



Review

Unpacking the complexity of the PET drink bottles value chain: A chemicals perspective

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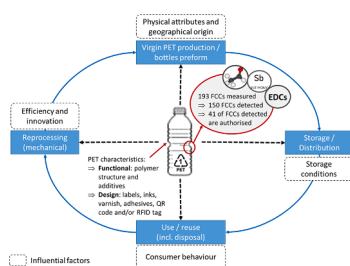
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HIGHLIGHTS

- Food contact chemicals (FCCs) migrating from PET bottles are examined.
- 193 FCCs have been investigated, of which 150 have been detected at least once.
- Research has focused on the migration of Sb, acetaldehyde and some well-known EDCs.
- Safety implications arising from reprocessing of PET bottles remain underexplored.
- Safety of rPET depends on transparency and cooperation across the value chain.

GRAPHICAL ABSTRACT



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ABSTRACT

Chemicals can migrate from polyethylene terephthalate (PET) drink bottles to their content and recycling processes may concentrate or introduce new chemicals to the PET value chain. Therefore, even though recycling PET bottles is key in reducing plastic pollution, it may raise concerns about safety and quality. This study provides a systematic evidence map of the food contact chemicals (FCCs) that migrate from PET drink bottles aiming to identify challenges in closing the plastic packaging loop. The migration potential of 193 FCCs has been investigated across the PET drink bottles lifecycle, of which 150 have been detected to migrate from PET bottles into food simulants/food samples. The study reveals that much research has focused on the migration of antimony (Sb), acetaldehyde and some well-known endocrine-disrupting chemicals (EDCs). It indicates and discusses the key influential factors on FCCs migration, such as physical characteristics and geographical origin of PET bottles, storage conditions, and reprocessing efficiency. Although, safety and quality implications arising from the

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recycling of PET bottles remain underexplored, the higher migration of Sb and Bishphenol A has been reported in recycled (rPET) compared to virgin PET. This is attributed to multiple contamination sources and the variability in the collection, sorting, and decontamination efficiency. Better collaboration among stakeholders across the entire PET bottles lifecycle is needed to ensure sustainable resource management and food contact safety of rPET.

Nomenclature		EDCs	Endocrine-disrupting chemicals
AGR	Annual growth rate	EG	Ethylene glycol
BBP	Benzylbutyl phthalate	EFSA	European Food Safety Authority
BPA	Bisphenol A	FCCdb	Food Contact Chemicals database
Br	Bromine	FCC	Food contact chemical
CAS	Chemical Abstracts Service	FCM	Food contact material
Cd	Cadmium	FDA	Food and Drug Administration
CLARITY-BPA	Consortium Linking Academic and Regulatory Insights on BPA Toxicity	IAS	Intentionally added substances
CMR	Carcinogenic, mutagenic, or toxic for reproduction	LoD	Limit of detection
CO ₂	Carbon dioxide	MSW	Municipal solid waste
COSTER	Conduct of systematic reviews in toxicology and environmental health research	Mt	Million metric tons
CPPdb	Chemicals associated with Plastic Packaging database	Ni	Nickel
Cr	Chromium	NIAS	Non-intentionally added substances
C&I	Commercial and industrial waste	OML	Overall migration limit
DBP	Dibutyl phthalate	Pb	Lead
DEHP	Di(2-ethylhexyl) phthalate	PET	Polyethylene terephthalate
DEP	Diethyl phthalate	PVC	Polyvinyl chloride
DMP	Dimethyl phthalate	rPET	Recycled PET
DMT	Dimethyl terephthalate	Sb	Antimony
DnOP	Di-n-octyl phthalate	Sb ₂ O ₃	Antimony trioxide
DRS	Deposit return schemes	SML	Specific migration limit
		TA	Terephthalic acid
		TDI	Tolerable daily intake
		TPA	Terephthalic acid

1. Introduction

Plastics, owing to their lightweight, flexible and durable nature, find use in multiple applications, including food packaging, and are considered to be the workhorse material of our modern society (Hahladakis, 2020). Among the most widely used types of plastics in the food packaging sector, polyethylene terephthalate (PET) is a thermoplastic produced by the polymerization of ethylene glycol (EG) and terephthalic acid (TA) or dimethyl terephthalate (DMT). PET is a particularly attractive polymer, due to its high transparency, high reflectiveness, easy processability, and relatively low prices, ranking it as the third largest polymer used in packaging applications. One of its most popular end-uses is in drink bottle (also known as beverage bottles) manufacturing (e.g. carbonated drinks, still drinks, fruit juices, beer and bottled water) (PlasticsEurope, 2018).

The widespread use of PET in drink bottles has received increased attention recently due to the short shelf life (around 6 months) of this packaging type (Ros-Chumillas et al., 2007), and, most importantly, its single-use nature (Hardesty et al., 2021). These properties along with socioeconomic and cultural factors (e.g. on-the-go consumption and poor or absent waste management infrastructure at the regional level) have rendered PET bottles one of the most commonly found plastic litter (Hardesty et al., 2021). Plastic pollution is now recognised as a planetary boundary challenge (Arp et al., 2021) and many countries around the world are committed to improving their local plastic recycling rates (Geyer et al., 2017). It is estimated that almost 58% of PET bottles were collected and sorted for recycling across the European Union (EU) in 2019 (PetcoreEurope, 2019). In 2020, the recycled PET (rPET) for bottle-to-bottle applications represented 32% of the rPET market in Europe, with an average recycled content of 14% (CPME, 2021). Yet,

according to the Directive on single-use plastics, PET bottles shall contain at least 25% of recycled content by 2025 (Directive, 2019/904, 2019), therefore this figure is set to increase. The European Directive on Packaging and Packaging Waste further mandates that by 2030, 55% of all plastic packaging should be recycled (Directive, 2018/852, 2018). PET bottles in particular shall contain at least 30% recycled content. This latter goal appears to be on track and is supported by the introduction of the EU Packaging Levy, a tax on non-recycled plastic packaging introduced in 2021 (European Commission, 2021a). Notwithstanding the controversies around the implementation of the levy, in the long term, it may encourage recycling and the development of a secondary commodities market to tackle the use of recycled plastic materials.

However, recycling of food contact materials (FCMs) may introduce new, or concentrate existing potentially hazardous chemicals, which may migrate into food (Geueke et al., 2018). Therefore, the increased use of rPET in FCMs presents a safety challenge that requires attention. Food contact chemicals (FCCs) are now recognised as a relevant route of chronic human exposure to hazardous substances (Geueke and Muncke, 2018; Muncke et al., 2020). In a global inventory of substances authorised for use in the manufacture of FCMs, 4742 have been assigned to plastic FCMs (Groh et al., 2021). In addition to these intentionally added substances (IAS) (such as *additives*, *monomers* and *catalysts*), non-intentionally added substances (NIAS) may also arise from impurities and reaction(s) by-products, or degradation of additives, during any stage of PET's life cycle (Groh et al., 2019; Wiesinger et al., 2021). Both IAS and NIAS may migrate into food and drinks.

To realise the required circularity and the multi-dimensional (i.e. environmental, economic, social, and technical) benefits envisaged by recycling plastic packaging waste, single-use plastic and packaging waste legislation must be closely aligned with regulations on FCMs.

These include:

- i) the European Regulation that provides general requirements on FCMs and food contact articles at the stage of manufacturing and processing (European Regulation (EC) No 1935/2004, 2004);
- ii) the European Plastics Regulations on plastic FCMs and food contact articles that define a list of authorised substances, known as the *Union list*, that can be added to plastic packaging materials under stipulated usage conditions and Specific Migration Limits (SML) (Commission Regulation (EU) No 10/2011, 2011; Commission Regulation (EU) 2020/1245, 2020); and
- iii) the European Regulation on recycled plastic FCMs that authorises recycling processes and introduces quality assurance requirements (Commission Regulation (EU) No 282/2008, 2008).

Current regulatory migration limits are based on traditional toxicological methods (Warner and Flaws, 2018), but the underlying data is often outdated or incomplete (Muncke et al., 2020; Muncke et al., 2017). For authorised substances included in the Union List, the overall migration limit (OML) from plastic FCMs to food simulants should not exceed 60 mg/kg food or 10 mg/dm² (expressed on a contact area basis). Due to gaps in knowledge about NIAS chemical identities and the lack of analytical standards (both IAS and NIAS), assessment and enforcement of migration and exposure limits for FCCs remain challenging, and this stands in the way of comprehensive safety determination (Muncke et al., 2017; Geueke, 2018; Thoden van Velzen et al., 2020). Regulation (EU) No 10/2011, (2011), acknowledges the likelihood of NIAS to be present in plastic FCMs and requires FCMs manufacturers to ensure safety for all migrating chemicals, including NIAS. In practice, an arbitrarily derived threshold limit of 10 µg/kg food is used for NIAS classified as not carcinogenic, mutagenic, or toxic for reproduction (CMR), while the toxicological threshold limit for potentially genotoxic NIAS was set at 0.017 µg/kg food for a worst-case scenario by the European Food Safety Authority (EFSA) (EFSA, 2011). EFSA also relies on this migration level to evaluate the safety of recycling processes for any unknown contaminant possibly present in rPET (EFSA, 2011), while the US Food and Drug Administration (FDA) has based the criteria for safety evaluation of recycled plastics on its Threshold of Regulation, set at 0.5 µg/kg food (US FDA, 2006).

These limits do neither account for mixture effects nor exposure via other routes which are non-negligible in the case scenario of ubiquitous materials such as plastics. Because of these known inconsistencies and insufficiencies, EU regulation on FCMs, including the recycled plastics Regulation (Commission Regulation EU 282/2008, 2008), is currently under revision (European Commission, 2021b). This revision also aims to address recyclers' need for increasing the recycling rates, while ensuring good quality output from a chemical perspective.

Given the scale of recycling ambitions in the food and drink packaging sector, there is an urgent need to interrogate current evidence about FCCs migrating from PET and rPET bottles to better understand which stages of a PET drink bottle's lifecycle (i.e., production, use, disposal and/or reprocessing and (re)integration back to the value chain as secondary material) may be prone to contamination. Therefore, this study aims to unpack the evidence and available information on the most frequently investigated and detected FCCs in PET and rPET, and analyse the conditions that may affect and/or control the presence of IAS and NIAS across the whole PET lifecycle. This offers insights into the impact of recycling processes on the potential chemical contamination related to the use of rPET. To this aim, the study exploits a subset of results from a systematic evidence map of the migration of FCCs into food. Here, we present the results, focusing on FCCs found in PET and/or rPET that have exceeded the current SML under specific test conditions at least once, and those that have been most frequently analysed. Then, we collate and critically assess the influential factors, practices, and conditions across all stages of the lifecycle, discuss the study's main findings, and make recommendations for further research.

2. Methodology

2.1. Literature search

Existing evidence about the migration of FCCs from PET and rPET was gathered as part of a larger evidence mapping effort carried out according to a published protocol Martin et al. (2018) adhering to the COSTER (conduct of systematic reviews in toxicology and environmental health research) recommendations (Whaley et al., 2020). Briefly, searches for peer-reviewed articles were conducted in five bibliographic databases (PubMed, Web of Science, Scopus, Google Scholar and ScienceDirect) using search algorithms combining 108 search terms. This strategy was completed by searches in grey literature databases (e.g. Environar, OpenGrey, Core) and targeted manual searches of relevant institutional websites (e.g. EFSA, JRC, USEPA, US FDA). This search yielded 15,915 studies that were screened for their relevance based on predefined selection criteria. The labour-intensive task of extracting information from the resulting 1211 eligible studies was assisted by a software tool (SciExtract) that allowed expert users to edit and extract relevant key terms from a domain-specific library automatically identified, categorized and highlighted using the third-party PDF annotation tool *tagtog*. This process generated a total of 22,601 database entries corresponding to unique investigations with 3240 unique chemicals in FCAs. The results of the full systematic evidence map will be published elsewhere (Geueke et al., *in preparation*). The records that specifically described the migration or extraction of FCCs from PET or rPET articles, such as films, bags, containers, bottles, trays, and other PET products and items, were obtained from this database. Studies that either conducted extraction experiments or analysed PET articles that were not associated with drink bottles were excluded. Therefore, this systematic evidence map included only studies that focused on the migration of FCCs from PET drink bottles, as well as precursors of PET drink bottles, i.e., virgin and recycled PET flakes and preforms, and PET containers (usually containing water).

