metals specified by the Commission Regulation (EU) 2020/1245. The relation of metals determined in migration experiments to the list of metals was also recorded. Although not all eligible studies were conducted in the EU, the determined FCCs migration was assessed based on the EU regulation for FCMs.
- Initial concentration of FCC (if available) in PE articles before migrating.
- Measured migrated concentration and the chemical analysis method including the limit of detection (LoD).
- Number of samples analysed during FCC migration that is given – specifying if the number refers to technical replicates (i.e., several measurements from the same sample) or distinct samples.
- Migration conditions (e.g., migration duration, temperature and other factors that may be investigated during migration testing) also including the type of food simulant or a food sample.
- The composition of the PE article, i.e., whether it was a monolayer consisting exclusively of PE (i.e., single-layer PE), a multilayer consisting of layers from different polymer types including also PE, or a multicomponent formed by plastic components of different polymer types (e.g., beverage bottle of which the main body was PET and the bottle cap was PE).
- The density of PE article expressed in LDPE or HDPE; if it was not specified by researchers, it was reported as PE.

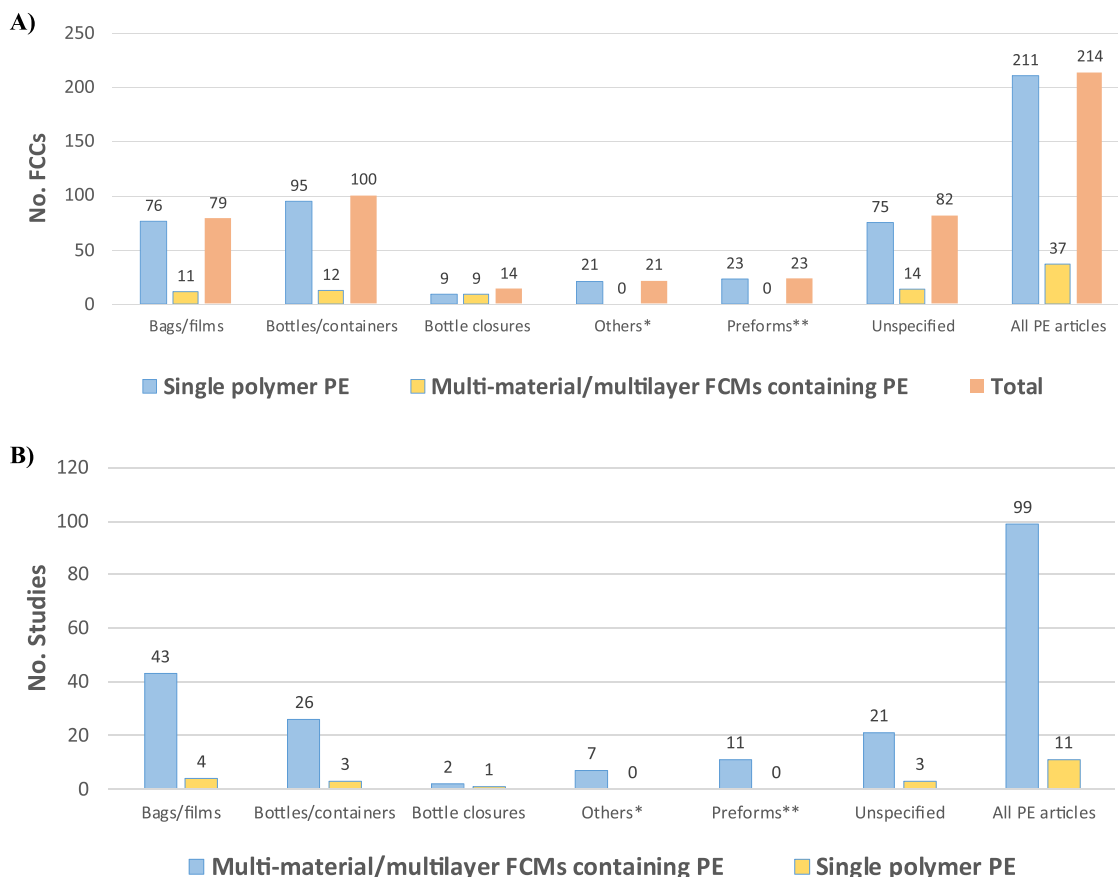


Fig. 1. FCCs migration from PE articles at detectable levels grouped by article type, form, i.e., single-layer PE articles and multi-material/multi-layer articles containing PE, and the sum of all PE-containing articles: **A)** Number of FCCs migrated at detectable levels at least once; and **B)** Number of studies that detected FCCs migration at least once. *Crate, jug, canister, cups, cutting boards; ** Granulate, beads, strip, sheet, pellet, plaque.

- Type of PE article as reported by the study, e.g., film, bag, bottle, packaging, specifying whether the PE was recycled or virgin; if this information was not provided, we assumed that PE was virgin.
- Purchase location of the PE article if available.
- The lifecycle stage covered by the study depends on the migration conditions and whether the material was recycled or virgin. For example, the investigation of storage conditions refers to the stage of storage and distribution, or migration experiments in virgin PE without the consideration of storage time and duration refer to the stage of production, or migration experiments in recycled PE provided insights on the stage of reprocessing.
- Graphical analysis of this dataset provided insights into: i) the most frequently investigated FCCs, ii) the relation of FCCs to inventory lists and regulatory authorization (Union list), iii) the FCCs that exceeded regulatory migration limits at least once, and iv) the interrelation of FCCs migration to the use and properties of PE articles. An in-depth analysis of FCCs' migration across the lifecycle of PE articles was subsequently performed to obtain insights on the aspects that gained research attention, as well as on research gaps.

4. Results

Findings are presented under two sub-sections: (1) an overview of FCCs migration from PE articles considering PE properties, existing FCCs databases and regulatory limits (Section 4.1); and (2) factors and processes that may affect FCCs migration across the PE articles' lifecycle (Section 4.2).

