Hazardous chemicals in recycled and reusable plastic food packaging

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Impact Statement

Society has benefited from plastic food packaging: many foodstuffs have become widely available to humanity throughout the year. However, a downside of plastic food packaging is its environmental persistence when local waste management fails or is not available at all. The increasing plastic pollution is being tackled by different means, one of them being a shift to using more recycled content in plastic articles. Another approach is to ramp up reusable packaging by introducing refillable containers. But both approaches – reusing and recycling plastic food packaging – must address the issue of chemicals that transfer from packaging into food, and that may lead to food safety issues due to the presence of hazardous chemicals that accumulate in plastics throughout their life cycle. In this review article, we zoom in on this issue of chemicals in reusable and/or recyclable plastic food containers, such as packaging and other plastic items that come into contact with food, like kitchen utensils and tableware. We highlight the scientific evidence and key knowledge gaps on chemicals in plastics and how some chemicals of concern found in plastics affect human health.

Abstract

In the battle against plastic pollution many efforts are being undertaken to reduce, reuse, and recycle plastics. If tackled in the right way, these efforts have the potential to contribute to reducing plastic waste and plastic's spread in the environment. However, reusing and recycling plastics can also lead to unintended negative impacts, because hazardous chemicals, like endocrine disrupters and carcinogens, can be released during reuse and accumulate during recycling. In this way, plastic reuse and recycling become vectors for spreading chemicals of concern. This is especially concerning when plastics are reused for food packaging, or when food packaging is made with recycled plastics. Therefore, it is of utmost importance that care is taken to avoid hazardous chemicals in plastic food contact materials, and to ensure that plastic packaging that is reused or made with recycled content is safe for human health and the environment. The data presented in this review are obtained from the Database on Migrating and Extractable Food Contact Chemicals (FCCmigex), which is based on over 700 scientific publications on plastic food contact materials. We provide systematic evidence for migrating and extractable food contact chemicals (FCCs) in plastic polymers that are typically reused, such as polyamide (PA), melamine resin (MelRes), polycarbonate (PC), and polypropylene (PP), or that contain recycled content, such as polyethylene terephthalate (PET). 1332 entries in the FCCmigex database refer to the detection of 509 FCCs in repeat-use food contact materials made of plastic. 853 FCCs are found in recycled PET, of which 57.6% have been detected only once. Here, we compile information on the origin, function, and hazards of FCCs that have been frequently

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detected, such as melamine, 2,4-di-tert-butylphenol, 2,6-di-tert-butylbenzoquinone, caprolactam and PA oligomers, and highlight key knowledge gaps that are relevant for the assessment of chemical safety.

Keywords

Plastic food packaging, hazardous chemicals, plastic recycling, reuse

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Introduction

Plastic materials are highly functional and economically used in today's globalized food systems (Millican & Agarwal, 2021). Plastics make for lightweight food packaging, can be engineered to extend shelf-life significantly (De Hoe et al., 2022), and at the same time, they enable profitable products by allowing for at-scale, high-throughput production and filling, globalized logistics, and retail selling (Matthews et al., 2021). Most food packaging is made of plastics (Poças et al., 2009), and around 20% of global plastics production is used for this purpose (Plastics Europe, 2022). The extensive use of plastic packaging for foodstuffs is also often justified as a means for preventing food waste (Heller et al., 2019). This makes single-use plastic food packaging an enabler of the current, globalized, processed foods system that provides convenience to consumers, making it very difficult to replace (Chakori et al., 2021; Chakori et al., 2022).

But despite its many advantages, the intense and increasing use of plastic food packaging is associated with serious environmental damage (Borrelle et al., 2020; Jambeck et al., 2015; MacLeod et al., 2021; Morales-Caselles et al., 2021; Persson et al., 2022; Wilcox et al., 2015) and has led to increasing calls for amelioration (Borrelle et al., 2020; Geyer et al., 2017; Lau et al., 2020). Therefore, the United Nations Environmental Program has been tasked with preparing a Global Plastics Treaty to "end plastic pollution" and develop "an international legally binding instrument" (UNEP, 2022). The call for reducing plastic pollution from (food) packaging waste has also been heard in several countries across the globe, and novel approaches are being developed that would allow for continued use of plastics materials in food packaging while addressing its end-of-life challenges (Matthews et al., 2021; Prata et al., 2019). This includes designing packaging so that it allows for recycling (De Hoe et al., 2022; Eriksen et al., 2019; Schyns & Shaver, 2021), for example, by using only certain polymer types as mono-materials with additional, specific material properties such as transparency and colorlessness.

However, the focus on plastic packaging recycling is a less favorable option according to the EU's waste hierarchy which sees reduction and reuse as preferable approaches (EEA, 2019). For this reason, there is an increasing push towards reducing overall plastics packaging waste, for example by setting binding national reduction targets and promoting the reuse of food packaging (EC, 2022b; EU 2019/904; Klemeš et al., 2021), even though this requires far bigger changes to food production, logistics, and retail, and is therefore more difficult to implement (Borrelle et al., 2020; Phelan et al., 2022; Wagner, 2022).

In this review, we focus on the important issue of chemicals, as this is an aspect that is often overlooked when solutions to end plastic pollution from food packaging waste are discussed (Dey et

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al., 2022; Wang & Praetorius, 2022). Indeed, plastics are chemically very complex materials, containing hundreds of different, synthetic compounds which are more often than not, poorly characterized for their hazard properties and which in many cases even remain unknown regarding their chemical identities (Crippa et al., 2019). Still, it is well-established that chemicals transfer from plastic food packaging into foodstuffs, and this process of chemical migration has been the focus of over 700 scientific publications (Geueke et al., 2022). At the same time, there is concern about the adverse health impacts of chemical migration when almost the entire population is ingesting plastic-associated chemicals that are often not studied adequately for their health risks (Groh et al., 2021; Landrigan et al., 2023; Muncke et al., 2020; Symeonides et al., 2021).

These concerns about migration of hazardous chemicals and their impacts on human health are especially relevant for plastic food contact materials (FCMs) made from recycled plastics (Cook et al., 2023; Geueke et al., 2018), because unknown and/or hazardous chemicals can accumulate in recycled material and then migrate into foodstuffs, leading to chronic human exposure, as has been shown in the case of beverage bottles made from polyethylene terephthalate (PET) plastic (Gerassimidou et al., 2022; Steimel et al., 2022; Tsochatzis et al., 2022). Illicit plastic recycling, where non-food grade plastics containing hazardous brominated flame retardants are used to make FCMs, is prevalent, as data from the European, US, and Korean markets reveal (Paseiro-Cerrato et al., 2021; Rani et al., 2014; Samsonek & Puype, 2013b; Turner, 2018). Additionally, technical limitations exist with respect to the recyclability of commonly used plastic food packaging into chemically safe recycled food packaging because of the inherent physico-chemical properties of the materials that hamper the efficient removal of chemical contaminants (Palkopoulou et al., 2016). Especially concerning is the use of recovered plastic waste, e.g., from ocean clean ups, for food contact applications, as persistent organic pollutants may be present (Gallo et al., 2018).

In addition, for reused plastic food packaging, there is concern about the migration of hazardous chemicals, for example from consumer (mis-)use of the packaging, or from detergents that can accumulate in the packaging (Tisler & Christensen, 2022). Indeed, food packaging is often soiled with food remains and needs thorough cleaning before reuse, but the plastic polymer may even absorb components of the food or cleaning agents, leading to discoloring and organoleptic changes, or even unwanted chemical contamination of the packaging that may migrate into the food during reuse. Also, non-packaging plastic items for food contact, such as kitchen utensils, tableware, baby bottles, water dispensers, and tubing of milking machines, are often used in repeated contact with food and are a source of chemicals that migrate into foodstuffs. Common plastic polymers used to make these items are polyamide (PA), polypropylene (PP), polycarbonate (PC), melamine resin (MelRes), and

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polyvinylchloride (PVC). At present, little attention is paid to this source of chemical food contamination.