The key terms used in our systematic evidence mapping and analysis are defined below. It must be noted that these terms are not necessarily used identically throughout the literature.

- **Food contact chemicals (FCCs):** several types of chemicals of both IAS and NIAS nature that are present in FCMs and can migrate into food (i.e., bottled drink) or food simulant (Martin et al., 2018).
- **Non-intentionally added substances (NIAS):** FCCs that are present in FCMs arising from impurities (i.e. residual contaminant), reaction products during FCMs manufacturing (i.e. oligomers) (Grob et al., 2009), and/or polymer and additive degradation (Nerin et al., 2013).
- **Intentionally added substances (IAS):** FCCs that are intentionally used in the manufacturing of FCMs, such as main constituents of the polymer chain (i.e. monomers), catalysts, or additives.
- **Additives:** a sub-category of IAS that improve the performance (e.g., during the shaping of the polymer through injection moulding, extrusion, blow moulding, vacuum moulding, etc.), functionality and ageing properties of the polymer. They can be grouped into *functional additives* (e.g., stabilisers, antistatic agents, flame retardants, plasticizers, lubricants, slip agents, curing agents, foaming agents, biocides); *colourants* (e.g., pigments, soluble azocolorants); *fillers* (e.g., talc, kaolin, clay, mica, calcium carbonate, barium sulphate); and *reinforcements* (e.g., carbon and glass fibres) (Hahladakis et al., 2018a).
- **Migration:** is the transfer of an FCC from an FCM into food or food simulant by diffusion, ion exchange/surface interaction, abrasion, or other processes under realistic and intended-use conditions (Martin et al., 2018).
- **Extraction:** is the transfer of an FCC from an FCM into a solvent under non-foreseeable use conditions arising from conditions that promote a strong interaction between the solvent and the selected

FCM – usually described as a worst-case migration (Martin et al., 2018).

- **Food simulant:** a liquid or solid substance with a well-defined composition (usually a solution or an oil) that is used in migration experiments aiming to simulate the chemical properties of specific foodstuffs (Martin et al., 2018) (information of the most prevalent food simulants reported in the Union list are given in Supporting Information – A).
- **Lifecycle:** stages of the PET drink bottles flow in the production-consumption-management system, including storage, distribution, and re-distribution back to the value chain (denotes circularity).

2.2. Data processing and analysis

Detailed information was extracted from the filtered records to compile a database of FCCs, including both IAS and NIAS, migrating from PET or rPET during each stage of the lifecycle, to document factors that may influence the migration of FCCs, such as physical properties of PET bottle (e.g. capacity, thickness and colour), storage conditions, food chemistry (i.e. type of food simulant and/or food sample / bottled drink), recycling process (reprocessing stage), and purchase location. Specifically, the resulting database (see Supporting Information – B) includes the following information:

1. Name of FCC along with its Chemical Abstracts Service (CAS) number, if available;
2. Type of FCC in terms of its role in PET (re-)processing (i.e., additive, catalyst, monomer, oligomer, impurity – or else referred to as residual contaminant, and degradation products);
3. Whether FCC was included in two relevant databases as well as recording its relation to PET based on these databases: Food Contact Chemicals database (FCCdb) which is a list of IAS potentially used worldwide in FCM manufacturing (FPF, 2021; Groh et al., 2021), and Chemicals associated with Plastic Packaging database (CPPdb), a list of chemicals associated with plastic packaging (Groh et al., 2019);
4. SML as set by Commission Regulation (EU) No 10/2011, (2011) along with the reference number of the substance as given in the Union list (FCM No);
5. Initial concentration of FCC (if available) in PET bottle before migrating;
6. Measured migrated concentration; limit of detection (LoD);
7. Number of samples analysed for the given migrated concentration;
8. Migration conditions (e.g., duration, temperature, UV exposure, pH, or any other procedure applied before migration test);
9. Food simulant or food sample;
10. Chemical analysis method;
11. Type of PET (i.e., virgin vs recycled, including mixed virgin-recycled content);
12. Use of PET article (i.e., single-use vs repeat-use);
13. Form of PET article (e.g., bottle, preform, pellet, bottle, container) including physical characteristics if available (e.g. capacity and colour); and
14. Purchase location of PET article.

The collated data were used to generate summary tables and graphs. In-depth analyses focus on the most frequently investigated FCCs that exceeded the SML in migration experiments at least once. Due to heterogeneity in reporting, some studies reported the migrated concentration of individual samples while others reported mean values across several samples with identical characteristics (i.e., type, use, form, and location). The arithmetic means measured by each study under identical migration conditions, analytical method, and food simulant were calculated.

3. Results

3.1. Overview of findings from the literature

To date, over 1000 migration tests on PET bottles and other relevant samples, such as flakes and pellets, have been reported by a total of 91 studies. As shown in Fig. 1, experiments have been conducted most frequently on bottles purchased in Spain, Germany and China, and rPET has been analysed on a global scale.

Collectively, the studies measured the migration rate of 193 FCCs from PET drink bottles into several food simulants and food samples (e.g., bottled water, soft drinks, fruit juices, milk, etc.) using a wide variety of test conditions across all lifecycle stages (see Supporting Information – B). Of these, 150 FCCs have been detected (> LoD) to migrate from PET bottles into food simulants/food samples. Summary findings according to FCC type and role in PET bottles (re)processing are shown in Table 1. It must be noted that the categorisation of FCCs according to type is indicative; their role in PET bottles (re)processing may be multivarious (e.g. residual contaminant and degradation product).

Fig. 2 illustrates the proportion of each type of FCCs in the total number of all FCCs that have been detected. Specifically, residual contaminants (41% of total FCCs) followed by degradation products (25%) constitute the most prevalent types of FCCs migrating from PET bottles. This finding indicates that the majority of FCCs migrate from PET bottles are NIAS.

The majority of FCCs measured by researchers (193 FCCs in total) has been included in FCCdb (131 FCCs) (FPF, 2021) and CPPdb (133 FCCs) (Groh et al., 2019). Only a few of these FCCs has been reported to be related specifically to PET – 13 and 33 FCCs based on FCCdb and CPPdb, respectively (Table 2).

Additionally, only 56 out of 193 FCCs are present in the Union list. Of the 150 detected in migration experiments of PET bottles, 41 are present in the Union list. These findings show that a high number of non-authorised substances may be intentionally or non-intentionally added across the lifecycle of PET bottles.

In the majority of tests, the migration of individual FCCs from PET drink bottles was found to be within current regulatory limits. There were cases where the migration of FCCs exceeded the SML (for authorised substances) or the regulatory threshold value (for non-authorised substances) (Table 3). As expected, the type of food simulant can significantly affect the migration of FCCs. For example, the highest migration of PET oligomers was observed in a fatty food simulant (i.e., 95% ethanol) (Ubeda et al., 2018), while the leaching rate of heavy metals (i.e., lead (Pb), cadmium (Cd), chromium (Cr), nickel (Ni) and antimony (Sb)) was detected in 5% citric acid at a rate higher than in deionized water (Whitt et al., 2016). Food simulants with lipophilic character, such as 50% and 95% ethanol, might cause swelling of the PET polymer matrix, which in turn could accelerate the migration of organic FCCs (Gehring and Welle, 2018; Franz and Welle, 2008; Franz et al., 2016). This is supported by our findings which suggest that most of the FCCs that exceeded regulatory migration limits have been found in fatty food simulants (i.e., 50% and 95% ethanol), or food types with lipophilic character (i.e., milk) (Table 3).

3.2. Focus on most studied FCCs: antimony (Sb), carbonyl compounds and endocrine-disrupting chemicals (EDCs)

3.2.1. Antimony

Antimony (Sb) trioxide (Sb₂O₃) is used as a catalyst in PET manufacturing (Kiyataka et al., 2018). With an SML of 40 µg/kg, the migration of Sb from PET bottles concerns PET stakeholders throughout its whole lifecycle. It can cause acute and chronic health issues, including cancer (Westerhoff et al., 2008; Buser et al., 2019), and its (Sb's) tolerable daily intake (TDI) has been set at 6 µg/kg body weight (WHO, 2017). Although the consumption of bottled water was not found to pose health risks related to Sb migration (Zmit and

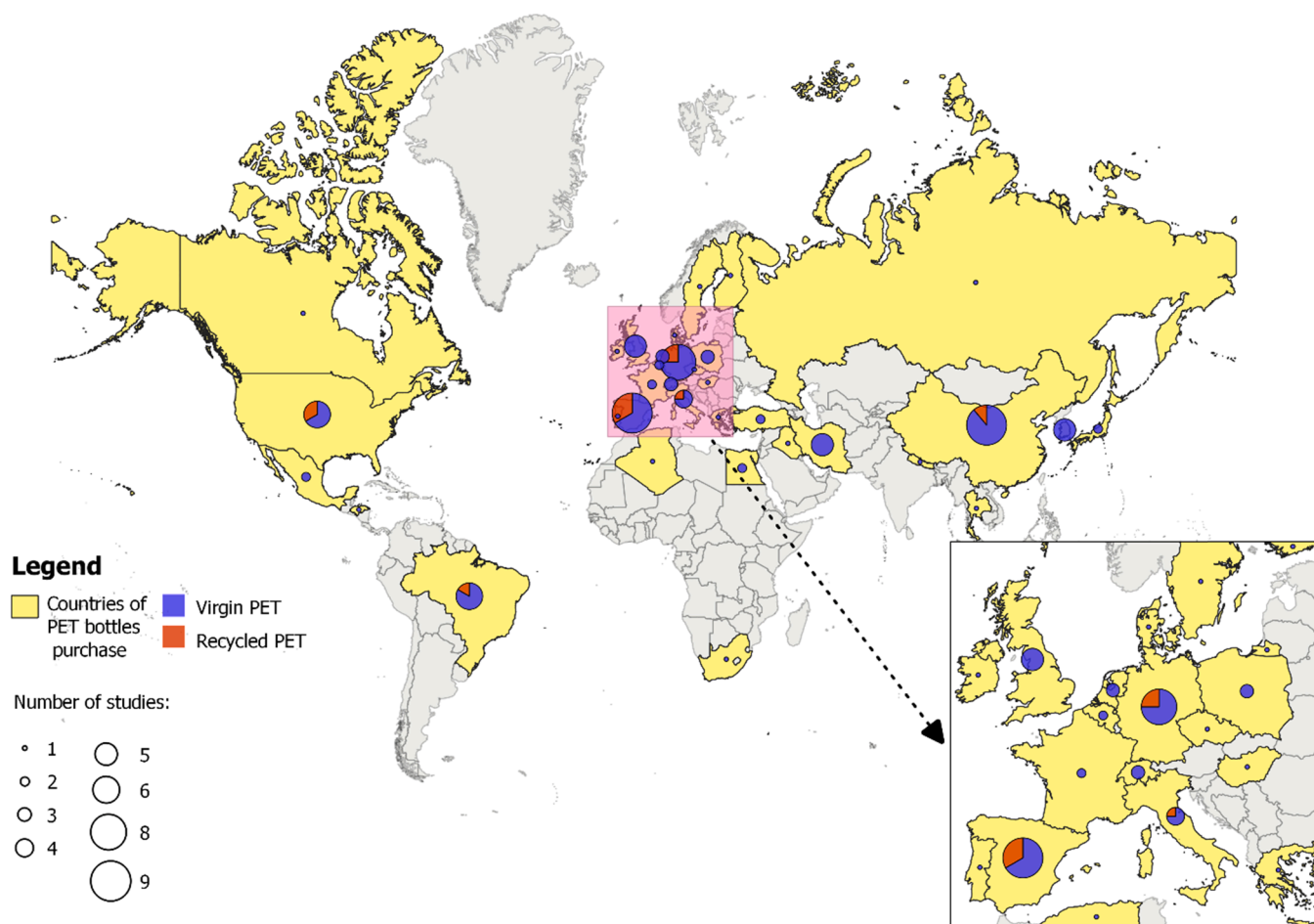


Fig. 1. Map of countries from which PET bottles were purchased to conduct migration experiments per study, including both recycled and virgin PET bottles.

