



Review

Food packaging in the circular economy: Overview of chemical safety aspects for commonly used materials



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ABSTRACT

Food packaging facilitates storage, handling, transport, and preservation of food and is essential for preventing food waste. Besides these beneficial properties, food packaging causes rising concern for the environment due to its high production volume, often short usage time, and problems related to waste management and littering. Reduction, reuse, and recycling, but also redesign support the aims of the circular economy. These tools also have the potential to decrease the environmental impact of food packaging.

In this article, we focus on chemical safety aspects of recycled food packaging, as recycling is currently seen as an important measure to manage packaging waste. However, recycling may increase the levels of potentially hazardous chemicals in the packaging and -after migration- in the food. Since exposure to certain chemicals migrating from food packaging has been associated with chronic diseases, it is of high importance to assess the safety of recycled packaging. Therefore, we describe recycling processes of commonly used food packaging materials, including plastics, paper and board, aluminum, steel, and multimaterial multilayers (e.g., beverage cartons). Further, we give an overview of typical migrants from all types of recycled food packaging materials, and summarize approaches to reduce chemical contamination. We discuss the role of food packaging in the circular economy, where recycling is only one of many complementary tools for providing environmentally-friendly and safe food packaging.

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1. Introduction

Based on the observation that today's take-make-dispose economy is one driver of global environmental change and ecosystems deterioration, the concept of a circular economy has been developed as a tool to further prevent and reduce those detrimental human activities (Geissdoerfer et al., 2017; Ghisellini et al., 2016; Kirchherr et al., 2017; Loiseau et al., 2016; Winans et al., 2017). The circular economy promotes closing loops in industrial systems, minimizing waste, and reducing raw material and energy inputs (European Environment Agency, 2016; Stahel, 2016). Over the past years, the concept has gained importance in policy making (EC, 2017; The Standing Committee of the National People's Congress China, 2008) and has been increasingly implemented in production, consumption and waste sectors all over the world (Ghisellini et al., 2016). Practical solutions aiming at a circular economy include eco-design, waste prevention programs, and extending the lifetime of products (European Environment Agency, 2016). "Reduce, reuse and recycle" are three important waste management options. The reduction principle targets the minimization of raw material use, energy input, and waste production whereas the reuse principle refers to the repeated use of products or components for their intended purpose (Ghisellini et al., 2016). Recycling is mostly used to save energy, resources, and emissions and decrease the environmental impacts of a material's use. Although reduction and reuse are "greener" options (Allwood, 2014), the use of recycled instead of virgin material is generally also perceived as a beneficial solution (Grosso et al., 2017). In the context of packaging waste, reuse and recycling are political means to initiate a change, which is expected to deliver both economic and environmental benefits (EC, 2015). However, only some types of material can be repeatedly recycled and/or reused (e.g., metals, glass). Accordingly, a material meeting these requirements was defined as:

"[...] *permanent* if its inherent properties do not change during use and through solid-liquid transformation, it can revert to its initial state. This is the case when the material consists of basic components, which are either chemical elements or robust chemical compounds, making repeated use and recycling possible without change of inherent material properties" (Conte et al., 2014).

In contrast to permanent materials, non-permanent materials either undergo gradual chemical changes during the use-phase and/or recycling leading to limited recyclability (e.g., thermoplastics, paper and board), or they are hardly recyclable (e.g., thermoset plastics). Nevertheless, non-permanent materials may be beneficially used in the circular economy, e.g., when using renewable material resources (Conte et al., 2014; Grosso et al., 2017).

In 2014, packaging waste generated in European households and all sectors of the producing industries was estimated at 82.5 million metric tons per year (Eurostat, 2017), and 69.6 million metric tons of packaging waste was collected as part of the municipal solid waste in the US (US EPA, 2016). 65.5% and 51.5% of the packaging waste was recycled in the EU and the US, respectively, but the share of food packaging in these waste streams has not been reported (Eurostat, 2017; US EPA, 2016). However, the markets for food and beverage packaging are large, with an estimated market share of 69% of all consumer packaging used in both categories in total (Rexam, 2012).

Recycling of food packaging waste into new food packaging presents particular challenges, especially with regard to safety issues. The use of recycled food packaging not only increases the possible sources of contamination, but often also the numbers and levels of chemicals that can migrate from the packaging into foods,

thereby potentially affecting human health (BMELV, 2012; Muncke et al., 2017; Pivnenko et al., 2016a; Vápenka et al., 2016). It is especially important to monitor recycled materials for the presence of non-intentionally added substances (NIAS), including (often unknown) impurities, reaction and break-down products. NIAS can reach higher levels in recycled food packaging, because (i) materials intended for recycling may contain intrinsic contaminants such as dyes, additives and their degradation products, (ii) the material may be degraded during use and/or recycling, (iii) chemicals may accumulate when materials are recycled several times, (iv) previous use and/or misuse of the packaging may contribute to the presence of unwanted and/or unexpected contaminants, and (v) non-food grade materials may enter the recycling stream.

However, regulation for food packaging in Europe and the US requires the same level of safety for chemicals migrating into foods for all recycled and virgin materials alike (21 CFR 170.3(i); EC 1935/2004; Muncke et al., 2017; EC 282/2008). In Europe, the use of recycled plastics in FCMs is specifically regulated under the Plastics Recycling Regulation (EC 282/2008). The Food and Drug Administration of the United States (US FDA) considers recycling processes for plastic food contact articles on a case-by-case basis and invites recyclers of plastic to submit information on their process for evaluation and comment (US FDA, 2006; 21 CFR 176.260). In the US, the use of recycled paper and board is regulated under 21 CFR 176.260. Accordingly, waste paper shall not contain any poisonous or deleterious substance that is retained in the recovered pulp and migrates into food, except those specifically regulated under 21 USC 346 and 21 USC 348. In Europe, no harmonized regulation exists for (recycled) paper and board FCMs while some EU Member States have introduced specific measures (Simoneau et al., 2016). Switzerland has banned the use of recycled paper and board in direct contact with food (FDHA, 2016).