This review provides a systematic overview of food contact chemicals (FCCs) detected in migrates and extracts of recycled plastic FCMs, with a special focus on recycled PET that is typically used in single-use packaging. Additionally, we provide evidence for migrating and extractable FCCs from reusable food contact articles (FCAs) made of plastics, , such as kitchen utensils, plates, cups, and containers. The data are obtained from the Database on Migrating and Extractable Food Contact Chemicals (FCCmigex) (Geueke et al., 2022). Human health implications of exposure to frequently detected FCCs are discussed. This work enables evidence-based decision making regarding the use of plastic food packaging in the circular economy.

Methods

Evidence for presence of FCCs in migrates and extracts

This review is based on the data and references of a systematic evidence map on FCCs measured in migrates and extracts of FCMs (Geueke et al. 2022). The results are accessible via an interactive tool, the FCCmigex dashboard (Food Packaging Forum, 2023). The latest data update considered all relevant and publicly available studies and reports through October 2022. On April 24, 2023, the FCCmigex dashboard included 24,810 database entries and 4266 FCCs. This information was retrieved from 1311 references. The terms FCC, FCM, and FCA were used according to the definitions in Muncke et al. (2017).

To find data on FCCs that were detected in migrates and extracts of recycled plastics, we first filtered the FCCmigex database for data and references on PET and recycled PET, which are listed as distinct FCM types if the relevant references provide this information. We also filtered the full dataset for “food contact material: plastics” and searched the term “recyc” in the titles and abstracts of the resulting references, which were then screened with respect to the recycled content of the investigated plastic FCMs.

For data and references on reusable plastics, we applied the filters “food contact material: plastics” and “food contact article: repeat-use” in the FCCmigex database. Additionally, we filtered for “detection: yes”.

We also searched the FCCmigex database for specific chemicals by using their Chemical Abstracts Service (CAS) Registry Numbers and combined these searches with the FCM of interest. For example, to obtain information about bisphenol A (BPA, CAS Registry Number 80-05-7) that was detected in migrates and extracts of reusable PC, we used the following search term and filters: CAS Registry

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Number: 80-05-7, food contact material: plastic > polycarbonate, food contact article: repeat-use, detection: yes.

Hazards of FCCs

For FCCs that were frequently detected in migrates and extracts of recycled and reusable plastic FCMs, we compiled the hazard properties according to the criteria mentioned in the European Chemicals Strategy for Sustainability (CSS) (EC, 2020). The CSS aims at removing the most harmful chemicals from consumer products, including FCMs. Chemicals that are carcinogenic, mutagenic, or toxic to reproduction (CMR), have specific target organ toxicity (STOT) or endocrine disrupting properties, were defined as “most harmful” by the CSS. Also, chemicals with persistence and bioaccumulation-related hazards (PBT, vPvB) and persistent and mobile chemicals (PMT/vPvT) were included as chemicals of concern in the CSS.

We applied the methodology as described by (Zimmermann et al., 2022) and referred to the following hazard sources: European Chemical Agency’s (ECHA) Classification and Labeling (C&L) inventory that is aligned with the Globally Harmonized System (GHS) for classification and labeling of chemicals (ECHA, 2023f), GHS-aligned classification by the Japanese Government (NITE, 2023), EU Registration, Evaluation, Authorisation, and Restriction of Chemicals (REACH) Substances of Very High Concern (SVHC) list (ECHA, 2023g), California's Office of Environmental Health Hazard Assessment’s (OEHHA) Proposition 65 List (OEHHA, 2023), substances identified as endocrine disruptors at EU level (Endocrine Disruptor List, 2022), PBT/vPvB assessments carried out under the previous EU chemicals legislation (ECHA, 2007), US Environmental Protection Agency's (EPA) list of PBT substances (U.S. EPA, 2023), US EPA’s archived list of Priority Chemicals (U.S. EPA, 2016), ECHA's PBT assessment list (ECHA, 2023a), Stockholm convention (POP) (Stockholm Convention, 2022), ECHA’s list for inclusion in POPs Regulation, ECHA’s list of substances subject to POPs Regulation (ECHA, 2023e), and German Environment Agency (UBA) report (Arp & Hale, 2019). All hazard sources were accessed between January 24-30, 2023.

Based on the GHS for classification and labelling, we defined chemicals with CMR properties that were assigned to categories 1A and 1B (known and presumed CMR, respectively) and chemicals with STOT that were classified as category 1 after repeated exposure as having hazard properties of concern. Chemicals with respiratory system hazards leading to a classification as STOT RE 1 were not included as they were not considered relevant for FCMs, where chemical exposure is oral.

FCCs that were not listed in any of the twelve sources above were labelled as “no data available”. For FCCs that have data in any of these sources, but were not categorized as chemical of concern, we searched for ongoing assessments and notifications in the respective Substance Infocard published

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by ECHA (ECHA, 2023b). We also added references from the peer-reviewed literature regarding potential hazards of concern if no priority hazards were assigned to a chemical according to (Zimmermann et al., 2022) or no ongoing regulatory assessments were reported by (ECHA, 2023b).

Results

Plastic data in the FCCmigex database

In the most recent version of the FCCmigex database, we included 824 scientific studies and reports on plastic FCMs. From these references, 13,958 database entries have been generated, where a database entry corresponds to one experimental finding (Geueke et al., 2022). More specifically, each database entry is linked to the reference from which it was generated and provides information about the FCC, what type of FCA (single or repeat-use) and which FCM(s) were investigated, whether the experimental set-up was a migration or extraction experiment and if the chemical was detected or not. Notably, a reference can contain multiple experimental findings, and therefore result in several database entries. In total, 3009 FCCs were detected in migrates and extracts of plastic FCMs. We integrated data from nine different types of plastic polymers (PA, PC, polyethylene (PE), PET, PP, PVC, MelRes, polyurethane (PU), and polystyrene (PS)). Additionally, plastic FCMs that consist of multilayers and those that were not further specified or made of another polymer, such as Tritan and polylactic acid, form two more categories of plastic FCMs in the database.

Recycled plastic FCMs

Recycled PET

The FCCmigex database contains 1436 FCCs detected in migrates and extracts of PET, represented by 2455 database entries. 22 of 156 references on PET specifically refer to the detection of FCCs in migrates and extracts of recycled PET (Figure 1). This percentage does not necessarily reflect the actual share of recycled content in the investigated samples as in many references no distinction was made between virgin and recycled PET.

Antimony and acetaldehyde are very often detected FCCs in migrates and extracts of PET (Table 1). Ortho-phthalates, such as di-(2-ethylhexyl) phthalate (DEHP), dibutyl phthalate (DBP), diethyl phthalate (DEP), dimethyl phthalate (DMP), and diisobutyl phthalate (DiBP), heavy metals, the monomers ethylene glycol and terephthalic acid, more aldehydes, cyclic PET oligomers, and 2,4-di-tert-butylphenol (2,4-DTBP) are also among the most frequently detected FCCs. On the contrary, 1014 chemicals that have been detected in any PET sample were found only once (corresponding to one database entry). 523 and 491 of these FCCs are found in virgin/unspecified PET and recycled PET, respectively (Figure 1), which is mainly the result of untargeted analyses of migrates and

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extracts (Aznar et al., 2020; Brenz et al., 2021; Jaén et al., 2021; Wu et al., 2022). Such untargeted screenings often lead to the detection of non-intentionally added substances (NIAS), including reaction by-products, contaminants, and degradation products (Table 1).

When focusing on the FCCs that have been detected in migrates and extracts of PET samples with confirmed recycled content, the data are sparse (Table 1). Antimony is most frequently detected, followed by limonene, a common aroma compound, that is considered a marker for recycled content (Fabris et al., 2010; Thoden van Velzen et al., 2020).

The FCCmigex contains data from a reference describing an untargeted analysis of volatile organic compounds (VOCs) where 1247 chemicals have been detected and tentatively identified in 45 virgin and 82 recycled PET samples (Li et al., 2022). In this study, 524 VOCs have been detected only in PET samples with recycled content, versus 461 chemicals that are present only in virgin PET. 262 chemicals are detected in both types of PET. 1139 of these 1247 chemicals reported by Li and colleagues have a CAS RN and are included in the FCCmigex interactive dashboard. 1017 of these 1139 chemicals (or 90%) have not previously been detected in any PET migrate or extract, which illustrates the potential of untargeted studies and also shows the large individual variations of FCAs made of the same polymer. Hydrocarbons and benzenoids are predominant categories for virgin and recycled PET samples, respectively. Slip agents, which are commonly used to control friction during polymer production, have been proposed as possible sources of hydrocarbons in virgin PET, and some of the benzenoids that are highly prevalent in recycled PET could have originated from food additives and degradation products of surfactants. To our knowledge, the results of this study form the most comprehensive, publicly available dataset systematically comparing chemicals in recycled and virgin PET samples.