Table 1

The total number of analysed and detected FCCs in migration experiments of PET bottles grouped according to their role in PET bottles (re)processing based on this systematic evidence map.

Type of FCCs	Description	IAS vs NIAS	Number of analysed FCCs	Number of detected FCCs	Number of studies
Catalysts & Co-catalysts	Substances added to regulate the incorporation of monomers into a polymer chain ^a .	IAS	6	5	25
Additives	Aids to achieve defined technological effects ^b .	IAS	34	24	19
Monomers & Co-monomers	Most important components and copolymer precursors to build up the polymer chains ^c .	IAS	12	8	9
Oligomers	Intermediates of polymerisation; low molecular weight polymers comprising a small number of repeat units ^d .	NIAS	15	14	7
Degradation products	Break-down products from polymer and additives degradation processes due to high temperatures and/or high irradiation energies ^e .	NIAS	46	38	32
Residual contaminants (impurities)	Undesirable substances accompanied by used substances (i.e. raw materials or additives) or equipment ^f .	NIAS	80	61	41

^a (Kaiser and Long, 2018).

^b (Commission Regulation (EU) No 10/2011, 2011).

^c (Yuan et al., 2012).

^d (Klaerner and Padmanabhan, 2016).

^e (Nerin et al., 2013).

^f (Grob et al., 2009).

Belhaneche-Bensemra, 2019), an association between drinking water antimony levels and cancer incidences has been reported (Colak et al., 2015). The mismanagement of PET drink bottles at their end-of-life stage (e.g., littering, dumping) can induce environmental pollution arising from Sb leaching from the bottle to the environment (Kiyataka et al., 2018).

Based on the collected data, the total concentration of Sb in PET

drink bottles ranged from 70 mg/kg (Chapa-Martínez et al., 2016) to 650 mg/kg (Kiyataka et al., 2018), depending on the use of other catalysts (e.g., titanium-based or germanium-based), and the reaction environment during processing (i.e., temperature and pressure conditions) (Westerhoff et al., 2008).

Fig. 3 shows Sb migration into three main food simulants: solutions with low per cent ethanol (migration tests used 10 and 15 v/v ethanol)

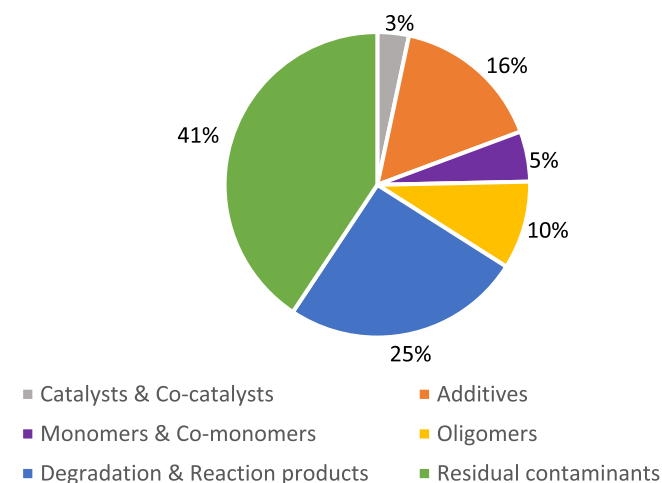


Fig. 2. Contribution of each FCCs type to the total number of FCCs that have been detected to migrate from PET drink bottles into food simulant/food sample according to the present systematic evidence map.

Table 2

Total number of FCCs measured and detected in migration experiments of PET bottles and the number (of them) included in FCCdb and CPPdb and with specified relation to PET.

	Evidence map	FCCdb ^a		CPPdb ^b	
		Included	Related to PET	Included	Related to PET
Number of analysed FCCs in PET bottles	193	131	13	133	33 (55% considered as NIAS in PET)
Number of detected FCCs in PET bottles	150	102	8	106	29 (59% considered as NIAS in PET)

^a (FPF, 2021).

^b (Groh et al., 2019).

(3-A); solutions with low per cent acetic acid (3–5% v/v) (3-B); and non-carbonated water (3-C) (Andra et al., 2011; Bach et al., 2014; Carneado et al., 2015; Chapa-Martínez et al., 2016; Cheng et al., 2010; Dogan and Cebi, 2019; Dutra et al., 2014; Fan et al., 2014; Fordham et al., 1995; Keresztes et al., 2009; Kiyataka et al., 2018; Reimann et al., 2012; Shoty et al., 2006; Westerhoff et al., 2008; Zmit and Belhaneche-Bensemra, 2019; Thompson et al., 1997; Lei et al., 2016). Sb migration values detected were all below the SML (Fig. 3). Migration of Sb into all three simulants was higher at the stage of reprocessing than at the stage of production, suggesting higher concentrations of Sb in rPET than virgin PET bottles. Dutra et al. (2014) suggested that Sb migration from rPET depends on the efficiency of the cleaning process. They supported the occurrence of higher levels of Sb migrating from rPET bottles when subjected to conventional cleaning versus more advanced cleaning processes. The importance of washing procedures was also highlighted by Cheng et al. (2010) who recommended that bottle fillers need to wash rPET bottles before first use.

Still (non-carbonated) bottled water or distilled water was used as a food sample or food simulant, respectively, in most studies (Fig. 3-C). The migration of Sb in food samples other than water (e.g., milk, coffee, acidic and carbonated drinks) can be higher than in still water under identical migration conditions (Fan et al., 2014). For example, Bach et al. (2014) and Keresztes et al. (2009) reported higher levels of Sb migration in carbonated water samples compared to non-carbonated bottled water.

Many researchers underlined the correlation between storage time

(Dogan and Cebi, 2019; Shoty et al., 2006; Payán et al., 2017) and temperature (Chapa-Martínez et al., 2016; Payán et al., 2017; Rungchang et al., 2013), and the higher Sb migration into non-carbonated water. Keresztes et al. (2009) stated that Sb is more likely to migrate from PET bottles in the first year of storage. This was confirmed by Zmit and Belhaneche-Bensemra (2019), who demonstrated that one year of storage can increase Sb migration by 88%. These authors also found that an increase in the storage temperature from 6 to 40°C can increase Sb migration by 50%. Most importantly, Sb migration from PET bottles can be increased considerably when non-carbonated water reaches a temperature between 40 °C (Reimann et al., 2012) and 60 °C (Westerhoff et al., 2008; Carneado et al., 2015). Although storage time and temperature are most often investigated, researchers have also found that ultraviolet (UV) exposure, and in-car storage (that combine UV radiation with high temperatures, especially during summer (Westerhoff et al., 2008)) can also increase Sb migration (Keresztes et al., 2009). Additionally, physical PET characteristics (Keresztes et al., 2009), initial concentration of Sb added during polymerisation, and quality of the water filled into bottles (e.g., conductivity and major ion composition) (Westerhoff et al., 2008) could potentially have a higher impact on Sb migration than storage conditions.

3.2.2. Carbonyl compounds

The most prevalent carbonyl compounds found to migrate from PET drink bottles are aldehydes, e.g., acetaldehyde and formaldehyde; with the former compound having received the most attention (Mutsuga et al., 2006). Formaldehyde has been classified by the World Health Organization (WHO) as carcinogenic to humans (WHO, 2018) and acetaldehyde as a probable human carcinogen (WHO, 2017). Both acetaldehyde and formaldehyde are included in the Union list with SMLs of 6 mg/kg and 15 mg/kg, respectively. In PET drink bottles, aldehydes occur as NIAS formed during manufacturing. Both acetaldehyde and to a much lesser extent formaldehyde, can be formed as thermal degradation by-products at the final stage of bottle preform production (polycondensation and melt processing) (Dąrowska et al., 2003; Dogan and Cebi, 2019). This, particularly in blow moulding, results in their enclosure in the bottle wall (Özlem, 2008; Baumjohann and Harms, 2015). Acetaldehyde scavengers are often used as a control measure to reduce the levels of aldehydes produced (Özlem, 2008; Baumjohann and Harms, 2015).

Fig. 4 presents the experimental results on the migration of aldehydes from PET bottles in mineral non-carbonated and carbonated bottled water (Bach et al., 2014; Dąrowska et al., 2003; Dogan and Cebi, 2019; Mutsuga et al., 2006; Nawrocki, Özlem et al., 2002, 2008). The migration of aldehydes is higher in carbonated water than in mineral water (Fig. 4). The positive correlation between water saturation with carbon dioxide (CO₂) and migration of aldehydes has been reported in previous studies (Dąrowska et al., 2003; Nawrocki et al., 2002; Bach et al., 2014; Baumjohann and Harms, 2015; Mutsuga et al., 2006). This might be attributed to the pressure in the bottle (caused by the CO₂ gas), rather than to the presence of CO₂ itself (Dąrowska et al., 2003).

Data and information on aldehydes migration from rPET are limited. One study showed a potential positive correlation between the recycled content in PET bottles and acetaldehyde migration (Baumjohann and Harms, 2015).

A couple of studies that tested acetaldehyde migration from virgin PET bottles into carbonated soft drinks at the stage of production (Linssen et al., 1995) and storage/distribution (Özlem, 2008; Linssen et al., 1995) reported acetaldehyde migration into carbonated beverages at levels higher than the SML (see Table 3). However, elevated levels of acetaldehyde in beverages other than water (e.g., juices) might also be related to the fact that acetaldehyde is naturally present in fruits and vegetables (Özlem, 2008). For that reason, most migration experiments use food simulants to determine the extent of chemical transfer from PET bottles into food, while food samples are selected to investigate mainly the impact of storage conditions.

Table 3

Literature evidence on FCCs migrating from PET articles at levels exceeding their respective SML set by (Commission Regulation (EU) No 10/2011, 2011).