With this article, we provide an overview of the most important properties of food packaging materials affecting their recyclability, as recycling is fundamentally relevant to achieving a circular economy. In addition, we review material-specific chemical contamination, impacting on the safe use of recycled FCMs. Recycling practices are exemplified for the different materials, along with decontamination options for removing chemicals of concern. Other aspects, such as energy use during packaging manufacture, renewable resources, energy recovery from waste, and composting of biodegradable food packaging are also important for selecting environmentally friendly packaging materials, but these are outside the scope of this article. Finally, criteria for successful reduction, reuse, and recycling are discussed with regard to permanent and non-permanent packaging materials.

2. Methods

We selected food packaging materials based on their estimated market shares (Poças et al., 2009): Plastic, glass, metal, paper and board, and multimaterial-multilayer were identified as most common types of food packaging materials.

We then searched Web of Science and Pubmed for keywords and combinations thereof. Material type keywords were "aluminum", "steel", "tinplate", "glass", "container glass", "plastics", "plastic", "polyethylene terephthalate", "polystyrene", "polyethylene", "polypropylene", "polyolefin", "paper", "paper and board", "beverage carton", and "tetrapak". Other keywords were "food", "food packaging", "food contact material", "decontamination", "chemical contaminant", "recycling", "reuse", "circular economy", "migration", and "leaching". Further, we also searched for "food packaging recycling". We additionally checked selected articles' references and chose appropriate studies. We searched the internet using a search engine for regulations, market share and

information on recycling processes. We accessed websites of the US FDA, the European Commission (EC) and European Food Safety Authority (EFSA) for information on food packaging recycling.

3. Food packaging materials: properties, recycling, and contaminants

In the following sections, we provide information on common food packaging materials and summarize their material properties affecting recyclability (Table 1). Further, we focus on current recycling technologies and give examples of typical contaminants and their sources.

3.1. Plastics

Plastics are highly diverse, and the most common materials used for packaging foodstuffs. Examples of plastic food packaging include bottles, foils, trays, bags, pots, cups, pouches, bowls, and many more. In 2015, almost 20 million metric tons of plastic packaging were used in Europe (Plastics Europe, 2016), generating approximately 30 kg of plastic packaging waste per inhabitant per year (Eurostat, 2017). Plastic waste can be recovered either by recycling, downcycling, or producing chemical building blocks, fuel or energy (Ignatyev et al., 2014). Recycling is most commonly achieved by mechanical and, to a much lower extent, by chemical processes. Mechanical recycling of plastics includes cleaning, grinding, remelting and regranulating steps. Chemical recycling processes depolymerize plastics into monomers which are subsequently used for repolymerization of virgin-like materials (Geyer et al., 2016). Economic and ecological issues currently limit the large-scale application of chemical recycling processes (Geyer et al., 2016; Ragaert et al., 2017); therefore, here we focus only on mechanical recycling processes for plastic FCMs, with a special emphasis on the presence and migration of contaminants.

Plastic packaging consists of single polymers, e.g., polyethylene terephthalate (PET), polypropylene (PP), high (HDPE) and low (LDPE) density polyethylene, polystyrene (PS), and polyvinylchloride (PVC), or plastic multilayers which are combinations of several types of plastic layers. All listed polymers belong to the group of thermoplastics that can undergo mechanical recycling. In contrast, plastic multilayers are currently not recycled into new food packaging (Life rPack2L, 2017), because they are often immiscible and may only be blended by the addition of compatibilizers generating non-food grade materials (Hopewell et al., 2009; Ragaert et al., 2017).

During mechanical recycling, polymer backbones are partially degraded because heating can lead to breakage of intramolecular bonds resulting in a lower molecular weight distribution and

changed mechanical and optical properties of the recycled polymer, e.g., increased brittleness and opacity (Conte et al., 2014; Curtzwiler et al., 2011; De Santis and Pantani, 2013; Ignatyev et al., 2014; Incarnato et al., 1998, 2003). Additionally, plastic polymers may already have been damaged by external influences during use or waste management, e.g., contact with acidic substances, UV radiation, and extreme temperatures. These inherent properties of plastics have led to their classification as non-permanent materials (although they are highly resistant to complete degradation in the environment). Some types of plastic additives, e.g., antioxidants and stabilizers, form intended reaction products, thereby losing their original function. To compensate such unwanted changes, mechanical recycling processes require the addition of virgin polymers and/or further additives (Achilias et al., 2008; Curtzwiler et al., 2011).

Plastic packaging is mainly made of fossil-fuel based raw materials. The use of renewable raw materials has steadily increased over the last years (European Bioplastics, 2017), but is still of considerably small market share. Recyclability is not influenced by the source of the raw materials, but by the type of plastic. Fully or partially bio-based plastics that have the same material properties as conventional plastics can be recycled in existing recycling streams (e.g., PE, PET). Other bio-based plastics are compostable under defined conditions, hence mechanical recycling of these materials does not have high priority (e.g., polylactide, starch- and cellulose-based materials). However, wrong or incomplete sorting of any of these materials may strongly disturb recycling and/or composting leading to products of low quality or even break-down of the processes (Hopewell et al., 2009; Weithmann et al., 2018). Thus, careful separation of all polymers into individual material streams is a prerequisite for successful recycling.

3.1.1. Safety evaluation of recycling processes for plastic FCMs

The application of mechanically recycled plastic in contact with food requires special attention, because it may not only contain degradation products of polymers and additives, but also incidental contaminants arising from previous use and misuse by consumers, cross-contaminations from waste disposal, and environmental contaminants. Furthermore, polymers of the same type, but not of food-grade quality, may also enter the recycling stream and increase the level of possible contaminants. Therefore, advanced sorting and mechanical recycling processes have been established for plastic FCMs including additional cleaning steps to reduce contaminant levels, e.g., by incubation at high temperature, vacuum or inert gas treatment, or surface treatment with non-hazardous chemicals (Welle, 2011). Extraction with supercritical carbon dioxide is another option for the purification of post-consumer plastic waste (Anouar et al., 2015; Palkopoulou et al.,

Table 1
General properties of food packaging materials with respect to their recyclability.