Other recycled polymers

The FCCmigex contains only a few references on the chemical migration from specific recycled polymers other than PET, such as PS, PP, PE, and Tritan. Typical FCCs reported in these references are volatile organic compounds, including styrene monomer and oligomers from recycled PS (Lin et al., 2017; Song et al., 2019), degradation products of antioxidants from recycled polyolefins (Coulier et al., 2007), and contaminations with bisphenols in recycled Tritan that may be explained by the ubiquitous presence of these substances (Banaderakhshan et al., 2022).

In the decade after 2010, the detection of brominated flame retardants and heavy metals in black plastic FCAs was an unexpected finding and it indicated that plastic waste from electrical and electronic equipment is illegally recycled into FCAs (Guzzonato et al., 2017; Puype et al., 2015; Puype et al., 2019; Samsonek & Puype, 2013a; Turner, 2018).

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Repeat-use plastic FCAs

In the FCCmigex, 1332 database entries from 177 references are related to the detection of 509 FCCs in repeat-use plastics. The polymer types for which the highest percentage of repeat-use articles has been studied are MelRes (95.6% repeat-use), PC (68.6%), PA (59.2%), and PP (17.1%) (Figure 2). Typical FCAs made of MelRes and studied for their chemical migration potential are reusable kitchen utensils and tableware, often especially designed for babies and children. Examples of repeat-use FCAs made of PP, PC, and PA that are included in the FCCmigex database are food containers, baby bottles, and kitchen utensils, respectively.

The most commonly used type of PC contains BPA as monomer. In the last decade, BPA-containing baby bottles have been banned all over the world due to health and safety concerns, leading to the replacement of BPA-based PC by other plastic polymers. PA is widely used in kitchen utensils, such as cooking spoons and spatulas, and other repeat-use FCAs, such as coffee mugs and electric kitchen appliances. Besides, single-use plastic packaging is also commonly made of PA, such as tea bags and multilayer plastic films. Food containers are often made of PP, for both single-use and repeat-use. Further food-contact applications of PP are, e.g., films, bags, and bottle caps.

Across all polymers, PA, PP, PC, and MelRes also have the highest total number of database entries for repeat-use FCAs (Figure 3). For four polymer types in the FCCmigex database (PE, PET, PS, and PVC), between 1.8 and 6.2% of their respective database entries are on repeat use (Figure 2). The FCM categories “multilayer plastics” and PU do not include any information on repeat-use FCAs, whereas 20.4% of the database entries refer to repeat-use in the category “plastics, non-specified or other.”

In migrates and extracts of PA and PP, 120 and 122 different FCCs have been identified, respectively, while 76 different FCCs originate from PC and 45 FCCs from MelRes (Figure 3). On average 4.4 and 3.6 FCCs per reference have been detected for PA and PP, respectively, which contrasts with only 1.7 FCCs per reference for PC and MelRes.

The frequencies of database entries for the most detected FCCs per polymer type are shown in Figure 4. For PC, 32.4% of the database entries are related to the detection of BPA, while the remaining 67.6% cover 75 other FCCs. Melamine and formaldehyde account for 50.6% of all database entries related to MelRes. In contrast, a much higher number of different FCCs has been detected in the migrates and extracts of PA and PP. Primary aromatic amines (PAAs), the monomer of PA6 (caprolactam) and cyclic PA oligomers are most frequently detected in PA. Plastic additives, e.g., Irgafos 168, Irganox 1010, and Irganox 1070, ortho-phthalates, silver, and degradation products

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of antioxidants (2,4-DTBP and 2,6-di-tert-butylbenzoquinone (2,6-DTBQ)) are found with the highest frequencies in migrates and extracts of PP.

Case studies of chemicals of concern

Table 2 summarizes the highly prevalent FCCs and groups of FCCs that have been detected in migrates and extracts of repeat-use FCAs and informs about their function, potential origin, hazards, and their presence on the Union list of authorized substances (EU 10/2011, 2011). Based on these data, we present three case studies to illustrate the implications of chemical migration from repeat-use plastic FCAs. In the following, we will focus in more detail on cyclic oligomers from PA, the degradation products of antioxidants commonly used in PP (2,4-DTBP and 2,6-DTBQ), and melamine from MelRes. All these FCCs are known to be present in plastics after manufacturing or formed during use, and they have the potential to migrate into foods. However, there is very limited information on the toxicity of the cyclic PA oligomers as well as 2,4-DTBP, and 2,6-DTBQ (Table 2, Table 3). The safety of melamine was assessed by the European Food Safety Authority (EFSA) in 2010 (EFSA, 2010), but further research on the human health and environmental hazards of melamine since then has led to its classification as a substance of very high concern and to its assessment as an endocrine disrupting chemical (EDC) and PBT (ECHA, 2023c).

Other FCCs that have been frequently detected in repeat-use plastic FCAs, such as ortho-phthalates, primary aromatic amines, silver, and BPA (Figure 4, Table 2), are not selected here as case studies. However, it is noteworthy that the European Food Safety Authority recently established a tolerable daily intake (TDI) of 0.2 ng BPA per kg body weight per day, which is based on BPA’s immunotoxicity (EFSA, 2023). In comparison with dietary exposure estimates for BPA, this TDI is exceeded by two to three orders of magnitude in all age groups. The human health effects of exposure to ortho-phthalates have also been recently reassessed (EFSA, 2022), and for silver-containing active substances human health risk assessment is under discussion (ECHA, 2021a, 2021b, 2021c; EFSA - ECHA, 2020). For PAAs, strict regulatory measures are already in place (EU 10/2011, 2011) (Table 2).

Case study 1: Cyclic PA oligomers

Caprolactam is a cyclic starting substance used in the synthesis of PA 6, whereas PA 6,6 is made from two linear monomers hexamethyldiamine and adipic acid. Both types of PA have global production volumes >1 million metric tons per year, of which a small proportion is used in the manufacture of repeat-use FCAs, such as kitchen utensils and appliances. Caprolactam and cyclic PA oligomers were reported to be the most abundant group of FCCs in migrates and extracts of repeat-use FCAs made of PA in general (Song et al., 2022). In contrast, the linear starting substances of PA 6,6 were typically not detected (Table 3). Early studies on caprolactam and cyclic PA oligomer migration from repeat-

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use PA FCAs were published in the 2000s (Brede & Skjevrak, 2004; Bustos et al., 2009; Skjevrak et al., 2005), but evidence for their migration has increased especially over the last decade (BfR, 2018, 2019b; Hu et al., 2021; Kappenstein et al., 2018) (Table 3). This development is reflected by improved analytical methods and identification approaches (Song et al., 2022), and the custom synthesis of reference standards for PA oligomers, which are not commercially available yet (Canellas et al., 2021).

None of the detected PA oligomers have been found in any of the sources which we consulted to identify hazard properties of concern. This absence of hazard data has already been discussed when PA oligomers were increasingly found in migrates and extracts of repeat-use FCAs, and a first safety assessment of PA oligomers in 2018 relied on the threshold of toxicological concern (TCC) concept to set specific migration limits (SMLs) of 90 µg/kg food for individual PA oligomers (BfR, 2018; Kappenstein et al., 2018). A year later, a group SML of 5 mg/kg food was proposed for PA 6 and PA 6,6 oligomers based on toxicity studies for 1,8-diazacyclotetradecan-2,7-dione, which is the smallest cyclic product of the PA 6,6 monomers hexamethyldiamine and adipic acid (BfR, 2019b). Nevertheless, oligomer migration from PA has been found to exceed the set values (BfR, 2018, 2019b; Hu et al., 2021).