4.1. Overview of literature findings on FCCs migration from PE articles

In total, 116 studies from 1994 to 2022 have determined the migration of FCCs from PE plastic food packaging (both single and multi-layer) leading to the determination of 394 FCCs in total, of which 377 FCCs were measured exclusively in single-layer PE articles. Section 4.1.2 includes all FCCs reported to migrate from both single and multi-layer PE for comparison purposes. Only the 377 FCCs derived from single layer PE articles are reported and discussed in Sections 4.1.3–4.2.4). Of these 377 FCCs, 211 were detected to migrate from PE articles into food or food simulants at least once, across 99 studies (more information can be found in the Supplementary Material (excel file)); in detail, 80 of 211 FCCs have been detected to migrate into food by 39 studies, and 175 of 211 FCCs have been detected to migrate into food simulants by 80 studies.

4.1.1. Most frequently determined FCCs migrating from PE FCMS

Table 1 presents the most frequently investigated FCCs. Phthalates, such as di(2-ethyl hexyl) phthalate (DEHP) and dibutyl phthalate (DBP), have been measured in more than 15 studies; the reason is that phthalates are a group of endocrine-disrupting chemicals (EDCs) of concern even at low exposure levels [44] and their use as plasticizers in food packaging raises concerns over safety [44,17]. Because of these concerns, there has been a growing substitution of DEHP for di(2-ethylhexyl) adipate (DEHA); hence the growing number of studies measuring this substance, DEHA, as well, as current information on DEHA's effects is limited [21]. However, the molecular structure of FCC may affect their potential migration from PE, e.g., [17] reported that long-chain phthalates such as DEHP are weaker bonded compared to DBP and therefore they are more prone to migrate under increased

Table 1

The most frequently studied FCCs among the 377 FCCs whose migration from single-layer PE articles was measured across the 116 studies included in our systematic evidence map.

FCC Group	Name of FCC	CAS no.	No. of studies that measured this FCC	No. of studies that detected this FCC at least once
Phthalates	Di(2-ethyl hexyl) phthalate (DEHP)	117–81–7	17	15
	Dibutyl phthalate (DBP)	84–74–2	17	13
	Di(2-ethylhexyl) adipate (DEHA)	103–23–1	10	7
Metals	Silver (Ag)	7440–22–4	12	11
Antioxidants	Irgafos 168	31570–04–4	14	9
	Irganox 1010	6683–19–8	12	8
	Irganox 1076	2082–79–3	10	7

temperature.

Furthermore, 11 studies investigated the migration of silver (Ag) from PE articles. This high interest is due to the increasing use of silver nanoparticles in PE food packaging as a filler material or as an antibacterial and antimicrobial agent while their migration remains poorly understood [51]. A considerable number of studies focused on the measurement of antioxidants (i.e., Irgafos 168, Irganox 1010 and

Irganox 1076) that are used in PE manufacturing to improve its stability at high temperatures and under UV exposure [41,88].

4.1.2. FCCs migration at detectable levels concerning PE properties

Fig. 1A presents the number of FCCs that were found to migrate from PE-containing articles at detectable levels by article type (e.g., bag/film, bottles/containers, bottle closures, etc.) and across different forms, i.e., single-layer PE article (211 FCCs) and multi-material/multi-layer articles containing PE (37 FCCs). The figure includes the FCCs detected in unspecified PE articles, as well as the sum of FCCs detected under each article type. Interestingly, most FCCs were detected to migrate from PE bottles/containers (95 FCCs for single-layer PE) followed by bags/films (76 FCCs for single-layer PE) and unspecified PE articles (75 FCCs for single-layer PE).

Fig. 1B shows the number of studies that measured FCCs migration from either single-layer or multi-material/multi-layer PE articles, by article type. It should be noted that there were studies that analyzed more than one type of PE article. Eleven studies have measured FCCs migration from multi-material/multilayer FCMs containing PE along with single-layer PE articles (Fig. 1B). The most frequently investigated type of PE article was films/bags (43 studies) followed by bottles/containers (26 studies), while 21 studies did not specify the type of PE articles (Fig. 1B).

In the case of multi-material/multi-layer articles, it is difficult to identify the exact source of migration into food (i.e., whether the detected FCCs originate from the PE or some other material present in the food contact article). This is the reason studies that investigated only the multi-material/multilayer FCMs containing PE were considered out of scope. It should be emphasised that hereafter, results solely refer to