Material	Material life-time	Recycling feasibility	# of cycles	Recycled material used in FCMs	Comments
Plastics	Non-permanent	Yes (thermoplastic polymers) No (plastic multilayers, thermoset polymers)	Limited –	Yes –	Mostly PET beverage bottles
Paper & board	Non-permanent	Yes	Limited	Yes	Recommendation/legal requirement in CH: not in direct contact with food
Metals	Permanent	Yes	Unlimited	Yes	
Glass	Permanent	Yes	Unlimited	Yes	
Multi-material multilayers	Non-permanent (paperboard)	Yes	Limited	Yes	Recycled paperboard not used in beverage cartons
	Non-permanent (plastic)	No	–	–	No efficient separation of plastic and aluminum layers possible
	Permanent (Al)	No	–	–	

2016). Such operations aim to reduce any chemical contamination to levels not posing a risk to human health. Risk assessments typically consider the quality of the input, the efficiency of the recycling process to remove contaminants, and the intended use of the recycled plastic (EFSA, 2008a). Specific challenge tests have been designed to demonstrate decontamination efficiency of recycling processes by simulating high levels of contamination, e.g., caused by consumer misuse (EFSA, 2008a; US FDA, 2006). During the challenge tests, plastic flakes are spiked with selected chemicals, so called surrogates, representing all groups of common contaminants and then submitted to all steps of the recycling process (US FDA, 2006; 21 CFR 170.39). After recycling, levels of residual surrogates are analyzed for assessing the process' efficiency to remove different types of contaminants. In order to make a recommendation for the maximum acceptable level(s) of contaminants in recycled plastic, the US FDA refers to the Threshold of Regulation approach, i.e., chemical contaminants shall not exceed migration levels of 0.5 ppb, leading to an estimated daily intake of 1.5 $\mu\text{g}/\text{person}/\text{day}$ (21 CFR 170.39; US FDA, 2006). EFSA bases the criteria for the safety evaluation of PET recycling processes on a human exposure threshold of 0.0025 $\mu\text{g}/\text{kg}$ bodyweight/day for any unknown contaminant possibly present (EFSA, 2011).

The efficiency of recycling processes and the quality of the recycled product mainly depend on the physico-chemical properties of the plastic, as well as the process conditions, and the purity of the input plastic waste. Recycling of PET beverage bottles has become standard due to the relatively high inertness of PET, its resistance to higher temperatures, and the establishment of collection systems for (mostly) food-contact grade PET (Welle, 2011). Thus, several dozens of bottle-to-bottle PET recycling processes have been developed in the last 25 years. In contrast, the recycling of other polymers is far less established, although they significantly contribute to the total quantity of food packaging (Poças et al., 2009; US FDA, 2007). Recycling of polyolefin packaging requires advanced cleaning and assessment protocols, because diffusion of a given substance is by orders of magnitude faster than in PET (Dole et al., 2006), strongly increasing migration rates, but also sorption of contaminants (Palkopoulou et al., 2016). Thus, tolerable residual concentrations of contaminants in recycled polymers are far lower in polyolefins than in PET. In addition, polyolefins are far less heat-resistant than PET, implying another challenge to efficient and effective decontamination.

Since 2010, EFSA adopted and published 79 scientific opinions evaluating more than 130 processes for recycled plastic to be used in FCMs (status: March 20, 2018) (EFSA, 2018). 95% of the processes describe recycling of PET, while the remaining 5% refer to polyolefins (PP and/or HDPE) (Fig. 1). EFSA has not received applications for any other types of plastic. At present (April 2018), none of the evaluated processes have been authorized by the EC. EFSA does not consider PET recycling as safety concern when processes are operated under the specified conditions. In most cases, input to the recycling process shall not exceed 5% of PET from non-food consumer applications. Further, it shall be ensured that potentially unknown contaminants do not migrate above a modelled migration level of 0.1 $\mu\text{g}/\text{kg}$ food. In some cases, the content of recycled material in the final product and/or its application are restricted. Polyolefin recycling processes were not considered of concern by EFSA if the recycled material originates from a product loop in a closed and controlled chain and the recycled material is used only in contact with certain food types (e.g., fruits, vegetables, and prepacked meat). However, in two scientific opinions for post-consumer polyolefins EFSA demanded additional data to be able to decide on the processes' safety.

Since 1990, the US FDA has issued 206 favorable opinions in the form of No Objection Letters for submissions on post-consumer

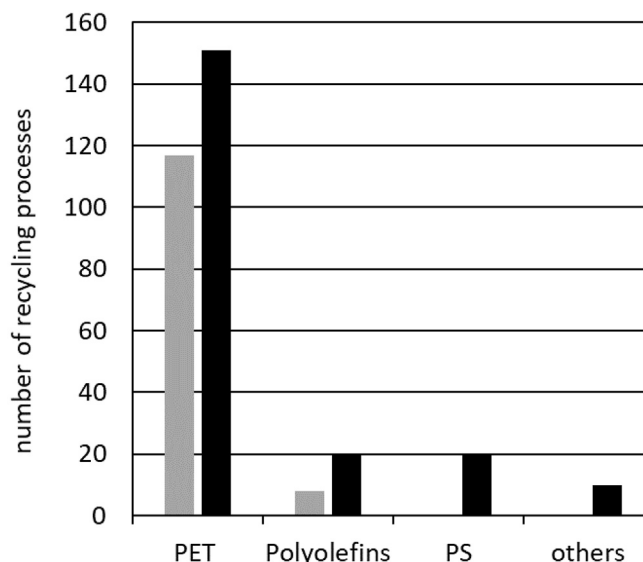


Fig. 1. Recycling processes for plastic FCMs with favorable opinions published by EFSA (grey columns) and the US FDA (black columns) (status: March 20, 2018).

recycled plastics for food contact articles (status March 20, 2018) (US FDA, 2017). 153, 22, and 21 submissions referred to PET, PS, and polyolefin recycling processes, respectively (Fig. 1). Six additional submissions dealt with specific issues related to PET recycling, e.g., coatings and functional barriers, and the remaining 4 submissions considered the recycling of polycarbonate (PC), PEN resin, PS and PP, and plastic grocery bags.