FCCs	FCC type	FCM No ^a	SML (mg/kg) ^a	Migration rate (mg/kg)	Food simulant / real food ^b	Lifecycle stage ^c	PET type	Location	Ref
Nickel (Ni)	NIAS (impurity)	–	0.02 ^d	0.03	3% acetic acid (B)	RE-PRO	Flake	Brazil	(Dutra et al., 2014)
Diethyl phthalate (DEP)	NIAS (impurity)	–	0.01	0.013–0.15	Fermented milk	S&D (4 °C for 3–6 months)	Bottle	Egypt	(Ahmed et al., 2017)
				0.018–0.038	Fruit juice	S&D (4 °C for 3–6 months)			
				0.048–0.063	Soft drink	S&D (4 °C for 6 months)			
				0.024	Distilled water	S&D (7 °C for 1 month)	Bottle	Korea	(Motahari et al., 2012)
				0.032	Fermented milk	PROD	Bottle	Egypt	(Ahmed et al., 2017)
Di-n-octyl phthalate (DnOP)	NIAS (impurity)	–	0.01	0.42–0.56	Fermented milk	S&D (4 °C for 3–6 months)	Bottle	Egypt	(Ahmed et al., 2017)
				0.46–0.82	Fruit juice	S&D (4 °C for 3–6 months)			
				0.53	Soft drink	S&D (4 °C for 6 months)			
				0.072 0.062–0.102	Bottled water Bottled water	PROD S&D (28–44 °C for 7–60 days)	Bottle	Italy	(Notardonato et al., 2019)
Di-isobutyl phthalate (DiBP)	NIAS (impurity)	–	0.01	0.05 0.052–0.114	Bottled water Bottled water	PROD S&D (28–44 °C for 7–60 days)	Bottle	Italy	(Notardonato et al., 2019)
				0.01–0.013	Bottled water	S&D (60 °C for 21–35 days)	Bottle	Thailand	(Leechart et al., 2015)
Dimethyl phthalate (DMP)	NIAS (impurity)	–	0.01	0.07–0.11	Distilled water	S&D (7–45 °C for 4–6 weeks)	Bottle	Korea	
				0.08–0.29	Fruit juice	S&D (4 °C for 3–6 months)	Bottle	Egypt	(Ahmed et al., 2017)
				0.08–0.15	Soft drink	S&D (4 °C for 3–6 months)			
				0.02	Tea	PROD	Bottle	China	(Wang et al., 2018)
Tinuvin 328	Additive	–	0.01	0.02 0.1	Carbonated drink	PROD	Bottle	China	(Wang et al., 2018)
Monohydroxyethyl terephthalate	NIAS (oligomer)	–	0.01	0.017	95% ethanol (D2)	PROD	Tray	NA	(Kim and Lee, 2012)
Second series acyclic dimer				0.062					
Second series acyclic trimer				0.036					
Second series cyclic dimer				0.042					
Second series cyclic trimer				0.015					
First series cyclic trimer			0.01	0.062					
Anthraquinone (SB 104)	Additive (colourant)	–	0.01	0.04–0.06 0.049–0.058	95% ethanol (D2)	PROD	NA	Europe	(O'Brien, 2010)
Quinophthalone (SY 114)	Additive (colourant)	–	0.01	0.02–0.025	95% ethanol (D2)	PROD	NA	China	(O'Brien, 2010)
Butylated hydroxytoluene (BHT)	Additive (antioxidant)	315	3	0.09–0.121	95% ethanol (D2)	S&D			
				3.3–5.59 3.51–6.12	Olive oil (D2) Sunflower oil (D2)	S&D (24–37 °C for 20–60 days)	Bottle	Brussels	(Tawfik and Huyghebaert, 1999)
2-aminobenzamide	Additive (acetaldehyde scavenger)	164	0.05	0.105–0.485 ^e 0.204–0.748 ^e	50% ethanol (D1) 95% ethanol (D2)	RE-PRO	Bottle	Germany	(Gehring and Welle, 2018)
				0.130 ^f 0.161 ^e 0.225 ^e 0.053 ^e	3% acetic acid (B) 10% ethanol (A) 20% ethanol (C) Isooctane				
				0.052–0.055	Carbonated water	S&D	Bottle	UK	(Franz et al., 2016)
				0.0105	95% ethanol (D2)	PROD	Bottle	China	(Cai et al., 2020)
					Carbonated beverage	S&D (40 °C for 2–6 months) PROD	Bottle	Turkey Netherlands	(Özlem, 2008) (Linszen et al., 1995)

^a As specified by the positive list of FCM (Commission Regulation (EU) No 10/2011, 2011).^b See Table 1.^c PROD: Production, S&D: Storage/distribution, RE-PRO: Reprocessing; Re-S&D: Re-storage / re-distribution.^d As specified by (Commission Regulation (EU) 2020/1245, 2020).

^e Concentrations were determined under several migration tests with a variety of contact times and temperatures as provided in (Commission Regulation (EU) No 10/2011, 2011); dash (-): Non-authorized substances / not included in the Union list.

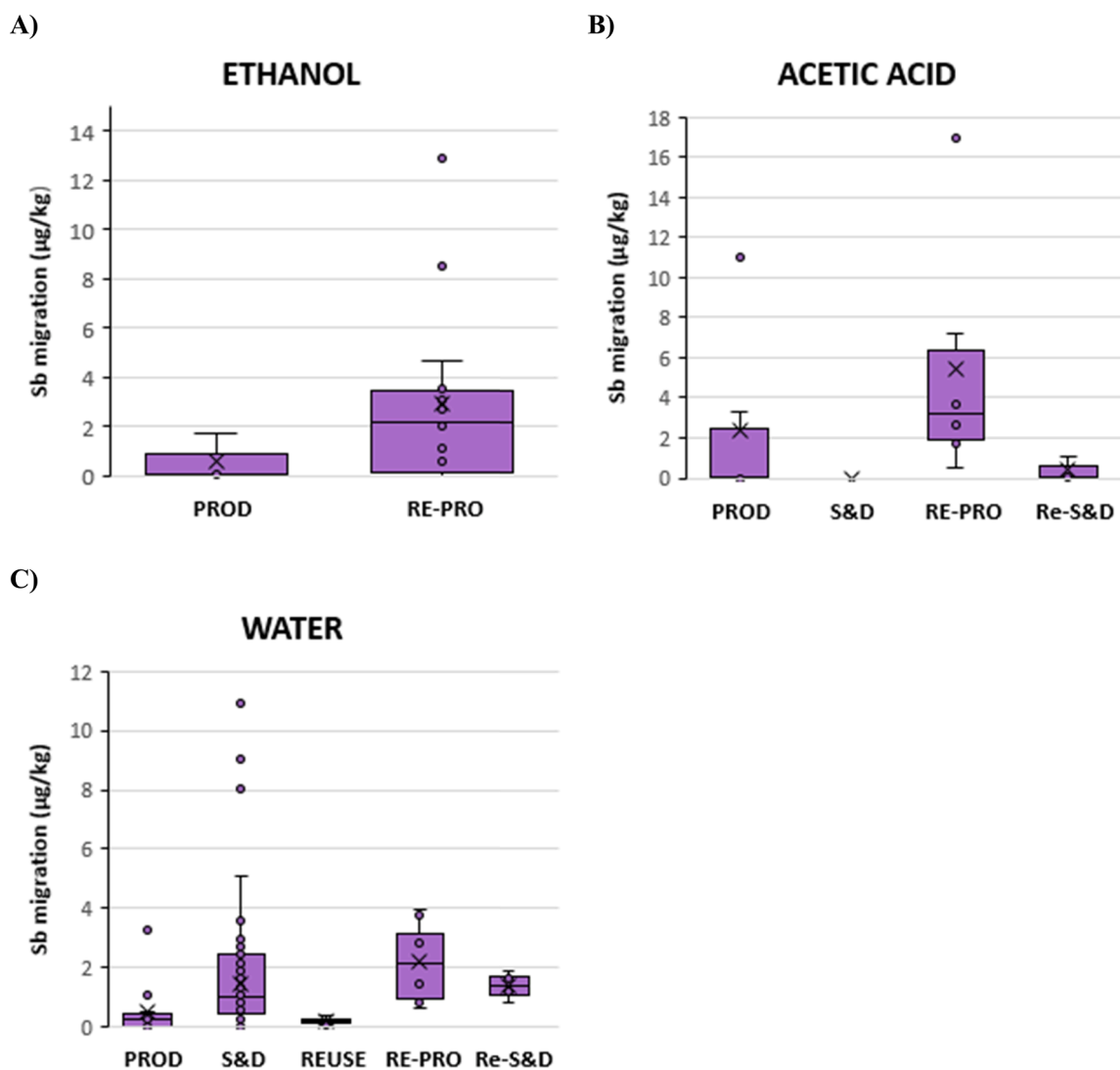


Fig. 3. Variability of migration of antimony (Sb) from PET drink bottles including pellets and flakes into food simulants A) ethanol (10–15% v/v); B) acetic acid (3–5% v/v); and C) non-carbonated water, across all stages of lifecycle (PROD: Production, S&D: Storage/distribution, REUSE: Reuse, RE-PRO: Reprocessing; Re-S&D: Re-storage / re-distribution) based on migration experiments.

Concerning storage/distribution, UV exposure has been reported as one of the most influential factors affecting the migration of aldehydes (Nawrocki et al., 2002; Bach et al., 2014; Baumjohann and Harms, Özlem, 2015, 2008). For example, after 10 days of UV exposure to PET bottles, an off-flavour in carbonated water might occur (Bach et al., 2014). About the storage temperature, migration of aldehydes begins to increase at 20 °C (Dogan and Cebi, 2019), while a storage temperature above 40 °C can significantly increase the migration of acetaldehyde in carbonated soft drinks (Özlem, 2008).

3.2.3. Endocrine-disrupting chemicals: phthalate esters and bisphenol A

Endocrine-disrupting chemicals (EDCs) can induce a disturbance in the endocrine system, leading for example to reproductive disorders, cardiovascular diseases, diabetes, obesity, and cancer in adults (Zoeller et al., 2012; Gore et al., 2015). Phthalates are well-known EDCs with evidence for affecting lower semen quality, neurodevelopment, childhood asthma, low birth weight, endometriosis, decreased testosterone, ADHD, Type 2 diabetes and breast/uterine cancer (Eales et al., 2022).

Phthalates are not chemically bound to polymer chains and therefore can easily migrate from PET bottles (Keresztes et al., 2009; Amiridou and Voutsas, 2011). Dibutyl phthalate (DBP), di(2-ethylhexyl) phthalate (DEHP), and benzylbutyl phthalate (BBP) are included in the Union list, with SMLs of 0.3, 1.5, and 30 mg/kg, respectively, whereas dimethyl phthalate (DMP), diethyl phthalate (DEP) and di-n-octyl phthalate (DNOP) that have also been detected in PET drink bottles are non-authorized substances.

DEHP is the most studied phthalate migrating from PET drink bottles (Ahmed et al., 2017; Amiridou and Voutsas, 2011; Aneck-Hahn et al., 2018; Arfaeina et al., 2020; Cacho et al., 2017; Cirillo et al., 2013; Farhoodi et al., 2008a; Farhoodi et al., 2017; Freire et al., 1998; Jeddi et al., 2016; Leechart et al., 2015; Li et al., 2013; Motahari et al., 2012; Notardonato et al., 2019; Otero et al., 2015; Penalver et al., 2021; Rastkari et al., 2018; Schmid et al., 2008; Wang et al., 2021a; Zaki and Shoeib, 2018; Rafiei Nazari et al., 2018). According to Commission Regulation EU 10/2011, (2011), DEHP should only be used as a plasticiser in repeated use materials and articles contacting non-fatty foods