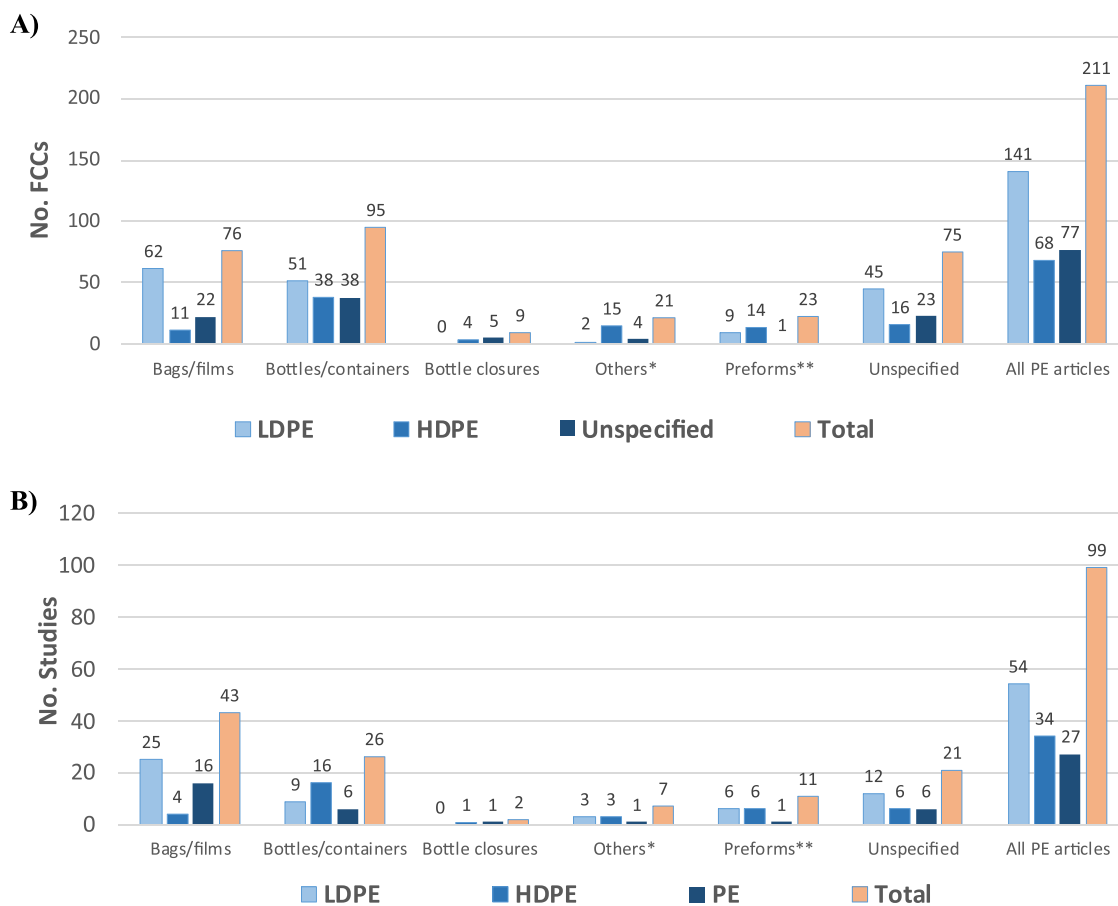


Fig. 2. FCCs migration from single-layer PE articles at detectable levels grouped by article type and density (i.e., LDPE, low-density PE; HDPE, high-density PE; and PE, when density was not specified): A) Number of FCCs migrated; and B) Number of studies that detected FCC migration. *Crate, jug, canister, cups, cutting boards; ** Granulate, beads, strip, sheet, pellet, plaque.

single-layer PE.

While, there is no evidence on the role of PE density in FCCs migration, it is worth mentioning that the density of the polymer is closely related to its molecular weight, which in turn determines its properties and therefore, potential FCC migration. That said, most FCCs have been detected to migrate from LDPE (141 FCCs), while 77 and 68 FCCs were found to migrate from unspecified PE density and HDPE, respectively (Fig. 2A). Compared to other plastic FCMs (e.g., PET), both HDPE and LDPE are considered to have higher diffusion and sorption characteristics [31,45,61]. Fig. 2B shows that more studies were conducted with LDPE (54 studies) than HDPE (34 studies), while a substantial number (27 studies) did not report the PE density. LDPE has been used as a worst-case scenario for FCC migration in many studies due to its high diffusivity [31,38,24,9,97], especially those using fatty food simulants [38].

4.1.3. Alignment with existing databases

Of the 211 FCCs detected in PE, 156 are also included in the FCCdb database [59,69], 143 are included in the CPPdb database [70], and 53 and 9 are included in the Union list (Commission Regulation (EU) No 10/2011) and the list of metals [22], respectively (Fig. 3). Only 29% of the total 211 FCCs detected are included in the Union list and the list of metals. Most FCCs found to be migrating from PE articles are therefore non-authorized substances in the EU that may be either IAS or NIAS (149 in total). This agrees with [36] reporting that many of the IAS used in the manufacturing of PE articles are not included in national and international regulatory lists and [122] who reported that only 45% of 29 FCCs (both IAS and NIAS) detected to migrate from LDPE was included in the Union list.

Sixteen FCCs were metals of which 9 are included in the list of migration limits for metals set by Regulation (EU) 2020/1245. The migration of metals is explained by residues of Ziegler-Natta or metal oxide catalysts used in the production of PE [55]. From the FCCs identified both in this work and in the FCCdb and CPPdb databases, only 11 and 47 FCCs, respectively, were specifically related to PE.

4.1.4. FCCs migration at levels exceeding regulatory limits

Nearly 21% of authorized FCCs included in the Union list (13 out of 53) and the list of metals (3 out of 9) were found to exceed the SML at least once (Fig. 4). Furthermore, nearly one-third of non-authorized FCCs (53 out of 158) exceeded the threshold value of 10 µg/kg at least once.

Sixteen EU authorized FCCs were found to exceed SMLs at least once in 35 migration experiments (Table 2), the majority of which used virgin PE articles as their substrate. In addition, most of these experiments used fatty food simulants (i.e., isooctane, olive oil and 95% ethanol) which are known for their positive correlation with the migration of lipophilic FCCs [122,127,130,45,17,18]. Fatty food simulants such as oil may penetrate PE and act as a plasticizer [18], and 95% ethanol may lead to swelling of the plastic, resulting in increased migration. For example, a study recently illustrated that fat content is positively correlated with

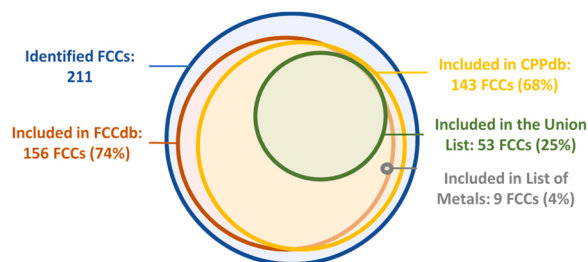


Fig. 3. The number of detected FCCs migrating from single-layer PE articles that are included in existing FCCs databases: i.e., FCCdb [59,69], CPPdb [70], Union List (Commission Regulation (EU) No 10/2011), and List of metals [22].

the migration of DEHA from PE films into three dairy products with increasing fat content, namely; milk (3% fat content), cheese (26% fat content) and butter (80% fat content) [21]. Most of the FCCs that exceeded the SML were either from PE bags/films or from an unspecified PE type, while limited evidence is available on PE density. In Table 2, there are some migration experiments conducted by EFSA in which a few FCCs exceed the SML. EFSA's conclusions were that the migration of these FCCs should not exceed the SML, but no additional explanation was provided.

However, it must be noted that migration testing entails a level of uncertainty arising from e.g., the heterogeneity of the packaging system, variations in the temperature and/or migration time. [104] followed a mathematical approach (i.e., probabilistic analysis using Monte Carlo simulation) to assess the uncertainty and variability in estimates of migration using HDPE bottles as FCM. They found that the affinity of the migrant to the food simulant is the most important source of variability (explaining 70% of the variance) followed by the thickness of FCM (explaining 23% of the variance) under controlled temperature conditions, while variations in the temperature and migration time may be responsible for more than 60% and 20%, respectively, of the variance in the migration estimates [104].