3.1.2. Contaminants in recycled plastics

Different groups of contaminants, e.g., oligomers, additives and their degradation products, as well as chemicals derived from previous (mis)uses, have regularly been reported in recycled plastic (Camacho and Karlsson, 2000; Dutra et al., 2011; Nerin et al., 2003). However, such studies often reflect the random distribution of contaminants, since their presence and identification depend on many factors including the production and recycling processes, the use and waste management phase as well as the applied analytical methods. Here, we provide a short overview of some typical groups of contaminants that have been found in recycled plastic derived from and/or intended for FCMs (Table 2).

3.1.2.1. Flavor, aroma, and odor compounds. Flavor compounds from previous uses belong to common contaminants in post-consumer plastic packaging. They may originate from intended applications, e.g., food, cosmetics and cleaning agents but also from misuse of the packaging by consumers, or cross-contaminations.

The presence of citrus-based essential oils has been attributed to soft drinks stored in PET beverage bottles (Bayer, 2002). Conventionally recycled PET contained traces of, e.g., limonene as well as γ -terpinene and *p*-cymene, but recycling processes including additional cleaning steps have efficiently removed such flavor compounds (Franz et al., 2004; Nerin et al., 2003; Triantafyllou et al., 2002). Other compounds from food-grade PET used in non-food applications such as mouthwash, personal hygiene and household cleaners contributed to the contamination of post-consumer PET (Bayer, 2002).

The fragrance and flavor compounds limonene, 3-carene, beta-myrcene, and terpinolene were measured in recycled, but not in virgin HDPE (Camacho and Karlsson, 2000). Soft drinks are typically sold in PET, but not in HDPE bottles. Thus, contaminations of

Table 2
Groups and examples of typical contaminants in recycled plastic FCMs.

Typical contaminants	Possible origins	Examples	References
Flavor, aroma, odor compounds	Previous use (e.g., sorption from food and non-food applications), cross-contaminations	Limonene (CAS 5989-27-5), γ -terpinene (CAS 99-85-4), <i>p</i> -cymene (CAS 99-87-6) and further citrus-based essential oils in conventionally recycled PET Limonene, 3-carene (CAS 13466-78-9), betamyrcene (CAS 123-35-3), terpinolene (CAS 586-62-9), 11 esters, 5 alcohols in recycled HDPE	(Bayer, 2002; Franz et al., 2004; Nerin et al., 2003; Triantafyllou et al., 2002) (Camacho and Karlsson, 2000)
Oligomers, monomers and derivatives	Production of virgin materials; degradation of polymers during use and recycling	Linear and cyclic PET oligomers, predominantly dimers and trimers in recycled PET Increased total migration of -presumably- oligomers from recycled low molecular weight PP Acetophenone (CAS 98-86-2) and benzaldehyde (CAS 100-52-7) in PS Increased total migration at elevated temperatures from recycled PS	(Bentayeb et al., 2007; López et al., 2014; Nerin et al., 2003; Triantafyllou et al., 2002) (Incarnato et al., 1998) (Vilaplana et al., 2007) (Kanwal et al., 2007)
Additives and their degradation products	Production of virgin material; (intended) degradation reactions during use and recycling; cross-contaminations	Seven UV absorbers (e.g., UVA-1 (CAS 3864-99-1), UVA-3 (CAS 1843-05-6)), four antioxidants (AOX-24 (CAS 93-43-9), AOX-26 (CAS 96-69-5), AOX-27 (CAS 90-66-4), HALS-3 (CAS 71878-19-8)), one plasticizer (diethylene glycol dibenzoate (CAS 120-55-8) in recycled PET Diisononyl adipate (CAS 33703-08-1), diisononyl phthalate (DINP, CAS 28553-12-0), Uvitex OP (CAS 7128-64-5), oleamide (CAS 301-02-0), Tinuvin 328 (CAS 25973-55-1), diethyltoluamide (CAS 13462-3) in recycled PET Bis(2-ethylhexyl) phthalate (DEHP, CAS 117-81-7), diisobutyl phthalate (DiBP, CAS 84-69-5), dibutyl phthalate (DBP, 84-74-2) and benzyl butyl phthalate (BBP, CAS 85-68-7) in recycled PET Adipates and erucamide (CAS 112-84-5) in post-consumer PET Nine phthalates in 20 waste, 8 virgin and 28 recycled plastic samples Irgafos 168 (CAS 31570-04-4), Irganox 1010 (CAS 6683-19-8), Chimisorb 944 (CAS 71878-19-8), and their degradation products in recycled polyolefins Degradation products of additives (e.g., 2,4-di-tert-butylphenol (CAS 128-39-2), and ethylbenzene (CAS 100-41-1) and xylene isomers) in recycled HDPE	(Bentayeb et al., 2007) (Dutra et al., 2014) (Keresztes et al., 2013) (Franz et al., 2004) (Pivnenko et al., 2016a) (Coulier et al., 2007) (Camacho and Karlsson, 2000; Dutra et al., 2011; Welle, 2005)
Contaminants from non-food grade plastic and sorted plastics; consumer misuse	Recycling of non-food grade or poorly consumer misuse	Brominated flame retardants in different types of black plastic FCMs 2-Methoxynaphthalene (CAS 93-04-9), sulfuric compounds and ethanol with fusel oil	(Puype et al., 2015; Samsonek and Puype, 2013) (Widén et al., 2005)
Inorganic elements	Catalysts; environmental origin; manufacturing processes		(Dutra et al., 2014; Nerin et al., 2003; Whitt et al., 2013, 2016)

HDPE with these aroma compounds may originate from the contact between post-consumer PET and HDPE bottles during collection (Welle, 2005). Furthermore, 11 esters and 5 alcohols were only identified in recycled, but not in virgin HDPE samples (Camacho and Karlsson, 2000). Their origin was assigned to the previous content of the packaging, e.g., personal hygiene products or cleaning agents.

3.1.2.2. Oligomers. Oligomers are unintentional byproducts formed during the synthesis of plastics. Additionally, they may also be generated during use and recycling of polymers. Regardless of their origin, oligomers may be present in the recycled material and migrate into food.