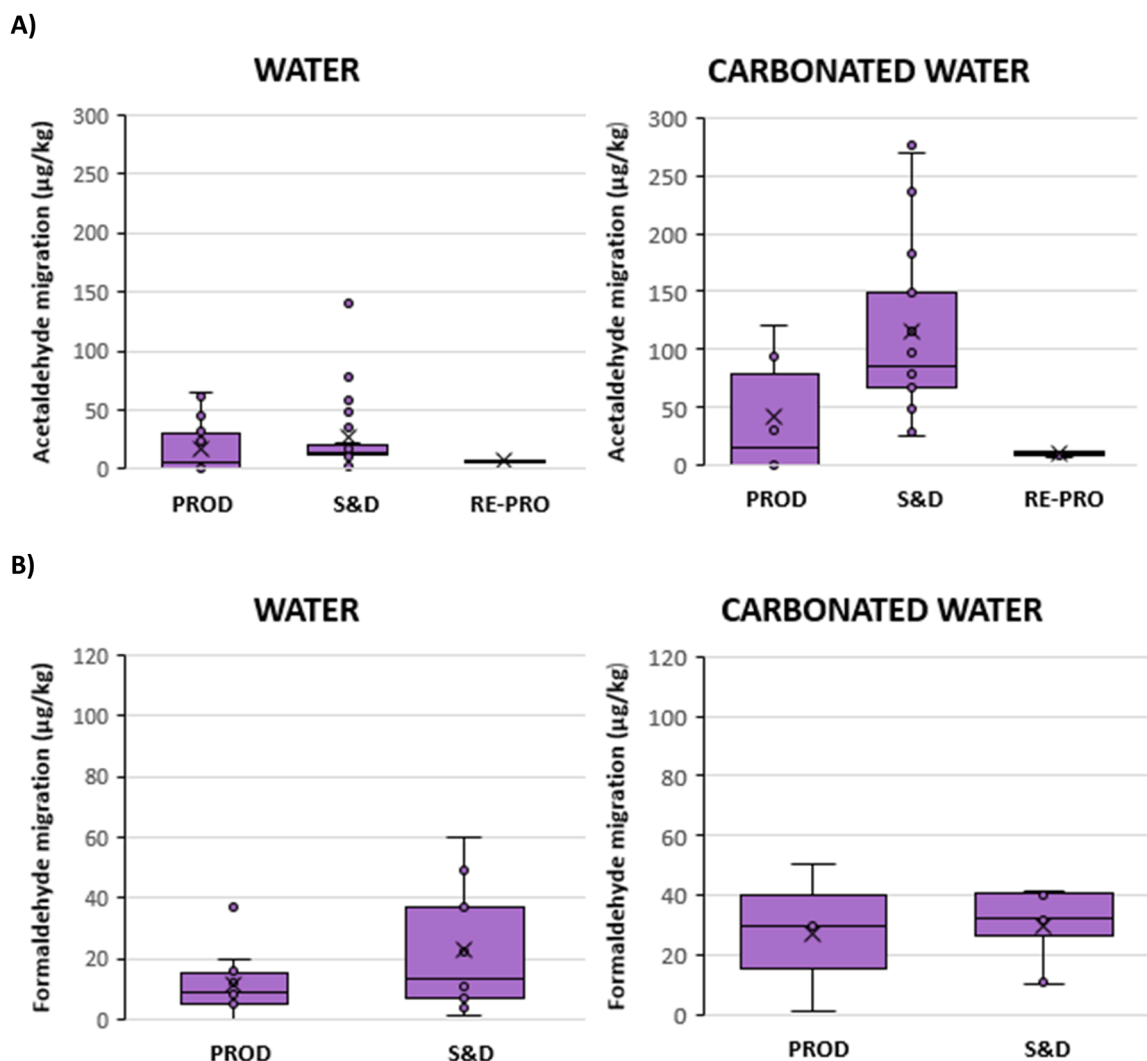


Fig. 4. Migration of aldehydes (acetaldehyde (A) and formaldehyde (B)) from PET drink bottles into non-carbonated and carbonated water at the stages of production (PROD) and storage/distribution (S&D).

and as a technical support agent in concentrations up to 0,1% in the final product. The second most studied phthalate is DBP (Aneck-Hahn et al., 2018; Jeddi et al., 2016; Li et al., 2013; Motahari et al., 2012; Otero et al., 2015; Peñalver et al., 2021; Rastkari et al., 2018; Salazar-Beltrán et al., 2018; Wang et al., 2021a; Zaki and Shoeib, 2018), followed by DMP and DEP (Ahmed et al., 2017; Amiridou and Voutsas, 2011; Arfaeinia et al., 2020; Leechart et al., 2015; Motahari et al., 2012; Notardonato et al., 2019; Salazar-Beltrán et al., 2018; Wang et al., 2021b).

Bisphenol A (BPA), another well-known EDC that has multiple adverse effects on human health at low levels (vom Saal and Vandenberg, 2020), is used as a monomer in the manufacture of polycarbonate and epoxy resins. As BPA is not known to be intentionally used in PET production, it is neither expected to be present nor to migrate from PET bottles (Dreolin et al., 2019; Fan et al., 2014). Nonetheless, many studies detected BPA migrating from PET bottles into beverages (Amiridou and Voutsas, 2011; Aneck-Hahn et al., 2018; Dreolin et al., 2019; Fan et al., 2014; Peñalver et al., 2021). Moreover, Dreolin et al. (2019) reported higher migration levels of BPA from rPET than virgin PET bottles placed in the European market (Dreolin et al., 2019).

The detection frequency (concentration > LoD) was 93.3% for DBP, 89.6% for DEHP, 76.2% for BPA, 52.9% for DEP, 43.9% for DMP, and 44.4% for DnOP, demonstrating that DBP, DEHP and BPA are commonly present in PET drink bottles. The migration of these compounds at the

stage of production, storage/distribution, and reprocessing (only for BPA) is illustrated in Fig. 5. The extreme values reported at the production stage are related to a study that determined the migration of DBP from ten commercial brands, reporting a wide range from <LoD (0.029 mg/kg) to 0.26 mg/kg and suggesting that the variable physical characteristics of PET samples could lead to variable migration of phthalates (Salazar-Beltrán et al., 2018). They reported, for example, a statistically significant correlation between thickness and phthalates content in PET bottles (95% confidence), as well as between DBP content in PET samples and its migration rate in non-carbonated bottled water (90% confidence).

Exposure to individual phthalate compounds via consumption of PET-bottled drinks was estimated to be low. Zaki and Shoeib (2018) found that the contribution of bottled water to TDI levels established by EFSA for DBP (10 µg/kg body weight per day) and DEHP (50 µg/kg body weight per day) did not exceed 0.16% and 0.04% for adults, respectively, and 0.72% and 0.16% for children, respectively. This is in agreement with another study that identified a low non-carcinogenic risk for phthalate intake (DEHP and BBP) via bottled water consumption reporting that PET-bottled water is not a significant contributor to phthalate intake (Jeddi et al., 2016). Ahmed et al. (2017) reported that the migration of phthalates from PET bottles into soft drinks was 3–135 times lower than the TDI. By contrast, Arfaeinia et al. (2020) conducted a risk assessment based on the oestrogenic potential of phthalates

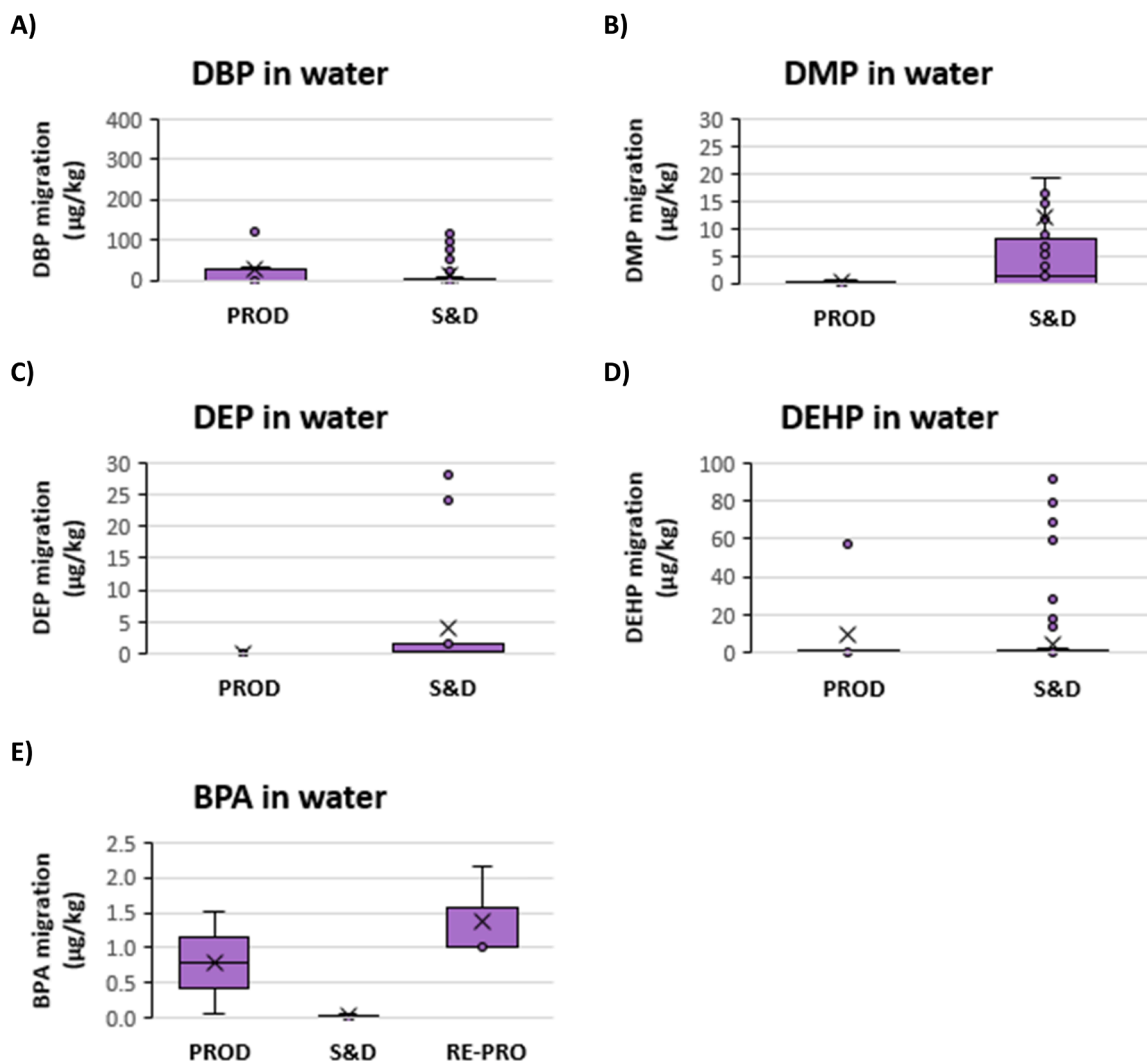


Fig. 5. Migration of EDCs from PET drink bottles into non-carbonated water: A) DBP migration; B) DMP migration; C) DEP migration; D) DEHP migration; and E) BPA migration.

indicating that the consumption of acidic juices in PET containers might induce long-term adverse health effects on consumers. Current human exposure to phthalates at levels well below regulatory thresholds are associated with many adverse effects (Maffini et al., 2021), indicating that existing TDIs may not be sufficiently protective.

The migration rate of EDCs in juices and carbonated drinks is reported to be related to the molecular weight of the migrant that determines its solubility in the food simulant, rather than the interaction between polymer and simulant (Farhoodi et al., 2008b). With phthalates having a higher solubility into fatty (Li et al., 2013) and acidic drinks (Rastkari et al., 2017), their migration rate in these drinks is expected to be higher. Arfaeinia et al. (2020) reported a statistically significant negative correlation ($p < 0.05$) between pH and the concentration of phthalates in bottled drinks, with DEHP migration from PET yoghurt drink bottles being reported to approach the SML.

The influence of several storage conditions on the migration of phthalates has also been examined. Temperature increase, storage duration and UV exposure have been found to increase the migration of phthalates. For example, Notardonato et al. (2019) recommended avoiding prolonged UV exposure to PET bottles. Storage temperature below 25 °C in an environment away from UV exposure has been suggested to minimise the migration of phthalates from PET bottles into still water (Jeddi et al., 2016). In addition, positive correlations between the migration of phthalates in non-carbonated water and acidic juices with

storage time and temperature have been found (Wang et al., 2021a; Zaki and Shoeib, 2018; Arfaeinia et al., 2020). A recent study found that DEHP might reach the maximum level of acceptable carcinogenic risk (10^{-6}) under 8.8 days storage at 40 °C of bottled water, or 7.7 days at 50 °C, or 6.1 days at 60 °C (Wang et al., 2021a). Likewise, an increase of storage duration and temperature, and particularly above 40 °C can enhance the migration rate of BPA from PET bottles (Aneck-Hahn et al., 2018).