4.2. FCCs migration across the lifecycle of PE articles

FCCs migration from single-layer PE articles has been investigated throughout their entire lifecycle. Focus has been placed on the PE production stage with 83% of the total studies analyzed (96 of 116 studies) having determined the migration of 91% of total FCCs (344 of 377 FCCs) at this stage. Therefore, 80% of the total number of detected FCCs (171 of 211 FCCs) was reported in 80% of studies (80 of 99 studies) focusing on the stage of PE production (Fig. 5).

Research attention on other lifecycle stages (i.e., storage, use/reuse, reprocessing) has been extremely limited, with the reprocessing stage being the least investigated. Only 3% of the total number of studies have investigated recycled PE articles. Interestingly, all of the FCCs investigated at the reprocessing stage (20 FCCs) were found to migrate in PE, in contrast to the other lifecycle stages. This highlights that further research into FCCs at the stage of reprocessing is, therefore, needed to obtain more clarity, as it is likely to uncover a higher number of FCCs migrating from PE-recycled articles.

4.2.1. Production

The factors affecting the migration of FCCs from plastic FCMs at the stage of production are related to the designed characteristics of plastics. For example, the thickness of plastics was found to positively affect FCC migration, including PE [36,42]. Evidence on FCCs migration from PE at the stage of production has focused, specifically, on the use of nanoparticles, design components like paints and inks, application of food preservation technologies and the geographical origin of PE production.

Nanoparticles are used in the production of PE articles, e.g., containers, bags, and films, to enhance antimicrobial, flexibility, barrier, and stability properties [30,31,50,16,85,93]. Due to their potential toxicity, nanoparticles have gained much attention but remain insufficiently studied [117,50]. Some studies detected the migration of total Ag (ion and nanoparticles form) from LDPE articles (e.g., bags [117,50] and PE cutting boards [3]), especially in acidic simulants [117,124,51,101], while other studies did not under same conditions [1,85]. Additionally, a study on the migration of Ag nanoparticles from PE cling films (surface coated with nano-silver) into real food samples found a higher migration rate compared to PE containers, although no chemical changes were observed in the food samples [93].

Some authors report that acidity may affect the migration of Ag nanoparticles in ionic form [124,101] due to their dissolution phenomena in acidic simulants [1]. For example, [1] found that 0.05% of total Ag contained in LDPE articles (i.e., plastic bag and cutting board) migrated into 3% acetic acid, while the migration of Ag into the water

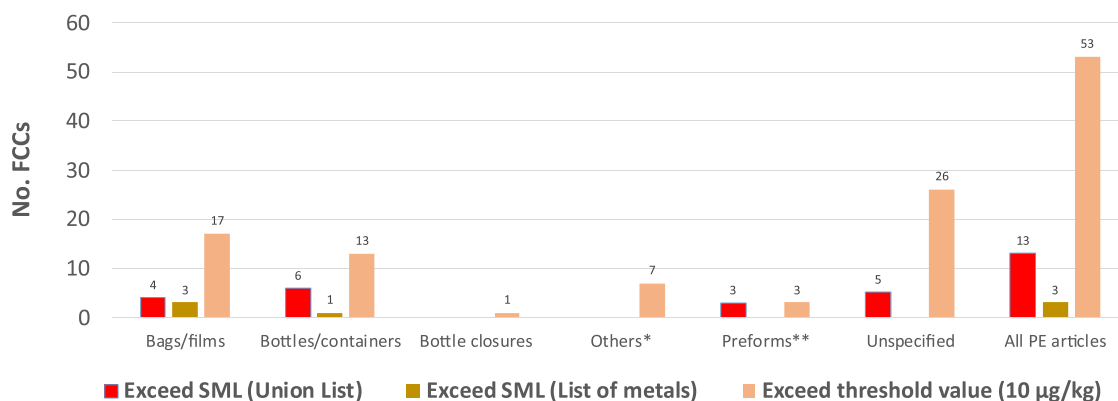


Fig. 4. Number of FCCs migrating from single-layer PE articles that exceeded the regulatory migration limits at least once grouped per article type. * Crate, jug, canister, cups, cutting boards; ** Granulate, beads, strip, sheet, pellet, plaque.

was significantly lower [1]. This was attributed to the oxidation of surface silver nanoparticles into Ag ions leading to the migration of Ag in ionic form rather than as nanoparticles [1]. Similar findings were reported for nano clay, e.g., the migration into 10% ethanol was 4–5 times lower compared to 3% acetic acid, indicating that the pH of the food simulant affects the migration rate of clay nanoparticles as well [51].

Furthermore, two EFSA scientific opinions reported relatively high levels of zinc (Zn) nanoparticles migration from LDPE into 3% acetic acid (7.6–17.3 mg/kg exceeding SML), while in other non-acidic simulants, migration was below the SML [16,9]. Zn is not expected to migrate in nanoform, but the solubilisation of Zn by acidic media leads to the release of ionic Zn; therefore, safety evaluations should focus on the migration of soluble ionic Zn [16,9].

Design components are considered the prevalent sources of migrating FCCs from PE bottles, originating from varnish, paints, or printing inks [110]. The potential migration of polycyclic aromatic compounds (e.g., benzo(a)pyrene) has been stressed when colourants are used in plastic packaging materials [60]. Heavy metals such as lead (Pb) used in pigments and printing inks at the stage of manufacturing can potentially migrate from the printed outer PE packaging into the food since it has been detected to migrate from some samples of PE containers and films [81,82]. Printing ink can be responsible for 98% of all additives leaching from printed LDPE bags into food; noting that leaching of FCCs from LDPE without print was found in an experimental study to be 10-fold lower compared to printed LDPE [115]. These findings are in line with a recent study that reported a strong correlation ($p < 0.001$) between printing ink and phthalates migration from HDPE sachets into water [15].