Linear and cyclic PET oligomers have been measured in recycled PET, with di- and trimers being the predominant species (Bentayeb et al., 2007; López et al., 2014; Nerin et al., 2003). Triantafyllou et al. (2002) stated that such oligomers are not recycling-related contaminants, but inherent to the PET matrix, whereas López et al. (2014) observed increased levels of cyclic oligomers in PET that had been reprocessed up to five times.

An increase in total migration levels and structural changes have been observed for different types of recycled PP when compared to the respective virgin materials (Incarnato et al., 1998). Especially for low molecular weight PP, the migration rates of -presumably-

oligomers increased after recycling.

Acetophenone and benzaldehyde are oxygenated derivatives of styrene, which have been found in higher relative abundances in recycled than in virgin PS samples, whereas virgin PS contained higher concentrations of styrene monomers and dimers than recycled material (Vilaplana et al., 2007). Kanwal et al. (2007) measured thermal degradation of PS after recycling, and observed increased levels of migration into vegetable oils at high temperatures.

3.1.2.3. Additives and their degradation products. Additives fulfill a large variety of different functions in plastic polymers by improving production processes as well as appearance and performance of the final products. In Europe, around 600 substances are authorized as additives and polymer production aids for plastic FCMs (EC 10/2011; EC 10/2011). Some additives are intentionally degraded during use (e.g., UV stabilizers and antioxidants), thus increasing the number of potential contaminants in the material. Additives are generally substituted during recycling to compensate losses in functionality while residual additives and their break-down products may remain in the plastic material. A special group of additives is intended to accelerate the degradation of oxo-degradable polyolefins in the environment by enhancing their oxidation. There is an ongoing debate on whether these materials pose problems in the

recycling of conventional polyolefins by destabilizing the recycled polymer (UNEP, 2015). Finally, the origin of many contaminants that have been measured in recycled plastics could not be identified, but may be explained by their abundance in the environment (e.g., phthalates) or cross-contaminations during waste management.

Seven UV absorbers that are known to be used in PET have been analyzed in four samples supplied by several PET recycling companies (Bentayeb et al., 2007). Furthermore, the plasticizer diethylene glycol dibenzoate and four antioxidants which are commonly used in PVC and/or polyolefins have been measured in the same set of samples. However, the levels of these contaminants have not been compared to virgin material. Dutra et al. (2014) measured several authorized plastic additives in recycled PET, including the plasticizers diisononyl adipate and diisononyl phthalate (DINP), the optical brightening agent Uvitex OP and the slip agent oleamide. Keresztes et al. (2013) detected the phthalates bis(2-ethylhexyl) phthalate (DEHP), diisobutyl phthalate (DiBP), dibutyl phthalate (DBP) and benzyl butyl phthalate (BBP) in water samples from PET bottles with 20–30% recycled content. In contrast, DiBP, DBP and BBP were found to be absent, and the levels of DEHP were strongly reduced in water samples from bottles composed of virgin PET. It remains unclear why these three studies reported plasticizers in the recycled material, because they are typically not needed as additives in PET (Enneking, 2006). Franz et al. (2004) proposed that phthalates as well as adipates and erucamide were introduced into post-consumer PET as irregular external contaminants derived from other polymer types. This is in line with the results of Pivnenko et al. (2016a) who analyzed the levels of nine phthalates in plastic waste samples as well as in recycled and virgin plastics. A strong correlation has been observed between the source of a plastic sample and its phthalate content, but neither the plastic resin nor the combination of sample source and plastic resin has been associated with the phthalate content. Significantly higher levels of DiBP, DBP and DEHP have been measured in waste plastic samples and recycled household plastics than in virgin and recycled industrial plastics.

For HDPE and PP, some additives showed increasing migration rates during repeated recycling (Coulrier et al., 2007). Higher migration levels of the stabilizers Irgafos 168, Irganox 1010 and Chimisorb 944 from recycled polyolefins were explained by the addition of these chemicals during each recycling step, but not by the processes themselves. Similarly, degradation products of additives typically used in HDPE and PP increased after several recycling sequences (Coulrier et al., 2007). Other studies also reported higher levels of degradation products in recycled HDPE which were derived from additives, e.g., 2,4-di-*tert*-butylphenol (Dutra et al., 2011; Welle, 2005) and ethylbenzene and xylene isomers (Camacho and Karlsson, 2000).

3.1.2.4. Contaminants derived from non-food grade plastics and consumer misuse. Brominated flame retardants (BFRs) have been measured in samples of black plastic FCMs from the European market indicating the recycling of plastic derived from waste electric and electronic equipment (WEEE) into FCMs (Puype et al., 2015; Samsoněk and Puype, 2013). This practice is not compliant with FCM regulations (EC 10/2011; EC 282/2008; Puype et al., 2017). Furthermore, the co-occurrence of other, non-food grade substances derived from illicit WEEE is to be expected.

Plastic bottles may also be used by consumers to store non-food items or liquids before recycling. In such cases, the plastic may have absorbed chemicals which subsequently could enter the recycling stream. A study by Widén et al. (2005) on chemicals in refillable PET bottles provided an overview of possible contaminants and their sources that could also be relevant for the recycling of post-

consumer PET. Examples of contaminants were 2-methoxynaphthalene (possibly derived from liquid fabric conditioners), sulfuric compounds (possibly derived from food products), and ethanol with fusel oils (possibly derived from home-distilled alcohol).

3.1.2.5. Inorganic elements. Dutra et al. (2014) investigated the migration of nine inorganic elements from 17 samples of virgin PET, recycled PET and HDPE into three simulants. In general, aluminum and iron were the elements with the highest migration levels, but their origin remained unknown. Antimony which is used as catalyst in the synthesis of PET commonly migrated from both recycled and virgin PET. Additional cleaning steps during recycling removed inorganic elements with different efficiencies. The levels of inorganic elements were generally lower in recycled HDPE than in PET. Silicon, calcium, sodium, iron, magnesium, aluminum, and zinc migrated from recycled PET samples, but the origin of these quite ubiquitous elements has not been clarified (Nerin et al., 2003). 29 out of 200 food packaging items made of recycled PET contained a combination of cadmium, chromium, nickel, antimony, and lead (Whitt et al., 2013). 22 of those 29 PET samples were further tested for the release of heavy metals into water and 5% aqueous citric acid. Migration of cadmium, lead or antimony was not measured, but most of the samples released chromium and/or nickel above detectable levels into 5% aqueous citric acid (Whitt et al., 2016).