Case study 2: Degradation products of antioxidants

In PP, antioxidants are needed to prevent oxidation and degradation of the polymer backbone during processing and service life, which would lead to, e.g., discoloration and reduced stability of the plastic product. Sterically hindered phenols (e.g., butylated hydroxytoluene, Irganox 1010, Irganox 1076) and phosphite antioxidants (e.g., Irgafos 168) are commonly used for this purpose (Dopico-García et al., 2007; Dorey et al., 2020). By intention, antioxidants fulfil their purpose by reacting in the polymer and forming new substances, of which 2,4-DTBP and 2,6-DTBQ were most frequently detected in extracts and migrates of repeat-use FCAs made of PP. 2,4-DTBP is a break-down product of Irgafos 168, whereas 2,6-DTBQ is a derivative of sterically hindered phenol antioxidants. Therefore, 2,4-DTBP and 2,6-DTBQ belong to the group of known and predictable NIAS.

2,4-DTBP is regularly detected in the migrates and extracts of baby bottles made of PP that have been used as substitutes for PC (da Silva Oliveira et al., 2017; Oliveira et al., 2020; Onghena et al., 2014; Onghena, Negreira, et al., 2016; Onghena, Van Hoeck, et al., 2016; Simoneau et al., 2012). Most of the database entries related to 2,4-DTBP in the FCCmigex are derived from untargeted studies (Carrero-Carralero et al., 2019; da Silva Oliveira et al., 2017; Onghena et al., 2014). Depending on the sample, migration levels of 10-100 µg/kg food are reported (Onghena et al., 2014). Degradation of Irgafos antioxidants and the formation and migration of 2,4-DTBP increases

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when PP is used at elevated temperatures and in contact with hydrophobic food simulants (Barkby, 1995). In another study, microwave heating shows stronger effects on the migration of 2,4-DTBP than conventional heating (Alin & Hakkarainen, 2011). 2,6-DTBQ is also frequently detected together with 2,4-DTBP, indicating the simultaneous use of sterically hindered phenols and phosphite antioxidants in the same FCAs (Carrero-Carralero et al., 2019; Onghena et al., 2014; Onghena, Van Hoeck, et al., 2016).

In 2019, 2,4-DTBP was measured at 'unexpectedly high' concentrations in human urine and a lack of hazard data has been stated (Liu & Mabury, 2019). In the EU, 2,4-DTBP is currently under assessment as endocrine disrupting chemical (ECHA, 2023d). In contrast, even less data are available for 2,6-DTBQ. For example, the EPA's CompTox Chemicals Dashboard does not list any hazard data, and the GHS-aligned classification results by the Japanese government do not include 2,6-DTBQ at all. However, 2,6-DTBQ recently has been found to have mechanistic evidence that indicates carcinogenic risk (Cui et al., 2022).

Case study 3: Melamine

Melamine belongs to the high-production volume chemicals with an estimated yearly production of almost 2 million metric tons in 2021 (NexanTECA, 2021). Together with formaldehyde, melamine is mainly used in the manufacture of MelRes that is commonly used in reusable tableware and kitchen utensils, often marketed for children. In 2007 and 2008, melamine became a high-profile public issue after several food-related scandals in which baby milk powder (Chan et al., 2008; Schoder, 2010) as well as pet food (Chen et al., 2009; Puschner & Reimschuessel, 2011) were adulterated using melamine. The high nitrogen content of the melamine molecule made it possible to use the industrial chemical as counterfeit for higher protein levels in feed and foods (Figure 5). In China, 50,000 infants were hospitalized because of these criminal food adulterations, and at least six died due to renal failure (Xiu & Klein, 2010).

The migration of melamine and formaldehyde from MelRes tableware has been known since 1986 (Ishiwata et al., 1986; Sugita et al., 1990). Since 2005, melamine has been regularly measured in migrates of tableware and kitchen utensils made of MelRes (Figure 5). Under typical migration conditions (70°C, 3% acetic acid, 2 hours, 3 repetitions), the SML is exceeded in several studies (BfR, 2019a; Mannoni et al., 2017; Osorio et al., 2020). Conditions that increase melamine migration are high temperature, low pH of the food/food simulant, and microwaving (Bradley et al., 2010; Ebner et al., 2020), as well as UV irradiation (Kim et al., 2021).

To simulate repeat-use, three repetitions of the migration tests are recommended because it is generally expected that migration levels decrease during use (EC 10/2011, 2011). For three

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consecutive cycles, there is evidence that the migration of melamine from MelRes follows these expectations (García Ibarra et al., 2016). However, other studies show a reversed trend when the actual use is simulated for more than three cycles, leading to MelRes degradation and increasing the release of its monomers over time (Mannoni et al., 2017; Mattarozzi et al., 2012).

Significant differences in melamine migration have been observed between samples from different suppliers that were tested simultaneously (García Ibarra et al., 2016). These results illustrate the heterogenous quality of MelRes FCAs, which may be caused by varying chemical compositions, impurities of the starting substances, and diverse manufacturing processes.

Additionally, evidence exists that samples have been labelled as MelRes but instead were made of urea-formaldehyde resin, using only a melamine coating on the surface (Poovarodom et al., 2011). Such counterfeit samples show formaldehyde migration exceeding the SML of 15 mg/kg after successive washing cycles (Poovarodom & Tangmongkollert, 2012).

In recent years, tableware made of MelRes and mixed with bio-based powders or fibers, such as bamboo, entered the market and was often labelled as “natural”, “compostable” and “eco-friendly.” However, the materials of natural origin are generally only used as fillers for MelRes, which itself is fossil-carbon based and not biodegradable. Therefore, such labelling is misleading and contains false claims. Even more, bio-based fillers decrease the materials’ stability, promote the migration of melamine and formaldehyde, and lead to the exceedance of SMLs for these FCCs (BfR, 2019a; Osorio et al., 2020). Consequently, the European Commission states that the use of bamboo and other plant-based fillers in plastic FCMs is not authorized according to Regulation (EU) 10/2011. Between May 2021 and April 2022, a European enforcement action plan on plastic FCMs resulted in 748 cases of plastic FCMs containing ground bamboo as filler that were destroyed, recalled, or taken off the market (EC, 2022a).

In 2011, the European Commission (EC) lowered the SML of melamine by a factor of 12 to 2.5 mg/kg food (Commission Regulation (EU) No 1282/2011), which is based on a tolerable daily intake (TDI) of 0.2 mg per kg body weight per day that was derived from the development of urinary bladder stones (EFSA, 2010; WHO, 2009). The EC also detailed the import conditions of kitchenware made of MelRes under Commission Regulation (EU) No 284/2011. In 2017, the FDA issued a recommendation on the use of melamine tableware (U.S. FDA, 2017), and two years later, the German Federal Institute for Risk Assessment (BfR) published a warning on melamine-type tableware (BfR, 2019a).

Besides being a renal toxicant (NITE, 2023; WHO, 2009), melamine is recognized as vPvM/PMT chemical (Arp & Hale, 2019; ChemSec, 2019; ECHA, 2023c). It is currently under assessment as an

EDC and PBT chemical (ECHA, 2023c). Melamine is suspected of damaging the fertility of the unborn child (ECHA, 2023c) and is possibly carcinogenic to humans (IARC, 2019). It may be metabolized to cyanuric acid by the gut microbiome, which supports kidney stone formation (Zheng et al., 2013). In a scoping review, Bolden et al. (2017) map evidence for neurotoxic properties of melamine and identify toxicological endpoints that are not well-studied, including immune, mutagenic/DNA damage, and hematological endpoints.

Discussion

Plastic is the most widely used packaging material for foods and beverages around the world. It generally turns into waste after being used a single time, leading to visible and invisible environmental problems, such as marine pollution by packaging items, microplastics, and chemicals (Gallo et al., 2018; Morales-Caselles et al., 2021). Recycling and reuse of materials have been proposed as measures to reduce the impact of plastic packaging on the environment (Lau et al., 2020). The information on chemical migration that is available in the FCCmigex database and summarized in this review shows that recycling and reuse of plastic FCAs implies that human exposure to hazardous chemicals increases if this aspect is not carefully managed.

Recycled PET has been widely used in food contact applications for over 20 years. Especially the use of recycled beverage bottles has increased due to the establishment of bottle-to-bottle recycling processes, for which decontamination processes have been developed to reduce chemical contamination (Welle, 2011). However, there is experimental evidence that recycled PET contains chemical contaminants that are introduced during use, waste handling, and recycling and that can migrate into the packaged beverages. Associations have been found between the presence of recycled content and the migration of, e.g., benzene and styrene (two carcinogenic chemicals) as well as the endocrine disrupting chemical BPA (Dreolin et al., 2019; Thoden van Velzen et al., 2020). Based on a systematic evidence map on chemical migration from PET bottles into beverages, other authors conclude that research comparing the chemical migration from virgin and recycled PET bottles is relatively sparse (Gerassimidou et al., 2022). This observation is based on the often-unknown level of recycled PET content in beverage bottles.