Food preservation technologies are often applied to extend the shelf life of packaged food with ionizing radiation being considered an effective approach. This technology has attracted the attention of researchers who investigated the migration of FCCs from irradiated PE articles [78]. The experiments showed a downward trend in migration levels with increasing γ -irradiation, and an almost negligible migration at elevated levels of γ -irradiation depending on the FCC, food simulant and irradiation levels [78]. High-pressure processing and modified atmosphere packaging (i.e., using gases such as oxygen, CO₂, and nitrogen) can also be applied to protect and extend the shelf life of food products. The latter has no significant impact on the migration of the antioxidant Irganox 1076 from PE articles [113]. In contrast, [129] reported that high-pressure processing, especially at high temperatures (i.e., 75 °C), may increase the migration of Irganox 1076 from LDPE films.

Geographical origin of PE articles may affect FCCs migration due to differences in the amounts of IAS and quality assurance measures implemented during processing and distribution across the world [64]. According to an FSA report, colourants for food contact plastics produced in China and India appear to contain more substances that could potentially migrate into food than those produced in the EU [62]. This

agrees with [81] who found higher levels of Pb in PE films used as candy packaging originating from China compared to other countries (e.g., South Korea, USA, Mexico, and others), attributing Pb presence to the ink pigments used in the outer printed surface. The migration of FCCs from identical types of PE articles (e.g., bags) may vary from one manufacturer to another making their potential toxicity highly variable [74]. For example, [114] identified the migration of isoborneol from recycled PE articles originating from China, while it was not detected in any of the samples from Spain.

4.2.2. Storage

The quality of packaged food samples may be affected under storage conditions, e.g., [109] reported a plastic-like odour in PE packaging and an off-taste of packed crisps induced by the migration of 8-nonenal – a NIAS. Several studies reported a positive correlation between FCC migration, such as phthalates, with temperature [106,15,56,17,89] and storage time [21,106,15,89]. For example, [15] found that the average cancer risk value due to DEHP for sachet-packed water exceeded the maximum recommended limit (1×10^{-5}) under storage conditions at high temperatures (i.e., 40 °C). However, some studies reported that the positive correlation of FCCs migration with storage time depends on the food simulant [107,126,17]. For example, [17] reported a linear correlation between temperature and phthalates migration into olive oil contained in PE bottles but no linear correlation was found for water as a food simulant. The type of food stored in PE articles is a crucial factor in FCCs migration [104].

4.2.3. Use/reuse

At the stage of use/reuse, surface desorption (i.e., use of acidic simulants) [1] and abrasion may affect the surface morphology of PE articles and can potentially increase nanoparticles migration during the use of PE cutting boards [3,4]. For example, surface abrasion was found to increase the migration of Ag in the form of nanoparticles indicating that the degradation of the polymer matrix by mechanical abrasion induces the dissolution of Ag nanoparticles aggregates into smaller particles; in turn, this results in a higher release of Ag nanoparticles compared to the unabraded cutting boards [3]. Additionally, the migration of DBP and DEHP from PE disposable tableware into the water was found to exceed the limit values suggested by the WHO indicating that the long-term and regular consumption of PE disposable tableware may pose risks to human health [86].

Furthermore, the use of ultraviolet (UV) sterilisation is widely used for kitchenware. The migration of FCCs from PE articles is resistant to UV exposure [80]. In contrast, the dishwashing process was found to increase the migration of additives from PE bottles into drinking water [116]. Microwaving could also mobilise phthalate molecules and in turn, increase phthalate migration from PE wrap films into food [89]. Saltwater was found to induce the leaching of FCCs compared to

Table 2

FCCs migrating from single-layer PE articles at levels exceeding their respective SML.

Chemical name	CAS No	FCM No.	SML (mg/kg)	Migrated FCC concentration (mg/kg)	Food sample/food simulant	PE density	PE article	Virgin or recycled	Ref.
Di(2-ethyl hexyl) phthalate (DEHP)	117–81–7	283	1.5	1.54–2.02 1.57–1.74	Olive oil	PE	Containers	Virgin	[17]
					Olive oil	PE	Plain bags (take-away bags)	Virgin	
				2.4–3.2	10% ethanol	HDPE	Bag	Virgin	[130]
				2.4–5.1	10% ethanol	PE	Food wrap film	Virgin	
				2.2–4.6	Distilled water	PE	Food wrap film	Virgin	
				2.9	Isooctane	PE	Film	Virgin	
				2.2	10% ethanol	PE	Film	Virgin	
2.3	10% sucrose solution	PE	Film	Virgin					
Dibutyl phthalate (DBP)	84–74–2	157	0.3	1.66	Olive oil	LDPE	Cling film	Virgin	[44]
				0.43–1.43	Olive oil	PE	Containers	Virgin	[17]
				0.55	10% ethanol	PE	Film	Virgin	[127]
				0.41	3% acetic acid	PE	Film	Virgin	
				0.61	10% sucrose solution	PE	Film	Virgin	
Di(2-ethylhexyl) adipate (DEHA)	103–23–1	207	18	18.36–24.56	Butter	PE	Films (37 µm thick)	Virgin	[21]
				54.7–109	Olive oil	HDPE	Sheet	Virgin	[97]
Irganox 1076	2082–79–3	433	6	7	95% ethanol	LDPE	Film	Virgin	[129]
				9.4–11.6	olive oil	LDPE	Film	Virgin	[97]
Irganox 1035	41484–35–9	690	2.4	11.8–58	olive oil	HDPE	Sheet	Virgin	[125]
				2.6	Distilled water	LDPE	Drink container	Virgin	
Lead (Pb)	7439–92–1	-	0.01 ^a	0.12	Basic (pH 10.0) reagent water	PE	Film	Virgin	[81]
				0.03–0.04	3% acetic acid	HDPE	Yogurt container (yellow lid)	Virgin	[82]
Chromium (Cr)	7440–47–3	-	0.01 ^a	0.17	Acidic (pH 4.0) reagent water	PE	Film	Virgin	[81]
				0.39	Basic (pH 10.0) reagent water	PE	Film	Virgin	
				7.6–17.3	3% acetic acid	LDPE	Film	Virgin	[16]
Alcohols, C12–14 secondary, beta-(2-hydroxyethoxy), ethoxylated	146340–15–0	802	5	12.3	NA	LDPE	NA	Virgin	[7]
N,N-bis(2-hydroxyethyl) dodecanamide	120–40–1	923	5	10.7	3% acetic acid	LDPE	NA	Virgin	[8]
				11.7	10% ethanol	LDPE	NA	Virgin	
				14.8	50% ethanol	LDPE	NA	Virgin	
2,4-bis(2,4-dimethylphenyl)-6-(2-hydroxy-4-n-octyloxyphenyl)-1,3,5-triazine	2725–22–6	452	5	16.2	olive oil	LDPE	NA	Virgin	[52]
				8.9	olive oil	LLDPE	NA	Virgin	
Phosphate form: phosphorous acid, mixed 2,4-bis(1,1-dimethylpropyl) phenyl and 4-(1,1-dimethylpropyl) phenyl triesters	939402–02–5	974	10	20.9	Olive oil	HDPE and LLDPE	NA	Virgin	[23]
1-Dodecene	112–41–4	268	0.05	0.18	95% ethanol	HDPE	70% recycled bottle and turnover box, 30% virgin PE	Post-consumer recycled	[114]
1-Tetradecene	1120–36–1	388	0.05	0.27	95% ethanol	PE	NA	Post-consumer recycled	[114]
Dodecyl acrylate	2156–97–0	437	0.05	0.10	95% ethanol	HDPE	Bottles and turnover box	Post-consumer recycled	[114]
Octocrylene	6197–30–4	492	0.05	0.17	95% ethanol	PE	Pellet	Post-consumer recycled	[114]