3.2. Paper and board

Paper and board are mainly used for the packaging of dry foods and as secondary packaging. Furthermore, coating or waxing of paper and board extend their applications to the packaging and serving of wet and fatty food types.

Since the 1960s, paper and board have increasingly been recovered and recycled into new fiber-based products in many countries worldwide. At present, paper fiber is recycled 3.5 times on average in Europe, but up to 7 cycles are technically feasible (ERPC, 2016). Recycled material is often favored over paper and board made from fresh fibers, because recycling reduces waste and saves raw materials and energy (Martens and Goldmann, 2016). For successful recycling, separate collection systems for paper and board have been established preventing contaminations by, e.g., food waste. However, in contrast to some types of plastic, it is not common to fractionate the collected material into food-grade and non-food grade streams before recycling. The production of recycled paper and board resembles the process for virgin fibers, but requires additional steps: After separating the recovered paper and board into defined technical grades, they are mixed with water to produce a pulp. Next, non-fibrous parts such as staples, textiles, and tape are removed. Subsequently, the pulp is ground in a disperser, water is removed in drum filters or screw extractors, and the fibers are cleaned by chemical, thermal and/or mechanical treatments. Optionally, bleaching and deinking may be applied to enhance the appearance of the final product. The entangled fibers are then mixed with fresh fibers to maintain quality and processed on a paper (board) machine producing the final material.

Chemical substances of various origins are generally present in waste paper and can eventually be carried over into the recycled product. They include additives introduced during production and often intended to be retained in the paper product, e.g., fillers, retention aids, sizing agents, coatings, biocides, and synthetic binders. Furthermore, paper is commonly printed, dyed, glued, or labelled leading to the presence of printing inks, adhesives, photoinitiators, solvents, plasticizers, surfactants, and pigments in the waste paper. Contaminants may also be introduced during use or waste management, because paper and board are prone to

absorbing chemicals. Therefore, recycled paper and board used as primary or secondary food packaging are usually made from rather undefined sources.

3.2.1. Contaminants in recycled paper and board

Two data compilations list more than 9000 and 6000 unique compounds that are potentially used in (printed) paper and board and may be carried over into recycled material, giving an indication of the extent and diversity of possible chemical migration from recycled paper and board (Pivnenko et al., 2015a; Van Bossuyt et al., 2016). By far not all, but hundreds of chemicals from these lists have been measured in packaging made from recycled paper and board and/or in the respective packaged food. More than 250 substances were counted in an attempt to determine all potentially health-relevant components in recycled paperboard used for food packaging (Biedermann and Grob, 2013). Their complete identification and toxicological assessment has been judged to be unrealistic (Grob, 2014). Additionally, the results of a comprehensive migration study showed that concentrations of known and unknown contaminants in recycled board can hardly be reduced to safe levels (BMELV, 2012). In 2016, Vápenka et al. (2016) identified 101 chemicals in 132 samples of paper-based food packaging from the Czech market. In this study, the levels of 10 typical contaminants, namely anthraquinone, bisphenol A (BPA), 4-methylbenzophenone, benzophenone, DBP, DEHP, diisodecyl phthalate (DiDP), DiNP, and two diisopropylphthalene isomers were compared in plain papers with recycled fibers content below 10% w/w and above 90% w/w. Except for anthraquinone, the concentrations of all contaminants were significantly higher in samples containing >90% recycled pulp. These substances represent groups of chemicals that have already been detected in food packaging made from recycled paper and board or in waste paper streams (BMELV, 2012). Here, we summarize further studies providing evidence for elevated levels of typical contaminants in recycled paper and board packaging (Table 3).

3.2.1.1. Mineral oil hydrocarbons. In recent years, particular emphasis has been placed on understanding the migration of mineral oil hydrocarbons (MOHs) from recycled paper and board. MOHs generally consist of a highly complex mixture of mineral oil saturated hydrocarbons (MOSH) and mineral oil aromatic hydrocarbons (MOAH). These chemicals have been measured frequently and at high concentrations in many FCMs made from recycled paper and board and in the packaged food (Barp et al., 2015; Biedermann et al., 2013; BMELV, 2012; Dima et al., 2011; EFSA, 2012; Lorenzini et al., 2013; Lorenzini et al., 2010; Vollmer et al., 2011). The recycling of newspapers printed with mineral-oil based inks has been identified as the most significant source of MOHs (Biedermann and Grob, 2010), but adhesives, waxes, processing aids, and other unknown sources may further add to MOH contaminations in recycled paper and board (Biedermann and Grob, 2012).

3.2.1.2. Bisphenols. The finding that bisphenols are spread through paper recycling has been supported by several studies showing their presence in waste paper and/or recycled paper and board (BMELV, 2012; Gehring et al., 2004; Liao and Kannan, 2011; Pérez-Palacios et al., 2012; Suciú et al., 2013). For example, an analysis of 15 types of paper from different fractions of household waste detected BPA and its structurally similar alternative bisphenol S (BPS) in 100 and 73% of the samples, respectively (Pivnenko et al., 2015b). For both chemicals, thermal paper receipts displayed the highest concentrations, because many of them contain unbound bisphenols. Other proposed sources of BPA in the recycling stream are ink and glue formulations added to paper products (Pivnenko

et al., 2016c).