Recent research aims at developing methods using untargeted screening of PET samples and machine learning algorithms to effectively discriminate between virgin and recycled content. Chemometric methods have tentatively identified hundreds of VOCs that are associated with plastic, food, and cosmetics and reveal significant differences among virgin and recycled PET as well as geographical regions where the recycled material was collected (Dong et al., 2023; Li et al., 2022; Peñalver et al., 2022). Such innovative studies provide highly valuable data on the chemicals that are

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present in recycled PET and other polymers (Su et al., 2021). However, whether this methodology can be used to reliably identify the recycled content in plastic food packaging on a routine basis remains to be seen. Even more, the question of how to assess the safety of the high number of chemicals found not only in recycled plastic polymers, but also in virgin plastics, needs to be urgently addressed.

Compared to recycled PET, even less information is available on the chemical migration from other mechanically recycled polymers. However, within the last five years, the US FDA issued an increasing number of favorable opinions on the suitability of recycling processes for producing FCAs made of polyolefins (U.S. FDA, 2023). These numbers may be a good indicator for the actual use of recycled polyolefins as FCMs. In the EU, it is expected that, besides PET, other types of recycled plastic polymers will be available on the market, as the new Commission Regulation EU 2022/2016 on recycled FCMs and FCAs provides the legal framework for such developments (EC, 2022c; EU 2022/1616, 2022). For example, in 2021, the first request for a safety evaluation of recycled PS was submitted to EFSA (OpenEFSA, 2021).

In addition to the evidence for chemical migration from FCMs with recycled content that is presented in this review, research exists on the chemical migration from recycled plastic polymers that are not used in direct contact with food yet but may be considered as FCMs in the future. However, these references were not included in the FCCmigex, because we focused on FCAs that were already on the market (instead of experimental materials under development), and on polymer samples intended for the manufacture of FCMs. For example, research as well as official assessments investigating the chemical safety of recycled polyolefins, which are not broadly approved as FCMs yet, show that chemical contamination and insufficient cleaning technologies limit the application in direct contact with food (EFSA, 2015, 2016; Horodytska et al., 2020; Palkopoulou et al., 2016; Su et al., 2021; Zeng et al., 2023). In this context, it is of concern that the new EU regulation on recycled plastic FCMs provides limited exemptions to allow FCMs produced with novel recycling technologies to be marketed until sufficient evidence has been gathered to decide on the suitability of the technology (EU 2022/1616, 2022).

FCCs that have been detected in migrates and extracts of PA, PP, PC, and MelRes can be categorized into starting substances, i.e., monomers and plastic additives, and NIAS, e.g., reaction by-products, contaminants, and degradation products (Table 2). Overall, these data indicate that especially some of the NIAS, such as the PA oligomers and degradation products of antioxidants, are still neglected by many regulators as they are only present in the final FCA or formed during use. Although there is evidence of the migration potential, toxicological data and risk assessment lag behind this

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knowledge. A solution could be to broaden the focus from testing the starting substances to also assessing the safety of the final FCA (after manufacture and over the life cycle of the FCA).

For PC and MelRes, most evidence is related to monomers that are detected in migrates and extracts. One reason for the frequent detection of BPA, melamine, and formaldehyde may be the focus of researchers on these well-known and hazardous migrants for which analytical methods and standards are available, but this knowledge-bias may result in other, equally relevant FCCs being overlooked. Alternatively, the abundance of these three FCCs may also be a strong indication for the instability of their respective polymer backbones, leading to migration of monomers that are released as a consequence of polymer degradation processes occurring during reuse and related cleaning. The literature is not clear on this, but there is evidence that PC and MelRes are degraded over repeated use cycles, and migration levels of these monomers increase when tested more than three times (Brede et al., 2003; Mannoni et al., 2017; Mattarozzi et al., 2012; Nam et al., 2010). Similarly, oligomers are also formed during manufacture or released during use of PC (Cavazza et al., 2021). Also for PA, there is clear evidence that cyclic oligomers are common manufacturing by-products (Jenke et al., 2005). Although decreasing concentrations of cyclic PA oligomers were reported after three subsequent migration tests (Kappenstein et al., 2018), it remains open whether degradation reactions will increase these levels over longer periods of use. Such cases are not reflected in the current regulation on plastic FCMs, where only three repetitions of the migration tests are required (EU 10/2011, 2011). Moreover, the recommended test conditions for repeat-use FCAs do not reflect realistic use conditions, such as dishwashing, that can, for example, lead to the adsorption of hundreds of dishwasher-related chemicals to the plastic material (Tisler & Christensen, 2022). Therefore, it would be highly desirable to revise the recommendations and regulatory requirements for repeat-use plastic FCAs to be able to monitor the stability of the polymers over time as well as the uptake of chemicals under more realistic use conditions.

The degradation of antioxidants in PP and other polyolefins is an expected and well-studied process (Dorey et al., 2020; Haider & Karlsson, 2002). However, typical degradation products, such as 2,4-DTBP and 2,6-DTBQ, have rarely been targeted in migration studies. Indeed, many of the results for these chemicals included in the FCCmigex are from untargeted screenings (Hu et al., 2021; Li et al., 2022; Skjevrak et al., 2005). Already in 2014 it was stated that these anticipated degradation products were not addressed in the European FCM regulation (Onghena et al., 2014), and since then the situation has not changed. This is especially concerning since 2,4-DTBP is under assessment as an EDC, and for 2,6-DTBQ limited hazard data indicate potential concern for carcinogenicity (Table 2). At the same time, these NIAS can be assumed to be present ubiquitously in PP packaging, leading to

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significant human exposure (Liu & Mabury, 2019). Therefore, hazard data for these substances are urgently needed to fill data gaps.

In this review, we showed that chemical migration from recycled and repeat-use FCAs is of concern, because FCCs with priority hazard properties are present in all investigated materials. What is more, for other frequently detected FCCs no or only limited hazard data exist, like PA oligomers and 2,6-DTBQ. Plastic recycling can introduce unknown or known hazardous chemicals originating from all stages of the life cycle as well as from illicit sources into food packaging and other plastic FCAs. Further concern stems from the observation that it is very difficult to discriminate virgin and recycled materials. Additionally, there is evidence for a potential increase in migration rates after prolonged use of reusable plastic FCAs, which should be better tested in the future.

Many of the data presented here have been acquired in targeted analytical studies. However, there is currently a shift towards untargeted screening studies, which are more suited to represent the chemical complexity of a migrate or extract. While the growing body of evidence in this area is highly appreciated, the question arises how this information can be used to increase the safety of plastic FCMs, because many of the chemicals detected in such screenings do not have any hazard data and cannot be tested one by one. In the future, one solution could be the routine implementation of bioassays to test the safety of migrates and extracts (Groh & Muncke, 2017; Muncke et al., 2023). Alternatively, a shift towards materials that can be safely reused due to their favorable, inert material properties could be a promising option to reduce the impacts of single-use food packaging on the environment and of migrating chemicals on human health. There is an urgent need for establishing suitable analytical methods with low limits of detection to assess the inertness of FCMs, and for including such considerations in FCM and packaging regulations all over the world.

Based on these data, we know that many hazardous chemicals have been found in migrates and extracts of plastic FCMs, and we have evidence for a potential increase in migration rates after prolonged use of some repeat-use plastic FCAs. Importantly, the introduction of unknown and known hazardous chemicals during plastics recycling is of concern, and we caution stakeholders on this matter.

Author Contribution statement

This overview review was conceptualized by BG and JM. Literature screening and data extraction was performed by BG and DP. Data were processed by LP. The original draft manuscript was written by BG and JM. All authors provided review and constructive feedback and approved the final version.

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Conflict of Interest statement

The authors have no conflict of interest to report. BG, LP and JM are employees of the Food Packaging Forum Foundation (FPF), and DP was paid as consultant by the FPF for this work. The authors were not restricted in any way to plan and execute this work.