^a Migration limits specified by [22]

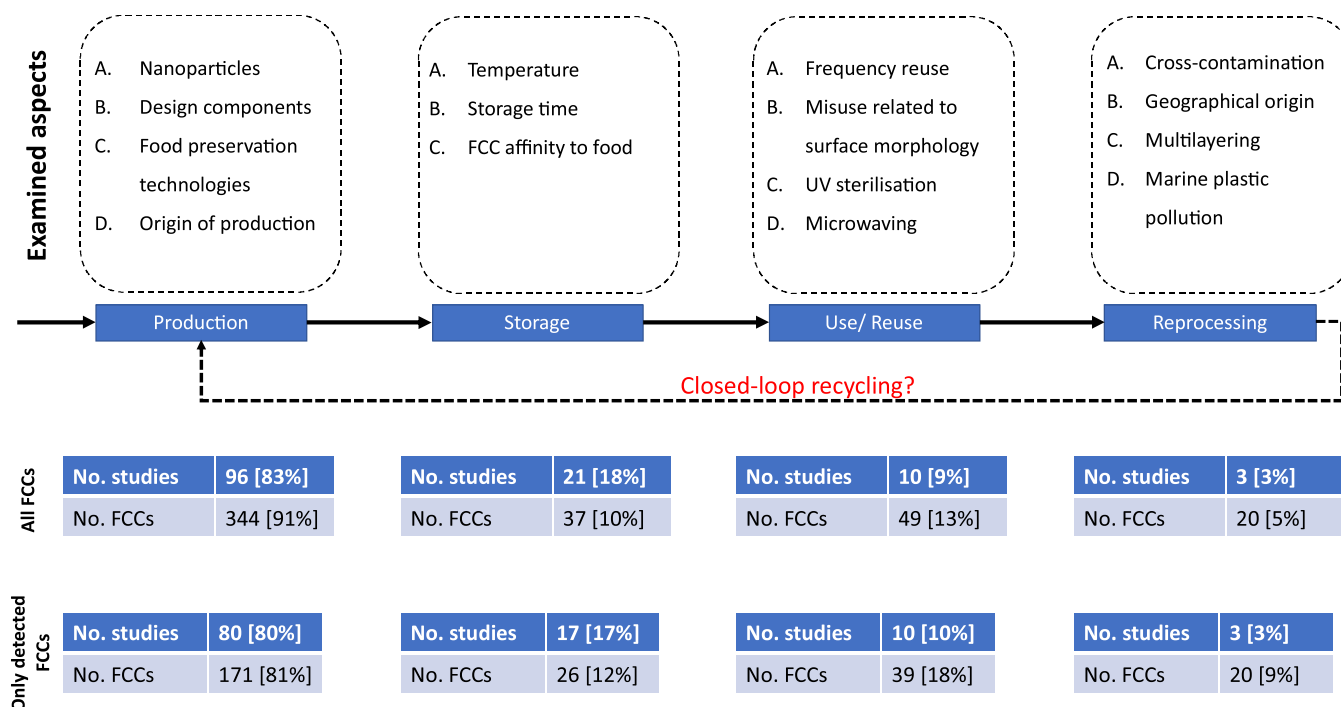


Fig. 5. Overview of FCCs migration-related aspects that were investigated across the lifecycle of single polymer PE food packaging articles based on this systematic evidence map, along with the number of studies and the number of FCCs determined at each stage. Note: percentages in brackets refer to the total number of studies or chemicals that were investigated or detected in total, accordingly.

deionised water. This probability depends on salinity variations, which may lead to complex changes in water and compound chemistry [115].

4.2.4. Reprocessing

Information on the FCC migration from recycled PE into food is very limited [37,39,45]. The migration of four additives widely used in PE manufacturing determined in both virgin and recycled HDPE crates under a different number of recycling cycles showed a slight increase in overall migration with the number of recycling cycles [39]. Authors attributed this increase to the higher concentration of additives with successive cycles of recycling rather than to the reprocessing process itself [39]. However, the migration of several degradation products of additives was detected in recycled HDPE [39] indicating that the reprocessing steps may also be related to FCCs migration. Post-consumer PE is used in the manufacture of multilayer packaging materials as a middle layer in food products and cosmetics [45]. The use of recycled PE in multilayer materials is because recycled PE packaging should not interact physically or chemically with their content [45]. Yet, the number of layers may affect the level of FCC migration. For example, [33] measured the migration of inorganic compounds from recycled HDPE pellets and two multilayer packaging materials - containing both virgin polymers (i.e., PE) and recycled HDPE in the middle layer consisting of three and seven layers, respectively, and found that the levels of migration from the multilayer sample with seven layers were higher due to the manufacturing process.