3.2.1.3. Phthalates. Phthalates have commonly been measured in food packaging made from recycled paper and board. Typical sources of phthalates in the recycled pulp are inks, lacquers and adhesives (Fierens et al., 2012; Poças et al., 2010). Amongst others, DEHP, DBP, DiBP, BBP, and diethyl phthalate (DEP) have regularly been identified in waste paper as well as recycled paper and board samples, and their migration contributes to phthalate levels in food (Ahmadkhanliha and Rastkari, 2017; Biedermann et al., 2013; BMELV, 2012; Bononi and Tateo, 2009; Bradley et al., 2013; Gärtner et al., 2009; Geueke and Muncke, 2017; Giannetti et al., 2017; Kappenstein et al., 2012; Pivnenko et al., 2016c; Poças et al., 2010; Suciú et al., 2013; Summerfield and Cooper, 2001; Vápenka et al., 2016; Vavrouš et al., 2016).

3.2.1.4. Diisopropylphthalenes (DIPN). In 1994, migration of six DIPN isomers from recycled paper and board packaging was reported for the first time (Sturaro et al., 1994). More recent migration studies have demonstrated that DIPN migration from recycled paper and board packaging is still common (Achilias et al., 2008; Gärtner et al., 2009; Giannetti et al., 2017; Lorenzini et al., 2013; Mariani et al., 1999; Sturaro et al., 2006; Summerfield and Cooper, 2001; Zhang et al., 2008). Mixtures of DIPNs are applied as solvents in carbonless copy paper substituting the previously used polychlorinated biphenyls (Barp et al., 2015). Thus, the recycling of office papers into food packaging is the main reason for the frequent detection of DIPN in packaging as well as in packaged food (Pivnenko et al., 2015a).

3.2.1.5. Photoinitiators. Photoinitiators are components of UV-curable printing inks and varnishes. They are intended to be consumed during application of the ink, but residual levels can remain in the ink due to incomplete reaction. The migration of more than 20 different photoinitiators from food packaging has been reported by Aparicio and Elizalde (2015). Of these, benzophenone and 4-methylbenzophenone have been identified most frequently in recycled paper and board (Anderson and Castle, 2003; Castle et al., 1997a; Giannetti et al., 2017; Koivikko et al., 2010; Song et al., 2000; Vápenka et al., 2016). Eight photoinitiators migrated from paperboard packaging into different food types, thereby transferring up to 24% of the total amounts measured in the packaging (Biedermann et al., 2013).

3.2.1.6. Inorganic elements. Paints and pigments have been identified as main sources of inorganic contaminants in paper and board (Mertoglu-Elmas, 2017). Food packaging made from fresh fibers contained lower concentrations of aluminum, copper, molybdenum, barium, and lead than recycled paper and board (BMELV, 2012). The heavy metals lead, cadmium, zinc, and copper were present in recycled corrugated cardboard samples from the Turkish market (Mertoglu-Elmas, 2017). Low levels of zinc and iron migrated from 10 samples of recycled paper and board into food (Parry and Aston, 2004), and cadmium, chromium and lead were measured in selected paper and board materials intended for food contact, but the content of recycled material was not stated (Castle et al., 1997b).

3.2.1.7. Others. 2-Phenylphenol (OPP), which is used as antimicrobial agent, fungicide, surface disinfectant and raw material for pigments and rubber additives, was detected in 20 out of 23 samples of paper materials used for food packaging and could be correlated to the content of recycled paper pulp (Votavová et al., 2014).

Phenanthrene, a polycyclic aromatic hydrocarbon (PAH)

Table 3
Groups and examples of typical contaminants in recycled paper and board.

Typical contaminants	Possible origins	Examples	References
Mineral oil hydrocarbons	Mineral-oil based printing inks, adhesives, waxes, and processing aids	Mineral oil saturated hydrocarbons (MOSH), mineral oil aromatic hydrocarbons (MOAH)	(Barp et al., 2015; Biedermann et al., 2013; BMELV, 2012; Dima et al., 2011; EFSA, 2012; Lorenzini et al., 2013; Lorenzini et al., 2010; Vollmer et al., 2011)
Bisphenols	Thermal paper receipts; ink and glue formulations	Bisphenol A (BPA, CAS 80-05-7) and bisphenol S (BPS, CAS 80-09-1)	(BMELV, 2012; Gehring et al., 2004; Liao and Kannan, 2011; Pérez-Palacios et al., 2012; Pivnenko et al., 2016c; Pivnenko et al., 2015b; Suciú et al., 2013)
Phthalates	Inks, lacquers, adhesives	DEHP, DBP, DiBP, BBP, diethyl phthalate (DEP, CAS 84-66-2), diisodecyl phthalate (DiDP, CAS 26761-40-0), DINP	(Ahmadkhanliha and Rastkari, 2017; Biedermann et al., 2013; BMELV, 2012; Bononi and Tateo, 2009; Bradley et al., 2013; Fierens et al., 2012; Gärtner et al., 2009; Geueke and Muncke, 2017; Giannetti et al., 2017; Kappenstein et al., 2012; Pivnenko et al., 2016c; Poças et al., 2010; Suciú et al., 2013; Summerfield and Cooper, 2001; Vápenka et al., 2016; Vavrouš et al., 2016)
Diisopropyl naphthalenes (DIPN)	Carbonless copy paper	2,6-Diisopropyl naphthalene (2,6-DiPN, CAS 24157-81-1), 2,7-diisopropyl naphthalene (2,7-DiPN, CAS 40458-98-8)	(Barp et al., 2015; BMELV, 2012; Pivnenko et al., 2015a; Sturaro et al., 1994; Triantafyllou et al., 2007)
Photoinitiators	UV-cured printing inks	Benzophenone (CAS 119-61-9), 4-methylbenzophenone (CAS 134-84-9), methyl-1-benzoylbenzoate (CAS 606-28-0), isopropylthioxanthone isomers	(Achiliás et al., 2008; Biedermann et al., 2013; BMELV, 2012; Gärtner et al., 2009; Giannetti et al., 2017; Koivikko et al., 2010; Lorenzini et al., 2013; Mariani et al., 1999; Song et al., 2000; Sturaro et al., 2006; Summerfield and Cooper, 2001; Vápenka et al., 2016; Zhang et al., 2008)
Inorganic elements	Paints, pigments		(BMELV, 2012; Castle et al., 1997b; Mertoglu-Elmas, 2017; Parry and Aston, 2004)
Other contaminants	Recycling of non-food grade paper & board; processing aids; reaction products; various additives	2-Phenylphenol (OPP, CAS 90-43-7) Phenanthrene (CAS 85-01-8), pyrene (CAS 129-00-0), anthracene (CAS 120-12-7), fluorene (CAS 86-73-7)	(Votavová et al., 2014) (BMELV, 2012)

commonly present in black coloring pigments of newspaper inks, was measured in all samples made from recycled board, but not in packaging composed of only fresh fibers (BMELV, 2012). In the same study, pyrene was measured regularly in recycled materials, while anthracene and fluorene were identified sporadically.