Data Availability statement

The most recent update of the FCCmigex database (version 2, release date: April 11, 2023) is publicly available as an interactive dashboard using Microsoft PowerBI under the following open access link (https://www.foodpackagingforum.org/fccmigex).

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Table 1. Overview of FCCs that were most frequently detected in migrates and/or extracts of FCMs made of PET (source: FCCmigex), their function and potential origin, hazard properties of concern, and presence on presence on the Union list of authorized substances (EU 10/2011).

FCC	CAS RN	FCCmigex		Function and potential origin in PET	Food contact chemical of concern, according to Zimmermann et al. (2022)	Other/not yet confirmed hazard properties of concern ECHA (2023b)	Primary literature indicates potential concern for*	Presence on the Union list; SML [mg/kg food or food simulant]
		No. of database entries (all PET/ only recycled PET)	No. of references (all PET/ only recycled PET)					
Antimony	7440-36-0	58/11	34/9	Catalyst	No priority hazards reported	A majority of data submitters agree this substance is toxic to reproduction	-	Yes; 0.04
Di-(2-ethylhexyl) phthalate (DEHP)	117-81-7	42/2	31/2	NIAS	CMR EDC	No	-	Yes[1]; 1.5
Dibutyl phthalate (DBP)	84-74-2	33/3	23/3	NIAS	CMR EDC	Under assessment as PBT	-	Yes[2]; 0.3
Acetaldehyde	75-07-0	29/3	18/2	NIAS (degradation product)	CMR	No	-	Yes; 6
Diethyl phthalate (DEP)	84-66-2	21/2	18/2	NIAS	No priority hazards reported	Under assessment as EDC	-	No
Dimethyl phthalate (DMP)	131-11-3	13/2	10/2	NIAS	No priority hazards reported	No	Immunotoxicity (Chi et al., 2022); EDC (Mei et al., 2019)	No
Decanal	112-31-2	13/2	9/2	NIAS	No priority hazards reported	No	-	No
PET cyclic trimer, 1st series	7441-32-9	13/1	10/1	NIAS (reaction by-product)	No data available	No data available	No data available	No

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Nonanal	124-19-6	12/2	8/2	NIAS	No priority hazards reported	No	No data available	No
Ethylene glycol	107-21-1	12/1	9/1	Monomer	CMR	No	-	Yes; 30 (group SML)
Cobalt	7440-48-4	12/1	8/1	NIAS (contamination)	CMR STOT	No	-	Yes; 0.05
Limonene isomers	138-86-3, 5989-27-5	11/5	8/4	NIAS (recycling-related contamination)	No priority hazards reported	Very toxic to aquatic life	-	No
Lead	7439-92-1	11/3	9/3	NIAS (contamination)	CMR STOT	No	-	No; ND
2,4-di-tert-butylphenol (2,4-DTBP)	96-76-4	11/1	9/1	NIAS (degradation product of antioxidants)	No priority hazards reported	Under assessment as EDC	-	No
Bisphenol A (BPA)	80-05-7	11/2	8/1	NIAS	CMR EDC	No	-	Yes[3]; 0.05
PET cyclic dimer, 2nd series	29278-57-7	11/2	8/1	NIAS (reaction by-product)	No data available	No data available	No data available	No
Terephthalic acid	100-21-0	10/0	9/0	Monomer	No priority hazards reported	No	Obesogenic properties (Molonia et al., 2022)	Yes; 7.5 (group SML)
PET cyclic trimer, 2nd series	873422-64-1	10/1	7/1	NIAS (reaction by-product)	No data available	No data available	No data available	No
PET cyclic dimer, 3rd series	16104-98-6	10/1	4/1	NIAS (reaction by-product)	No data available	No data available	No data available	No
Diisobutyl phthalate (DiBP)	84-69-5	9/2	8/2	NIAS	CMR EDC	Some data submitters indicate they consider this substance as PBT	-	No
Cadmium	7440-43-9	9/3	7/3	NIAS (contamination)	CMR STOT PBT/vPvB	No	-	No; ND (LOD 0.002)

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2-Methyl-1,3-dioxolane	497-26-7	9/3	5/2	NIAS (reaction by-product)	No priority hazards reported	No	No data available	No

Abbreviations: SML – specific migration limit, NIAS – non-intentionally added substance; CMR – carcinogenic, mutagenic or toxic to reproduction, STOT – specific target organ toxicity, EDC – endocrine disrupting chemical, PBT – persistent, bioaccumulative and toxic, vPvB – very persistent, very bioaccumulative, vPvM – very persistent, very mobile, ND – the substance shall not migrate in detectable quantities, LOD – level of detection.

*Primary literature was only consulted when no priority hazards were assigned according to Zimmermann et al. (2022) or no ongoing assessments were reported by ECHA (2023b).

[1]Only to be used as: (a) plasticizer in repeated use materials and articles contacting non-fatty foods; (b) technical support agent in polyolefins in concentrations up to 0,1 % in the final product. [2]Only to be used as: (a) plasticizer in repeated use materials and articles contacting non-fatty foods; (b) technical support agent in polyolefins in concentrations up to 0,05 % in the final product. [3]Not to be used for the manufacture of PC infant feeding bottles and PC drinking cups or bottles which, due to their spill proof characteristics, are intended for infants and young children.

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Table 2. Overview of FCCs that were most frequently detected in migrates and/or extract of repeat-use plastic FCAs (source: FCCmigex), their function and potential origin, hazard properties of concern and presence on the Union list of authorized substances (EU 10/2011).

Polymer type	FCC	CAS RN	FCCmigex		Function and potential origin in PET	Food contact chemical of concern, according to Zimmermann et al. (2022)	Other/not yet confirmed hazard properties of concern ECHA (2023b)	Primary literature indicates potential concern for*	Presence on the Union list; SML [mg/kg food or food simulant]
			No. of database entries	No. of references					
PA	4,4′-methylene-dianiline	101-77-9	11	11	NIAS (potential contamination from azodyes)	CMR STOT	No	-	No[1]; ND (LOD 0.002)
	Aniline	62-53-3	12	12	NIAS (potential contamination from azodyes)	CMR STOT	No	-	No[1]; ND (group SML 0.01)
	PA cyclic oligomers	see Table 3	91	8	Reaction by-products	No data available	No data available	No data available	No
	Caprolactam	105-60-2	7	5	Monomer	No priority hazards reported	No	High aquatic mobility and concern for toxicity (Montes et al., 2022)	Yes; 15
PP	2,4-DTBP	96-76-4	13	10	NIAS (degradation product of phosphite antioxidants)	No priority hazards reported	Under assessment as EDC	-	No
	2,6-di-tert-butylbenzoquinone (2,6-DTBQ)	719-22-2	9	6	NIAS (degradation product of sterically hindered phenol antioxidants)	No priority hazards reported	No	Carcinogenicity (Cui et al., 2022)	No
	Silver	7440-22-4	12	5	Active substance	No priority hazards reported	Under assessment as EDC;	-	No

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							some data submitters indicate they consider this substance as toxic to reproduction		
	DBP	84-74-2	9	8	Technical support agent	CMR EDC	Under assessment as PBT	-	Yes[2]; 0.3
	DiBP	84-69-5	5	5	NIAS	CMR EDC	Some data submitters indicate they consider this substance as PBT	-	No
	BPA	80-05-7	5	5	NIAS	CMR EDC	No	-	Yes[3]; 0.05
	Irgafos 168	31570-04-4	8	7	Plastic additive	No priority hazards reported	Under assessment as PBT	-	Yes; no SML
	Irganox 1010	6683-19-8	6	4	Plastic additive	No priority hazards reported	No	No data available	Yes; no SML
	Irganox 1076	2082-79-3	4	4	Plastic additive	No priority hazards reported	No	No data available	Yes; 6
PC	BPA	80-05-7	46	38	Monomer	CMR EDC	No	-	Yes[3]; 0.05
MelRes	Melamine	108-78-1	26	23	Monomer	STOT PMT, vPvM	Under assessment as PBT and EDC	-	Yes; 2.5
	Formaldehyde	50-00-0	18	17	Monomer	CMR	No	-	Yes; 15 (group SML)

Abbreviations: SML – specific migration limit, PA – polyamide, PP – polypropylene, PC – polycarbonate, PAA – primary aromatic amine, NIAS – non-intentionally added substance; CMR – carcinogenic, mutagenic or toxic to reproduction, STOT – specific target organ toxicity, EDC – endocrine disrupting chemical, PBT – persistent, bioaccumulative and toxic, vPvM – very persistent, very mobile, ND – the substance shall not migrate in detectable quantities, LOD – level of detection.