3.3. FCCs across the lifecycle of PET bottles

Critical aspects relevant to FCCs migration from PET bottles were grouped according to the lifecycle stage at which they were reported including the production, storage/distribution, reuse and reprocessing (Fig. 6). These were examined to identify knowledge gaps and actions required for the transition towards more sustainable management.

3.3.1. Production

3.3.1.1. Virgin PET. FCCs can either derive from substances that are used as aids (i.e. additives and catalysts), or from basic PET components (i.e. the monomers EG, TA, and DMT), their by-products (produced during processing) and/or residual contaminants present in raw materials or processing equipment (see Supporting Information – B).

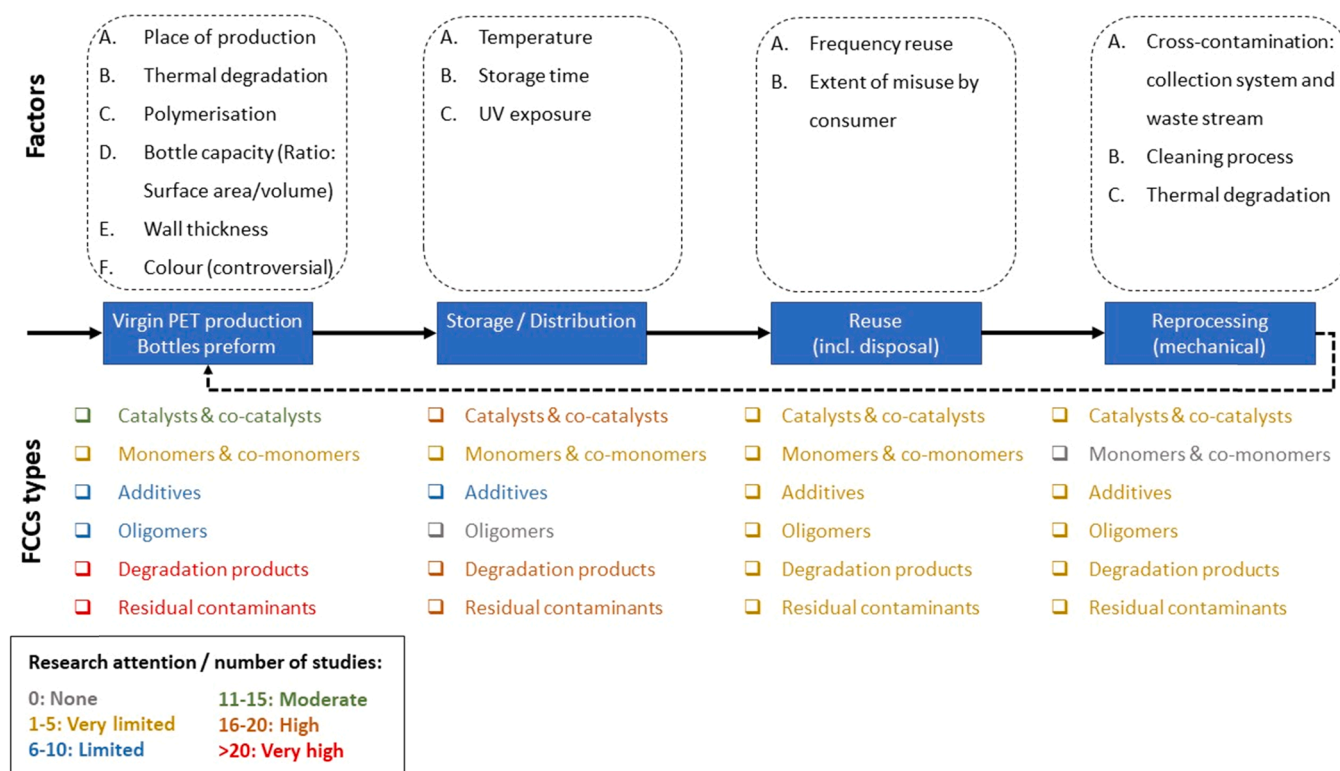


Fig. 6. A simplified overview of the types of FCCs recorded at each stage of the PET drink bottles value chain from production to reprocessing (closed-loop) with a focus on the influential factors on FCCs migration including related research attention to the types of FCCs from PET bottles at each stage.

Oligomers (i.e., substances composed of a few PET monomers), for example, constitute some of the most common NIAS in PET polymers. The migration of 14 oligomers from PET bottles was recently reported, highlighting the need to characterise the composition of oligomer in virgin PET polymer production and the material's suitability for food contact applications (Ubeda et al., 2018).

The country of origin of PET bottles was found to be relevant for the migration of FCCs. Schmid et al. (2008) measured the migration of phthalates into distilled water contained in PET bottles purchased from around the world (Switzerland, Honduras, and Nepal), and found a statistically significant difference among countries (e.g., significantly higher DEHP migration from bottles in Honduras than from bottles in Switzerland). The country of origin of bottles was reported as the most distinctive parameter (against temperature and UV exposure) for phthalate migration using distilled water as a simulant (Schmid et al., 2008). Considerable differences were found in the Sb migration rates in non-carbonated water among PET bottles purchased from several European countries (i.e., Germany, France, Switzerland, Finland, Czech Republic, Denmark, Spain, Poland, Belgium, Netherlands, and Italy (Shotyk et al., 2006). Sb leaching can also significantly vary among different commercial brands of drinking water (Chapa-Martínez et al., 2016). Sb migration from PET bottles into the water was higher for international water brands compared to national (i.e., Spain) brands under identical storage conditions (Payán et al., 2017).

The physical properties of PET bottles may also affect the migration potential of FCCs into bottled drinks. Characteristically, the capacity (i.e., filling volume) of PET bottles has been reported as an influential factor on FCC migration. Dogan and Cebi (2019) reported a higher migration rate of Sb, cobalt, and acetaldehyde in non-carbonated water contained in small PET bottles (i.e. 0.5 L) as opposed to bottles with a high capacity (i.e. 1.5 and 5 L). This is attributed to the correlation of migration ability with the ratio of surface area to water volume (Zmit and Belhaneche-Bensemra, 2019; Keresztes et al., 2009). Likewise, the thickness of PET bottles has been correlated with the migration of Sb

(Zmit and Belhaneche-Bensemra, 2019) and phthalates (Salazar-Beltrán et al., 2018): the thicker the plastic walls, the higher the migration rate which is expected since migration levels are most strongly determined by concentration in the FCM (Biryol et al., 2017). The colour of PET bottles and its influence on FCCs migration remains unclear. Some studies reported that the colour of PET bottles had no effect on the Sb migration rates (Payán et al., 2017; Shotyk and Krachler, 2007), while Carneado et al. (2015) found slightly higher leaching rates of Sb in coloured bottles than in clear bottles.

3.3.1.2. Virgin PET mixed with varying amounts of rPET (recycled content). Research comparing the migration of FCCs between virgin PET and rPET is still relatively sparse. Some authors report higher migration of some chemicals from rPET whilst others found no differences. Ubeda et al. (2018) observed no difference between virgin PET and rPET pellets with regards to cyclic and linear oligomers migration. Heavy metals (i.e., Pb, Cd, Cr, Ni and Sb) may be present in rPET articles as a result of cross-contamination by other plastic components and products such as polyvinyl chloride (PVC) (Whitt et al., 2016). Whitt et al. (2016) nonetheless concluded that the consumption of rPET articles is safe concerning the leaching of these inorganic elements (i.e. heavy metals) since their migration levels were below the LoD. Moreover, Welle and Franz (2011) reported that the levels of Sb used in PET bottle manufacturing lead to a low migration from both virgin and 100% recycled PET bottles into beverages and edible oils that would not exceed 1% of the current TDI (6 µg/kg body weight per day (WHO, 2017)). Nonetheless, Thoden van Velzen et al. (2020) reported a clear relationship between the migration of two NIAS - benzene and styrene - into non-carbonated water and the recycled content of PET bottles. Their presence in the rPET matrix originated from the thermal degradation of polystyrene (i.e., styrene) and PVC (i.e., benzene) products (Thoden van Velzen et al., 2020). In addition, the migration of BPA from rPET into 20% ethanol and 3% acetic acid was found to be increased with higher recycled content (0–100%) (Dreolin et al., 2019).

3.3.2. Storage / distribution

Storage time, temperature, and UV exposure are the variables mostly evaluated in the literature with regards to the impacts of storage/distribution on the migration of FCCs from the PET bottle to the foodstuff. Temperature increase can lead to enhanced polymer degradation, which in turn accelerates the speed of diffusion of additives from the polymer wall to the foodstuff (Leechart et al., 2015). However, the extent to which storage factors affect the migration rate differs from one FCC to another – e.g. UV exposure was found to be more influential on aldehydes migration than other storage factors, while the temperature is more influential on Sb migration. This highlights the need for these factors to be separately investigated for each FCC. To date, storage conditions have been investigated mainly for Sb, carbonyl compounds, phthalate esters and BPA, and to a lesser extent for other FCCs, such as metals (Reimann et al., 2012), colourants (O'Brien, 2010), antioxidants (Tawfik and Huyghebaert, 1999), monomers (Farhoodi et al., 2008b), volatile organic compounds (Pandey and Kim, 2011) and other additives (Franz et al., 2016; Begley et al., 1991; Wang et al., 2018).

3.3.3. Consumption / reuse

The reuse of PET drink bottles before their disposal is quite common particularly in developing countries, but the influence of reuse, and its frequency, on FCCs migration has not received much attention to date. Andra et al. (2011) found that the frequency of reuse can have a greater impact on chemical leaching from PET drink bottles than storage conditions. These authors examined the migration of Sb and bromine (Br) from PET bottles into water under a range of 0–27 rounds of reuse and reported that the frequency of reuse was the most significant factor among UV exposure, storage time and temperature. Specifically, a reuse up to 20 times can increase Sb and Br migration up to 2–3 times under identical storage conditions (Andra et al., 2011). Konkol (2004) added that sorbed compounds can migrate from PET bottles during reuse. In contrast, Jetten and Kruijff (2002) reported no significant change in the migration rates of PET oligomers, monomers and other degradation products into several simulants (i.e., 3% acetic acid, 50% and 95% ethanol, olive oil and isooctane) by the repeated use of PET bottles (simulated by subjecting the PET bottles to repeated cycles of washing procedures) (Jetten and Kruijff, 2002; Jetten, 1999).

In developing countries, consumers could sometimes refill empty PET bottles with highly acidic (e.g., vinegar and lemon juice), or fatty (e.g., oil) foodstuffs and may keep them under adverse storage conditions (Arfaeina et al., 2020). These types of foodstuffs could enhance the migration of FCCs, e.g. PET oligomers (Ubeda et al., 2018) and phthalates (Li et al., 2013; Rastkari et al., 2017), which could be further exacerbated by the reuse frequency and storage conditions. For this reason, UV exposure, extreme storage temperature, and storage time of more than three months should be avoided (Notardonato et al., 2019; Leechart et al., 2015). The presence of sodium chloride (NaCl) content in aqueous solutions was found to decrease the migration of Sb and phthalates due to their reaction with NaCl (Leechart et al., 2015). A common practice in developing Asian countries is the reuse of PET bottles by filling them with condiments (e.g. fish sauces). Characteristically, Leechart et al. (2015) demonstrated that fish sauces might contain lower amounts of these FCCs compared to distilled water due to considerable amounts of salt contained therein (ca. 23–28%).