5. Discussion

Our in-depth analysis reveals the migration potential of 377 FCCs from PE-FCMs, of which 211 FCCs have been found to migrate at detectable levels at least once. Grouping the detected FCCs into IAS and NIAS was not possible, as there is a lack of clear indications on whether an FCC was intentionally or non-intentionally added to the PE food packaging. All the FCCs identified, and those which were included in databases and lists (i.e., FCCdb and CPPdb), could be considered as IAS. However, these databases and lists also contain several NIAS often found

in PE and, hence, they cannot provide detailed polymer-specific information [64].

An example of NIAS in PE food packaging that is usually included in databases is BPA, not expected to be present in PE articles from intentional use. However, BPA has been detected in PE bottles at levels below the SML [35,83,84]. BPA is included in the databases as it is an IAS for other polymers (e.g., PVC or PC). The SML of BPA may not guarantee human safety [72], as evidenced by a recent EFSA opinion that has suggested lowering the tolerable daily intake (TDI) 100,000 times compared to the current TDI [53]. The duality that some FCCs, such as BPA, exhibit (i.e., can be either IAS or NIAS) needs to be further investigated. Phthalates are ubiquitous and FCMs constitute a considerable source of exposure. This can increase the risk of chronic effects on human health and raising awareness of the regular use of FCM including PE might be needed [90]. Assessment of the exposure limits for FCCs remains a challenge not only for NIAS, but also for authorised FCCs for which current regulatory migration limits are often based on outdated or insufficient toxicological data [95,94].

Most FCCs that were found to migrate from PE are non-authorised substances (149 of 211 of the detected FCCs). They are derived from design components [115], such as printing inks containing, e.g., toluene and ethyl benzene, and propyl acetate [61]. Fifty-three non-authorised FCCs exceeded the threshold limit value of 10 µg/kg at least once in migration experiments (see [Supplementary Material \(excel file\)](#)). This threshold limit is a pragmatic value rather than a science-based toxicological threshold [64], that can be used for the migration of non-authorised IAS classified as not being carcinogenic, mutagenic, or toxic for reproduction (CMR) and that are used behind a so-called functional barrier (EU 10/2011). Nevertheless, it is also applied to NIAS for which safety evaluations have not been conducted [64], indicating that there is often limited information on their actual risk. Official guidance on risk assessments of NIAS advances in analytical techniques and exposure assessments, as well as, the development of *in silico* models and databases of toxicity data are needed to assess NIAS [65].

Overall, our study shows that PE, despite being a high-value plastic waste stream that is increasingly recycled, lacks sufficient evidence to

support that it can be safely recycled into new food-grade packaging articles. A considerable number of studies measuring the migration of FCCs from PE food packaging offered deficient information on the use (e.g., bag or container) or physical characteristics (e.g., density or thickness) of PE (24 and 37 of 116, respectively). This hinders our ability to comment and explore the relation of these properties with the level of migration. The absence of key details regarding PE articles assessed in many studies could be related to their focus either on validating a migration method [71,75,82,87] or on investigating other plastic polymers (e.g., PET) and using PE only for comparison [118,40].

Moreover, our analysis revealed that several influential factors, such as design characteristics, frequency of reuse and reprocessing techniques, have been underexplored, while existing information on their influence in FCCs migration from PE is highly fragmented. This lack of information points to our inability to acknowledge and control potential trade-offs arising from FCCs migration across the PE articles' lifecycle. Most importantly, the recycling of PE articles, and the quality of recycled PE (rPE) that is, and will be increasingly, used in the production of new packaging food-grade materials poses critical questions from a chemical safety perspective. The role of several critical aspects defining the success of the reprocessing stage, such as decontamination technologies, waste collection system and design components, and influence on FCCs migration from rPE remain underexplored, as opposed to post-consumer PET bottles [64]. The database of contaminants in the recycled plastic FCMS, and, therefore, in rPE is still very limited indicating the need to improve the identification techniques for these contaminants [119].

It is worth noting that the diffusion coefficient in PE is higher than that of PET, leading to a higher absorption of chemicals in polyolefins than in PET, which, in turn, makes the PE decontamination step even more challenging [114]. The knowledge that particular physical-chemical properties of PE (density, intrinsic viscosity, glass transition, melting point) hamper decontamination at higher temperatures during the reprocessing stage can be useful [102,57]. This has major consequences for the assessment procedures of PE mechanical recycling, which differ from those used for PET, following the established criteria [10,19]. More criteria have been set for the closed-loop mechanical recycling for PET bottles than PE [5,6]. Presently, only a few technologies for the mechanical recycling of polyolefins (PE and PP) have been positively evaluated by EFSA that allow the use of recycled polyolefins in FCMS under defined conditions [2,12].

[34] recently reported that additives used in the design and production of plastics are not optimized for recycling, but only for their processing and first use. At the same time, toxic NIAS can be generated during reprocessing [34], especially for flexible films [58]. The adoption of 'design-for-recycling and potentially, also, traceability principles at the start-of-life stage (e.g., physical characteristics, RFID labelling, avoidance of hazardous chemicals and controlled selection of labels, printing inks, varnish, adhesives, and best-before-date-print)' is an important measure for increasing the circularity of plastic food packaging [64]. The action plan of the EU Chemicals Strategy for Sustainability (CSS) as part of the EU's zero pollution ambition aims at developing a framework for the definition of safe and sustainable by design (SSbD) criteria for chemicals and FCMS [33]. SSbD is an approach that integrates critical aspects for sustainability such as safety, circularity and functionality of chemicals and materials across their entire lifecycle from the stage of design to the final EoL management stage [33]. Recently, an OECD report provided key considerations and criteria for the selection of inherently sustainable plastics from a chemical perspective including materials selection processes across all stages of the plastics lifecycle [98]. As a follow-up, OECD held a workshop and developed a workshop report and two background reports aiming to understand challenges from a chemical perspective that are related to the design of more sustainable flexible food-grade packaging [99]. According to these reports, there is not one single solution that can improve the sustainability of plastics, while policy support across the plastics lifecycle is needed with chemical

transparency and safety being key priority areas [99].