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*Primary literature was only consulted when no priority hazards were assigned according to (Zimmermann et al., 2022) or no ongoing assessments were reported by (ECHA, 2023b).

[1]"ND" if primary aromatic amine on REACH Annex XVII (detection limit 0.02 mg/kg); if not listed: 0.01 mg/kg (group SML). [2]Only to be used as: (a) plasticizer in repeated use materials and articles contacting non-fatty foods; (b) technical support agent in polyolefins in concentrations up to 0.05 % in the final product. [3]Not to be used for the manufacture of PC infant feeding bottles and PC drinking cups or bottles which, due to their spill proof characteristics, are intended for infants and young children.

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Table 3. Polyamide (PA) monomers and cyclic oligomers in extracts and migrates of repeat-use FCAs made of PA. Cyclic oligomers are reaction by-products formed during the manufacture of PA 6 and PA 6,6.

FCC		CAS RN	FCCmigex		Presence on the Union list; SML [mg/kg food or food simulant]
			No. of database entries	No. of references	
PA 6 cyclic monomer	Caprolactam	105-60-2	7	5	Yes; 15
PA 6 cyclic dimer	1,8-diazacyclotetradecane-2,9-dione	56403-09-9	9	5	No
PA 6 cyclic trimer	1,8,15-triazacycloheneicosane-2,9,16-trione	56403-08-8	11	7	No
PA 6 cyclic tetramer	1,8,15,22-tetraazacyclooctacosane-2,9,16,23-tetrone	5834-63-9	10	6	No
PA 6 cyclic pentamer	1,8,15,22,29-pentaazacyclopentatriacontane-2,9,16,23,30-pentone	864-90-4	10	6	No
PA 6 cyclic hexamer	1,8,15,22,29,36-hexaazacyclodotetracontane-2,9,16,23,30,37-hexone	865-14-5	10	7	No
PA 6 cyclic heptamer	1,8,15,22,29,36,43-heptaazacyclononatetracontane-2,9,16,23,30,37,44-heptone	16056-00-1	4	3	No
PA 6 cyclic octamer	1,8,15,22,29,36,43,50-octaazacyclohexapentacontane-2,9,16,23,30,37,44,51-octone	16093-69-9	2	2	No
PA 6 cyclic nonamer	1,8,15,22,29,36,43,50,57-nonaazacyclotrihexacontane-2,9,16,23,30,37,44,51,58-nonone	50694-79-6	1	1	No
PA 6,6 linear monomer	Hexamethyldiamine	124-09-4	0	0	Yes; 2.4
PA 6,6 linear monomer	Adipic acid	124-04-9	0	0	Yes; no SML
PA 6,6 'cyclic monomer'	1,8-diazacyclotetradecane-2,7-dione	4266-66-4	12	8	No

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PA 6,6 cyclic dimer	1,8,15,22-tetraazacyclooctacosane-2,7,16,21-tetrone	4238-35-1	11	7	No
PA 6,6 cyclic trimer	1,8,15,22,29,36-hexaazacyclodotetracontane-2,7,16,21,30,35-hexone	4174-07-6	10	7	No
PA 6,6 cyclic tetramer	1,8,15,22,29,36,43,50-octaazacyclohexapentacontane-2,7,16,21,30,35,44,49-octone	4266-65-3	1	1	No

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Figure captions

Figure 1. Aggregated numbers from the FCCmigex database on FCMs made of recycled and virgin/unspecified PET. Numbers of references, FCCs, and FCCmigex database entries are shown in blue, yellow, and green, respectively. FCCs that were detected only once in any of the PET samples are shown in light yellow. Filter applied in the FCCmigex: Detection – yes.

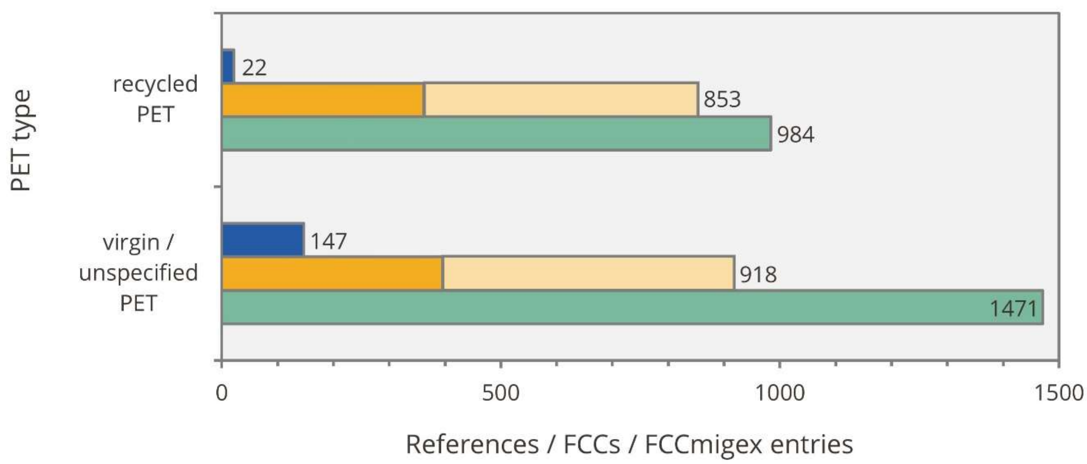

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Figure 2. Number of FCCmigex database entries for eleven categories of plastic FCMs. The plastic FCMs are divided into nine different polymers (PE, PP, PET, PS, PVC, PA, PC, MelRes, and PU) and two other categories ("multilayer plastics" and "plastics, non-specified and others"). Each bar displays the number of database entries for single-use FCAs (blue), repeat-use FCAs (yellow), and FCAs that were not specified (green). The data labels show the percentage of repeat-use FCAs for each category. Filter applied in the FCCmigex: Detection – yes.

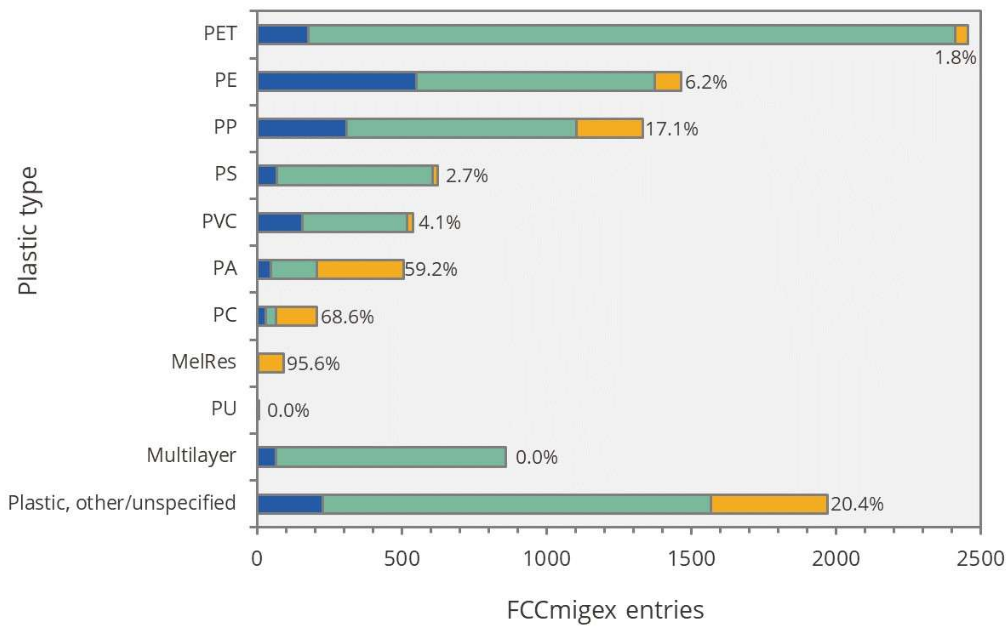

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Figure 3. Aggregated numbers from the FCCmigex database on repeat-use plastic FCAs by polymer type (polyamide (PA); polypropylene (PP); polycarbonate (PC); melamine resin (MelRes), plastic, other/non-specified). Numbers of references, FCCs, and FCCmigex database entries are shown in blue, yellow, and green, respectively. Filters applied in the FCCmigex: Detection – yes, FCA – repeat-use. For example, for PA, the FCCmigex contains 27 references with 120 FCCs detected and results from 277 experimental findings.