3.3.4. Reprocessing

The impact of post-consumer PET bottle reprocessing on FCCs migration has remained largely underexplored. Still, contamination of post-consumer PET-bottle waste has already been linked to: i) design components, such as labels, adhesives and additives (Dutra et al., 2014); ii) misuse of PET bottles by consumers (EFSA, 2011); iii) cross-contamination from non-food contact PET articles and other plastics at the collection (due to kerbside collection, commingled vs separated; household waste recycling centres (HWRCs), also known as 'civic amenity sites'; and bring sites/banks), sorting (due to techniques

employed and size of material recovery facilities) and reprocessing stages (due to technologies efficiency) (Hahladakis et al., 2018b); iv) co-occurrence of impurities (e.g. glass, paper, metals) in PET recycling streams; v) sorption of food and non-food residues into PET drink bottles (Dutra et al., 2014; Thoden van Velzen et al., 2020; EFSA, 2011); vi) formation of degradation products and oligomers during thermal reprocessing (Thoden van Velzen et al., 2020); and vii) use of certain chemicals in the reprocessing stage such as detergents and caustic soda (Schyns and Shaver, 2021; Welle, 2011). All these factors can downgrade the rPET quality (Hahladakis and Iacovidou, 2018; Iacovidou et al. 2019). Cross-contamination of PET FCs with non-food PET and other plastic components and products that may accidentally enter the PET waste stream has received the most attention. According to EFSA guidelines on plastics mechanical recycling, the fraction of non-food PET articles in the input reprocessing stream of rPET bottles should not exceed 5% (EFSA, 2011). However, the proportion of non-food post-consumer PET articles may be highly variable depending on the kerbside collection system used that may contain ca. 0.04–6% non-food PET articles (Franz and Welle, 2020). This can delimit the potential of some decontamination technologies to sufficiently process post-consumer PET bottle waste. The reason for this can be two-fold: firstly, due to the conditions outlined above, and secondly, due to the technological performance of the reprocessing infrastructure.

Several cleaning processes can be employed for the decontamination of post-consumer PET drink bottles, e.g. a) conventional and b) super-cleaning processes, providing different quality of resulting PET recyclates. Conventional cleaning, or decontamination, is a water-based washing technique that uses 2–3% caustic soda and 0.5% detergents at relatively high temperatures (70–90 °C) applied to PET flakes grounded from post-consumer bottles (Hossain and Mozumder, 2018; Welle, 2011). Conventional washing is sometimes combined with separation steps to remove non-PET material, such as polyolefins, and the efficiency of the process depends on the residence time of flakes washing, the water temperature and concentration of washing additives (Welle, 2011). However, conventional washing is a surface cleaning process not able to remove organic components absorbed into PET. Therefore, conventionally washed PET flakes might not be suitable for food contact recyclates, even if re-melting and re-extrusion are applied to the washed flakes (Welle, 2011). Conventionally recycled PET flakes can be used in food contact applications if they are covered by a layer of virgin PET as a functional barrier to prevent contaminant migration into food. However, the virgin PET layer can also be contaminated with post-consumer contaminants by the recycled layer due to high temperatures at the stage of production (Welle, 2011).

Super-cleaning decontamination can be applied to conventionally washed PET flakes and consists of three typical processing steps: high-temperature treatment; vacuum or inert gas treatment; and surface treatment with non-hazardous chemicals (Welle, 2011). There are several commercially available super-cleaning processes able to decontaminate post-consumer PET at levels similar to virgin PET since low molecular weight contaminants and/or volatile solvents can be efficiently removed by high-temperature treatment (Welle, 2011; Franz and Welle, 2020). The decontamination efficiency of super-cleaning recycling without artificial contamination can be assessed by using limonene, a natural fragrant characterised as NIAS at very low concentrations but always detectable in post-consumer PET bottles (Thoden van Velzen et al., 2020; Horodytska et al., 2020), as an internal indicator (Welle, 2009).

The diversity of technologies indicates variability in the efficiency of post-consumer PET decontamination processes, and, thereby, variable rPET quality. Dutra et al. (2014) investigated the migration of inorganic elements and non-volatile organic compounds in post-consumer rPET pellets and flakes subjected to several cleaning processes, such as conventional cleaning (i.e., use of water), deep-cleaning (i.e., use of detergent for hot caustic washing, friction washing and drying), extrusion, and super-cleaning (i.e., a combination of deep cleaning, extrusion and

solid-state polycondensation). They showed that there was a significant reduction in the migration of inorganic elements and non-volatile organic compounds when deep- and super-cleaning processes were applied (Dutra et al., 2014). It is recommended that a super-cleaning process should be employed for rPET that was originally collected under conditions of limited control over cross-contamination (Dutra et al., 2014) (e.g. highly heterogeneous waste streams: municipal solid waste (MSW) versus commercial and industrial waste (C&I); and/or collection systems with low efficiency: kerbside collection vs deposit return schemes (DRS)). However, none of these cleaning processes was able to eliminate the migration of non-volatile organic compounds including both IAS and NIAS, indicating the need for a more efficient PET recycling chain (Dutra et al., 2014).

Since 2010, EFSA has published 133 favourable Scientific Opinions on the recycling of post-consumer PET into FCM, and more than 180 applications have been evaluated so far (EFSA, 2022). These processes were based on more than 40 different decontamination technologies consisting of a combination of cleaning treatments (e.g., conventional cleaning, pre-drying, infra-red treatment, crystallisation, extrusion, and solid-state-polycondensation). In these evaluations, the decontamination efficiency was tested for specific hazardous chemicals, known as surrogates. These “surrogates are substances with different molecular weight and polarity” (EFSA, 2008) and they represent general chemical contamination of post-consumer plastics. Examples of surrogates recommended by the US FDA are toluene, benzophenone, lindane and chlorobenzene (US FDA, 2021). Surrogates were monitored during the recycling processes and their levels found in plastics after the processes demonstrated different decontamination efficiencies, depending on the technology and surrogate. EFSA Scientific Opinions also highlight that operating parameters (e.g., temperature, residence time, pressure and gas flow) are critical for the performance of cleaning treatments and therefore they determine the decontamination efficiency (EFSA, 2022).

According to Cheng et al. (2010), metal contamination in PET reprocessing is mainly attributed either to contaminants remaining on the plastic surface, which can be easily removed by rinsing, or to catalyst residuals used in the stage of manufacturing which cannot be easily removed. Based on this perspective, two parallel experiments for metal migration in rPET bottles were conducted to evaluate the effect of several treatments such as cooling with ice-cold water, heating with boiling water, microwaving, incubation with low-pH water, outdoor sunlight irradiation, and in-car storage; with the sample washing before each treatment, or not, being the changeable variable (Cheng et al., 2010). Notably, the washing procedure that was applied before treatments reduced the migration of Sb, suggesting that the contamination of rPET bottles at the stage of reprocessing could be avoided by washing rPET bottles before first use.

4. Discussion

The study reveals that 150 FCCs could be migrating from PET bottles, 18 of which may exceed EU regulatory limits (Table 3). Of these 150 FCCs, 109 are non-authorized substances, while 48 and 44 FCCs are not included in the FCCdb, a database of over 12,400 FCCs, and CPPdb, a database of over 4200 plastic packaging-associated chemicals, respectively. Most of these substances are NIAS: in fact, 113 out of 150 detected FCCs are NIAS. Additional chemicals that have not been studied or reported on, are also likely to be migrating from PET, and there is insufficient information available in the public domain. Additionally, only 8 and 29 of the detected 150 FCCs in PET could be specified as related to PET based on information in the FCCdb and CPPdb, respectively. However, it should be clarified that none of these two databases provides comprehensive and polymer-specific detailed information. As an example of additional information, a recent review reported that acetaldehyde, formaldehyde, 2-methyl-1,3-dioxolane, PET oligomers, toluene, xylenes and cyclopentanone are mainly present as NIAS in PET bottles (Horodytska et al., 2020).

While the current legal limit for non-authorized substances (10 µg/kg) can be applied for the migration of non-authorized IAS behind a functional barrier (that are not CMR), it could be misinterpreted as a toxicologically relevant limit, leading to underestimating the actual risk of NIAS for which hazards have not been identified and characterised. Notably, this regulatory threshold is a pragmatic limit value and not a science-based toxicological threshold. Official guidance by authorities on risk assessments of NIAS has not been provided so far, and NIAS assessments are currently based on limited toxicity data or in silico predictions which are associated with high uncertainty (Van Bossuyt et al., 2017, Bschor, 2017). This indicates a need for advances in analytical techniques and exposure assessments, as well as developing databases for the identification of NIAS (Geueke, 2018).

Our in-depth analysis of the PET-related findings from the systematic evidence map revealed that researchers have placed increased focus on specific FCCs including Sb, aldehydes and particularly acetaldehyde, and certain EDCs (DBP, DMP, DEP, DEHP and BPA). Increased scrutiny of these FCCs may have been driven by an intended focus on FCCs that could provide insights on the typical quality of PET in terms of contamination (i.e., Sb), quality (i.e., aldehydes) and adverse health effects (i.e., EDCs). Sb migration from PET bottles into non-carbonated water was found not to exceed the EU regulatory limit (SML: 40 µg/kg) (Fig. 3) according to FCMs regulation, but it was found to exceed the regulatory limit for natural mineral water (i.e., from natural underground reservoirs and springs) of 5 µg/L set by the EU regulation (Commission Directive, 2003/40/EC, 2003) under certain conditions. This points to questions of practice, whereby on the one hand Sb migration in PET-bottled water is considered to be safe at 40 µg/kg, whilst regulation on drinking (tap) water is more stringent. Moreover, this is particularly questionable in light of findings of carcinogenicity from consumption of waters with Sb content below 20 µg/L (Colak et al., 2015). The difference between regulatory limits for bottled and tap water further suggests that politics may underlie the bottled water provision in the food packaging system (Gerassimidou et al., 2021). Bottled water provision and consumption is the fastest growing market segment in the beverage industry globally, valued at USD 217.66 billion in 2020 and is expected to see an annual growth rate (AGR) of 11.1% from 2021 to 2028 (Hawkins, 2009; Bevindustry, 2021; GVR, 2021). In Europe, bottled water and soft drinks are experiencing an 11% increase in 2019, making it one of the most lucrative segments of the beverage industry (FoodDrinkEurope, 2020).

The increased attention on the potential migration of aldehydes from the PET bottle to the beverage (Fig. 4), usually water, can also be explained by the politics of bottled water provision. Aldehydes, and particularly acetaldehyde, can be highly soluble in water (Linszen et al., 1995). Acetaldehydes migration from the bottle to the water even at low levels can result in an undesirable odour and off-taste, described as fruity and plastic-like (Baumjohann and Harms, 2015). This can make PET bottle manufacturers and water fillers (i.e., shareholders upstream in the value chain) particularly anxious about their brand image as it can affect the marketability of their product. The main premise on which bottled water is being sold globally is due to claims that bottled water is healthier, safer, and tastier than tap water (Hawkins, 2009; Hu et al., 2011). Clearly, this is not the case; tap water is controlled much more stringently (Commission Directive, 2003/40/EC, 2003). Nonetheless, bottled water is now a major commodity associated with convenience and status, and is in high demand; if the above qualities are not upheld by the brands, this could severely impact their profit margins and the product proliferation of the bottled water industry. Understanding the conditions under which aldehydes are present, and migrate from, the PET bottles, are very important to the bottled water industry.