Borealis – a European company specialized on polyolefin recycling – has recently provided 10 design for recyclability (DfR) codes that can be adopted at the stage of product design and are aimed at improving the recyclability of polyolefins at their end-of-life stage [29]. These DfR codes can make PE packaging "recycling-ready" and are based on the following three principles [29]:

- i. design food packaging articles consisting of 'as few different polymer types, components and materials as possible' highlighting that multicomponent or multilayer food packaging materials should be avoided – the safety challenges, as well as the challenges of recycling multi-layer plastic food packaging are reported elsewhere [20];
- ii. design food packaging articles in a way that is easy to wash off or strip all design components (e.g., labels, sleeves, adhesives, printing, and inks) from the main body of the article;
- iii. by no means of affecting the food or product preservation/protection, design food packaging articles under requirements and not over-engineering.

Whilst we commend the efforts to change the design of PE plastic packages, it is imperative to point out that such attempts should be matched with the right evidence; because, such transitions require informed and careful planning in order to deliver the desired results and transform the PE plastic packaging value chain. For instance, in relation to i. - there needs to be a clear emphasis and concurrent action on regulating the additives used in the design of the PE food packages; in relation to ii. - design principles should effectively discourage the use of printing inks, adhesives and labels that can play an important role in FCCs migration; and finally, in relation to iii. - understanding that in multinational markets products need to be over-engineered to cater for diverse needs is imperative, and therefore, information on what this entails should become available to design changes in a way that can be efficiently and effectively managed across different contexts. The food and beverage sector strongly relies on PE to provide goods to billions of people every day, making it unlikely that PE will be replaced in the food packaging sector soon. Therefore, it is essential to understand the consequences of the production and use of PE food plastic packaging from a sustainability and chemical safety perspective, especially as many nations rely on the flexible single-layer PE articles that are vastly placed on the market and are grossly mismanaged, posing safety risks to both the environment and human health. Fundamentally, plastic packaging production should be reduced, and adherence to the SSbD principles should be prioritized during its design and production stage to prevent unintended consequences to the environment and human health. By designing system changes holistically and via a lifecycle, systems thinking approach [77], SSbD goals can be sufficiently supported, e.g., through selecting materials with low hazard and risk profiles, and planning the product's commercial 'afterlife' to ensure minimum waste generation and targeting for secondary feedstock [98].

The Department for Environment, Food and Rural Affairs (Defra) in the UK has launched a series of stakeholder workshops to inform on the UK chemicals strategy aiming to ensure the safe use and management of chemicals [108]. This is in line with the EU CSS action plan towards the safer use and management of chemicals and materials [33]. Inspired by these actions, key enabling conditions are recommended:

- i) Improving the communication between waste operators and PE product designers aiming to support and implement SSbD criteria;
- ii) Increasing the transparency and traceability of PE food packaging quality across the supply chain through initiatives such as product passports and labelling systems;

- iii) Developing technologies and databases able to support transparency of processes and decision-making across the entire lifecycle of PE food packaging with a focus on reprocessing;
- iv) Developing a monitoring mechanism to control the quality of PE food packaging in compliance with SSbD and to identify gaps in the regulatory framework aiming to build a robust and up-to-date policy of FCCs;
- v) Focusing on FCCs, e.g., EDCs, that prevent closed-loop recycling for PE food packaging accompanied with plastic waste tracking. In that case, the development of methodologies to assess the toxicity of chemical mixtures with similar behaviour rather than substances individually can be insightful; and
- vi) Incentivising better product design through economic instruments such as taxation, extended producer responsibility, and waste management technologies that reflect SSbD criteria.

Different stakeholders are involved in the value chain of PE food packaging and therefore building effective partnerships and trust, especially between PE manufacturers and regulators would mobilise the transition towards a closed-loop PE food packaging system with the minimum trade-offs from a chemical perspective.

6. Conclusions

Our study revealed that at least 211 FCCs have the potential to migrate from single-layer PE food packaging materials. More than a quarter of these chemicals are not included in inventory databases and lists and only 25% are included in the Union List, which highlights the lack of data traceability and sufficient harmonisation of existing regulations. Above all, the currently insufficient state-of-the-evidence available for PE-FCMs hinders our ability to identify the right points of intervention in the PE value chain. This emphasises the criticality of further research on: FCCs migration from specific, well-characterized PE articles, especially, those collected for recycling; and the effect of various designed (e.g., ink, density, hardness) and created (i.e., physically induced during use) characteristics [76], on FCCs migration along the PE value chain. While limited evidence prevents us from reliably identifying the strongest points of intervention across the entire lifecycle of PE food packaging, it also highlights the crucial need for the collection and assembly of all the information needed to better understand and monitor PE food packaging quality from a chemical perspective across its entire lifecycle and allow careful and well-informed changes in the PE value chain. A reduction in the volumes of PE-FCMs produced, as well as the adoption of SSbD practices at the design and production stage, would be a good starting point to enable the transition to a sustainable and circular plastics value chain.

Environmental implications

This study looks into the migration of food contact chemicals (FCCs) from polyethylene (PE) food packaging articles to food and food simulants. Many of the FCCs identified are phthalates, which are considered to be endocrine disruptors and thus, of hazardous nature to the environment and human health. Shedding light on the migration potential of such chemicals points to the need for understanding and monitoring the quality of PE food packaging in compliance with sustainable and safe by design criteria. This will illuminate gaps in the regulatory framework and help to build a robust and up-to-date policy for FCCs in plastic packaging.

Declaration of Competing Interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Olwenn V. Martin is a member of the management board of the European Chemical Agency, and sits on the Scientific Advisory Board (SAB)

of the Food Packaging Forum. Ksenia Groh sits on the Scientific Advisory Board (SAB) of the Food Packaging Forum.

Data Availability

Data is provided in the supplementary material at the Attach File step.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at [doi:10.1016/j.jhazmat.2023.131422](https://doi.org/10.1016/j.jhazmat.2023.131422).

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