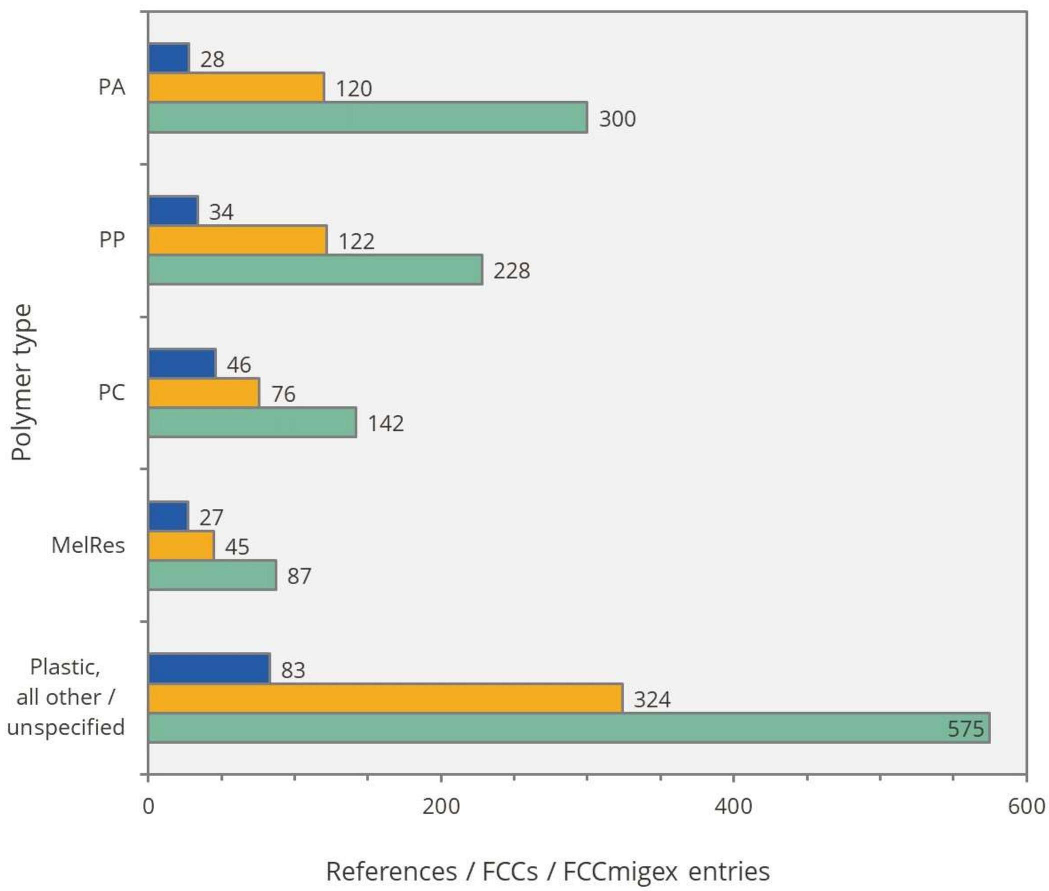

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Figure 4. Relative frequency of FCCmigex database entries per FCC for four repeat-use plastic FCAs by polymer type (polyamide (PA); polypropylene (PP); polycarbonate (PC); melamine resin (MelRes)). Function and potential origin of the most frequently detected FCCs were coded by colors: red – restricted substances, yellow – reaction by-products, blue – monomers, green – authorized plastic additives, light green – degradation products of antioxidants (NIAS), gray – not authorized for plastic FCMs in the EU. Filters applied in the FCCmigex database: Detection – yes, FCA – repeat-use.

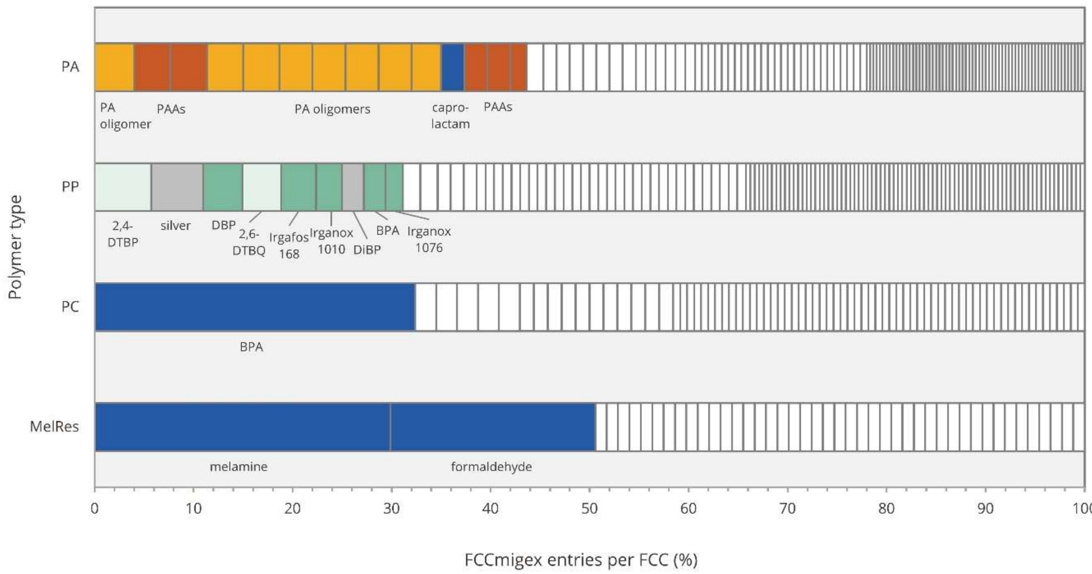

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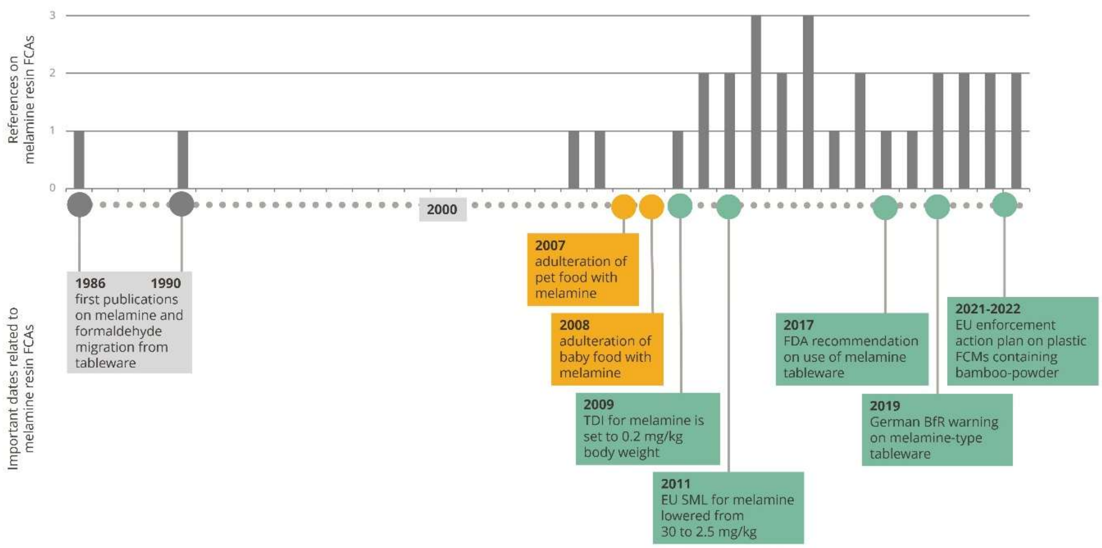
Figure 5. Evidence for chemical migration from melamine resin FCAs into foods and food simulants represented by number of publications by year and important dates related to melamine and food safety.



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