Phthalate esters and BPA are not expected to be present in PET bottles, but they are prevalent at all stages of the PET bottles value chain (Fig. 5). Their presence signifies that these substances are ubiquitous in the environment, and thus, widely dispersed in the FCM system (Wee and Aris, 2019). In FCMs, these compounds can be used as plasticizers

(Salazar-Beltrán et al., 2018; Otero et al., 2015), and maybe also found in adhesives, paints, inks and adhesives (Otero et al., 2015; Amiridou and Voutsas, 2011) and cap sealing resins (Keresztes et al., 2013). Packaging practices for branding purposes and marketability, such as PET drink bottles with colourful labels, as well as the caps used (Cincotta et al., 2018) may be sources of migration for these substances. The presence of phthalates and BPA in PET bottles may also arise from the machinery, and raw material contamination in the bottling factory (virgin PET) (Keresztes et al., 2013), as well as cross-contamination during disposal, collection and reprocessing (post-consumer PET bottle to rPET) (Dreolin et al., 2019). Despite the reported low migration levels of phthalates detected in PET-bottled water (below the TDI), attention should be given to the combined and cumulative effect of the continuous exposure to EDCs, via the consumption of beverages as well as from other sources (Hahladakis et al., forthcoming). Recently, the Consortium Linking Academic and Regulatory Insights on BPA Toxicity (CLARITY-BPA) project – a collaborative programme among US agencies and academic researchers – has shed light on that aspect revealing that low-dose exposure to BPA, a well-studied EDC, might induce adverse health effects at doses below the regulatory limits (2.5 µg/ kg weight per day versus the current TDI of 4 µg/ kg weight per day) highlighting the need for the re-establishment of ‘safe’ limits by regulators (Vandenberg et al., 2019; Patisaul, 2020; Kumar et al., 2020). And recent studies have questioned whether current safety limits for phthalates are sufficiently protective of human health, as adverse effects in humans have been found up to 8000 times below the threshold (Maffini et al., 2021, Trasande et al., 2022, Eales et al., 2022). These statements are further supported by the context of a recent EFSA scientific opinion which proposes a lowered TDI for BPA which is 100,000 times below the current TDI – this implies that the SML for BPA will be lowered significantly, and this, in turn, would have consequences also for systemic contamination of rPET (EFSA, 2021). As the new scientific evidence of harm unfolds in the case of BPA, this could be the case potentially for many more chemicals in the future. In PET bottles manufacturing, the positive correlation of thickness and bottle capacity with FCCs migration (i.e. the thicker the bottle the higher the migration), has been favourably viewed as “killing two inefficiency birds with one stone” (Section 3.3.1). This is because the industry is keen on making reductions in the thickness of the bottle to meet their ‘sustainability’ commitments. However, there are further trade-offs related to the thickness of the bottle; in the case of non-carbonated water, the thinner the bottle, the more likely it is for FCCs in labels and adhesives to migrate to the bottled water. This, in turn, means that bottles with large labels or shrink-wrap, as well as storage conditions, can impact the FCCs’ migration potential. The thickness of PET bottles is also associated with the drinks’ quality as it prevents the oxidation of the contained liquid (e.g. juices) or provides the mechanical properties necessary to withstand the gas pressure (Becerril-Arreola and Bucklin, 2021). For example, a PET bottle with an average capacity of 1 litre filled with carbonated beverage requires 3.13–10.28 g PET, and with an acidic beverage (i.e. juices) requires 17.80–25.40 gr, while a bottle with the same capacity (i.e., 1 L) for non-carbonated water requires 3.21 g on average (Becerril-Arreola and Bucklin, 2021). This indicates the interdependency among product category (i.e., type of bottled drink), PET physical characteristics and FCCs migration.

It is conceivable that storage conditions affect the migration rate of FCCs from PET bottles, but in the published literature, a focus is placed on storage time, temperature and UV exposure (Section 3.3.2). However, there is also evidence supporting that the origin of PET bottles may also influence FCCs migration (Payán et al., 2017; Shotyky et al., 2006; Schmid et al., 2008). Therefore, the geographical origin of PET bottles can be an indicator of PET-bottled drinks quality from a chemical perspective since the amount of IAS (Zmit and Belhaneche-Bensemra, 2019; Westerhoff et al., 2008), the physical characteristics of PET bottles, the quality assurance measures obtained during PET (re)-processing and/or trading (i.e. control of FCCs migration) can strongly differ from

region to region. Better monitoring of the global cross-border flows of plastic PET drink bottles and rPET from a chemical perspective could provide a better understanding of the role of geographical origin in FCCs migration from PET bottles. A high-performance mechanism to control the quality of rPET is also needed to increase rPET use. At present, the lack of data traceability and harmonisation on regulations on the quality of rPET creates uncertainty over its use. More research should be dedicated to addressing the identified data gaps, which may be strengthened through closer collaboration between academic scientists and industry.

The presence and fate of IAS and NIAS during the reprocessing of post-consumer PET bottle waste, and in the resulting rPET, has not yet been fully elucidated. Existing literature highlights that the reprocessing stage may lead to a downgrade in the quality of the output material (rPET). NIAS can migrate from rPET bottles into foodstuff and some may also deteriorate material properties and/or enhance polymer degradation (Möller et al., 2008). For example, acetaldehyde and oxygen scavengers at the reprocessing stage can be partially degraded during recycling, turning rPET flakes brown and leading to their use in the production of lesser quality products. Further, NIAS with low molecular weight, such as degradation products mainly formed during reprocessing, have higher migration potential (Möller et al., 2008; Welle, 2011; Delva et al., 2018). The inclusion of IAS (e.g., antioxidants, plasticizers, chain extenders and fillers) and compatibilization processes (e.g. blending technologies), can potentially minimise this degradation of the rPET material properties. However, there is a lack of standards for grades of recycled polymer, and these polymer degradation minimising approaches might bring significant variations in the quality of the rPET produced by different mechanical reprocessing facilities, resulting in price fluctuations for rPET (secondary material) (Schyns and Shaver, 2021). Currently, recyclers remain in a regulatory grey zone arising from a complex interplay between regulations on the chemical safety of plastic FCM and recycling targets (De Tandt et al., 2021). This reality, along with practical and logistical aspects, make bottle-to-bottle recycling difficult (De Tandt et al., 2021). The ongoing revision of the EU’s FCM regulation (European Commission, 2021) reveals several gaps concerning the assessment processes of FCMs. In this regard, it is important to note that the exact chemical composition of rPET is essentially not known unless measured. Certain hazardous FCCs may or may not be present in rPET on the market and only a case-by-case analysis can establish the presence, levels and safety risks of potential contaminants of concern.

Shareholders in the value chain of PET drink bottles (i.e., recyclers and soft drinks companies) are required to comply with the EC regulation in FCMs (Commission Regulation (EU) No 10/2011, 2011, Commission Regulation (EU) No 282/2008, 2008), and therefore need to meet criteria for migration limits, NIAS risk assessment requirements, and receive an EFSA opinion in favour of authorising the recycling process (EFSA, 2011). Still, a maximum level of recycled content in PET drink bottles in terms of food safety and quality cannot be specified without further risk assessment of the migrating NIAS (Thoden van Velzen et al., 2020). Additionally, the variety of decontamination processes that are commercially available (Welle, 2011), and therefore the efficiency of reprocessing (Dutra et al., 2014) along with the existence of numerous contamination sources in post-consumer PET bottles at the stage of disposal, collection and sorting, make the definition of an optimum recycled content in PET bottles from a chemical perspective challenging.

The use of DRS is a step in the right direction but requires investment and consideration of where reverse vending machines should be placed. Forward-thinking and collaborative initiatives are required for making it easy for consumers, in the household and on the go, to give away, or drop into different collection points (perhaps small shops), their PET bottle waste for a deposit, rather than disposing of it to street bins. Besides the use of DRS systems, design-for-recycling is key towards increasing the circularity of PET bottles. The quality of rPET is determined by the homogeneity of feedstock input into reprocessing (Allassali

et al., 2021). A recent study on post-consumer plastic packaging recycling in the Netherlands revealed that chemical contamination of recycled plastics originated mainly from their design components (e.g., adhesives and inks) (Brouwer et al., 2019). This evidence implies that highly recyclable products, such as PET drink bottles, can be inapt for closed-loop recycling when poorly designed, indicating the need for greater adoption of design-for-recycling principles and improvements at the waste management infrastructure level (Brouwer et al., 2019). Improving PET drink bottles' design, complemented by efficient decontamination technologies, can streamline the production of good quality rPET (Alassali et al., 2021). The end-of-life phase of plastic FCMs, including that of PET drink bottles, must thus be considered at their start-of-life phase (conception and design) as this is essential for achieving a circular plastics economy (BPF, 2016). What's more, better communication between all the direct and indirect stakeholders in the PET bottles value chain is required to realise progress in the right direction (Gerassimidou et al., 2021).

5. Conclusions

To date, initiatives that aim to increase the recycling of PET bottles are ongoing worldwide, while the safety and quality implications arising from reprocessing of PET bottles remain underexplored. By unpacking the PET drink bottles value chain to understand chemical challenges, we revealed a long list of FCCs that can migrate from PET drink bottles, collected from several geographical regions and time periods, which raises questions about the safety of PET drink bottles and the quality of rPET produced. This can be linked to the lack of official guidance on risk assessment of NIAS and insights into their potential hazards, as well as the lack of safety and quality standards at the reprocessing stage or clarity on how these are implemented and monitored. These issues are important gaps that need to be considered by the regulatory community to capture and mitigate the safety risks across the entire lifecycle of PET drink bottles and in closing the loop.

Thus far, emphasis has been placed on chemicals that are of interest to the PET bottles manufacturing industry (Sb, aldehydes, phthalates and BPA). This bias could be influenced by the 'politics' of bottled drinks manufacturers. There is an economic forfeit for making sure rPET is safe to use, a cost that both the production and recycling industries will need to tackle very soon to make PET bottle-to-bottle recycling successful and safe. At the production stage, solutions include rethinking the thickness of the bottle and designing its labelling (branding), whilst at the management (recycling) stage, solutions could be oriented towards investing in super cleaning processes for minimizing the likelihood of rPET to contain higher levels of hazardous chemicals than virgin PET. At the consumption stage, retailers, local governments and consumers need to coordinate their efforts to keep the PET drink bottles in good storage conditions and collect the post-consumer PET bottles in a way that maximises their value. This points to the need for collaboration across the entire value chain to ensure that the following key requirements are met to increase the use of recycled content in PET bottles whilst ensuring chemical safety:

- i) 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);
- ii) monitoring the presence of chemicals at the PET bottles production/filling stage (i.e., impurities of hazardous chemicals in raw materials and machinery contamination);
- iii) controlling the storage conditions that bottled drinks are subjected to (e.g., controlled temperature, UV exposure, storage time and outdoor (i.e., ambient) conditions);
- iv) promoting consumer behaviour change (e.g., use bottles with higher capacity and eliminate misuse of repeated-use bottles);
- v) improving the collection, sorting and reprocessing infrastructure of PET drink bottles (e.g., separate collection, DRS, sorting technologies, super-cleaning decontamination technologies) and introducing a traceability mechanism;
- vi) reaching bilateral agreement on what constitutes good quality rPET between the industry and the regulator and putting in place a compliance mechanism; developing a monitoring mechanism to control cross-border flows and quality of finished goods (drinks), PET preforms and rPET in compliance with i), v), vi);
- vii) revising the chemical risk assessment approaches that are used as a basis for setting the current regulatory limits, considering current scientific knowledge.

To facilitate progress towards the realisation of the above requirements greater transparency and improved communication in the entire production-consumption- management system is needed. The goal must be a functional economy where wasted resources find their way back into the system with the least trade-offs, especially those relevant to health and safety.

Declaration of Competing Interest

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

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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.2022.128410](https://doi.org/10.1016/j.jhazmat.2022.128410).

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