Perfluoroalkyl substances (PFASs), a group of persistent chemicals used to convey non-stick properties and oil and water repellency to paper and board, have regularly been measured in paper and board food packaging (Begley et al., 2008; Trier et al., 2011; Vápenka et al., 2016), but it is currently not known whether recycling contributes to elevated levels of PFASs.

3.2.2. Approaches to reducing migration

The frequent detection of contaminants in recycled paper and board initiated a discussion on safer solutions. Technical and regulatory improvements have already been achieved, but the issue is not generally solved yet. Most recycling processes for paper and board include deinking steps detaching printing inks from the paper fibers and removing them from the pulp. Although deinking significantly reduces the levels of certain contaminants, it does not

suffice for guaranteeing the chemical safety of recycled paper and board (Jamnicki et al., 2015). The complete substitution of mineral-oil printing inks by, e.g., vegetable-based inks may eventually decrease the levels of MOHs in recycled paper and board. However, this solution will not quickly solve the complete migration issue, but only decrease the load of MOHs in a long-term view. To avoid extensive analytical and toxicological tests of recycled paper and board, alternative technical approaches aim for the highest possible reduction of migration. This can be achieved by using internal bags, applying barrier layers on the inner surface of the packaging or including adsorbents in the paperboard. Testing the efficacy of internal bags revealed that paper or polyolefin bags hardly reduced migration, whereas plastic multilayers slowed down migration efficiently, and an additional layer of aluminum was considered as absolute barrier (Biedermann et al., 2013; Richter et al., 2014).

3.3. Metals

Metal food packaging includes cans, tubes, other containers, caps and closures, films, and barrier layers. Cans made of aluminum

and steel are the most widely used types of metal-based beverage and food packaging. Metal cans achieve high recycling rates, but other types of metal food packaging are not commonly recycled. Irrespective of the source and the type of the metal, cans are generally lacquered with an organic coating to prevent reactions between the metal and the food as these can lead to corrosion and subsequent damage of the packaging, or off-tastes in the food. Can coatings reduce metal ion migration, although the coatings themselves are a well-known source of chemicals migrating into food (Grob et al., 2006).

3.3.1. Aluminum cans

Aluminum is widely used for beverage cans and other types of food packaging (e.g., foils, trays, tubes, beverage cartons, coffee capsules). Aluminum cans represent 90% of beverage cans worldwide (Rexam, 2016). Aluminum is a temperature resistant, permanent material with good barrier properties. In addition, it does not alter organoleptic properties of food and is relatively non-toxic (EFSA, 2008b). Aluminum recycling can continue almost indefinitely. However, untreated aluminum surfaces are prone to oxidation which can cause some loss of material during recycling. Aluminum recycling has environmental benefits in terms of energy savings, air pollution, and water use but it produces toxic gaseous and particulate emissions and waste products such as dross (Tsakiridis, 2012).

Global recycling rates of aluminum cans reached around 70%, with regional recycling rates differing between 30 and well-above 90% (IAI, 2009; US EPA, 2016). For other types of food and beverage packaging, aluminum recycling is practically non-existent due to collection difficulties and technical limitations (Marsh and Bugusu, 2007). Recycling processes for aluminum cans typically start with mechanical shredding, followed by a decoating or delacquering step to remove all internal and external lacquers, coatings, and printing inks. After this, aluminum scrap goes through melting into new production phases (Baeyens et al., 2010a).

Closed-loop recycling of aluminum cans enables high rate continuous recycling (Buffington, 2012; Das, 2006; Rexam, 2016). However, repeated recycling of the same aluminum sources may result in accumulation of unwanted metals and metalloids, introduced either as impurities, alloying elements, or coating residues left due to incomplete decoating before recycling (Li and Qiu, 2013; Løvik and Müller, 2014; Nakajima et al., 2010; Niero and Olsen, 2016). Indeed, raw materials used for aluminum production may be initially contaminated with low levels of geogenic metals. Mixed scrap may contain other metals eventually entering the aluminum recycling stream. Furthermore, aluminum is rarely used as a pure metal, but instead comes in a variety of alloys which are used for specific applications, e.g., typical alloys used for beverage can bodies differ from those used for can lids (Das, 2006; Løvik and Müller, 2014). Additional metal contaminants may originate from the organic coating and decoration of the cans which often contain pigments based on, e.g., titanium dioxide and zinc oxide.

The presence of other metals may result in downcycling, because the precise alloy composition is crucial for ensuring the required properties of the recycled aluminum (Nasu et al., 2001). Therefore, contaminations with metals need to be controlled and, ideally, avoided. Nakajima et al. (2010) analyzed alloy metals and impurities during aluminum scrap recycling and showed that most metal impurities are difficult to remove, because the majority of elements distributed among the metal phase but not among the slag or gas phases. Thus, most of the metal impurities can be expected to gradually increase with each recycling loop (Das, 2006). Depending on the specifications of the final alloy, necessary amounts of different metals can be supplemented. Addition of

primary aluminum is practiced often, as this helps reducing the end concentration of various impurities. Furthermore, modeling has shown that manganese concentrations decrease with increasing recycling rates during closed-loop recycling which makes it necessary to adjust the level of this alloying metal (Niero and Olsen, 2016). Thermal decoating is broadly used during aluminum recycling and carried out by blowing hot air on shredded can pieces. However, such a treatment may only remove the coating partially (Wang et al., 2007), possibly leading to a significant contamination of the recycled aluminum with titanium (Løvik and Müller, 2014; Rabah, 2003). Alternatively, chemical or combined decoating methods may offer higher levels of decoating efficiency along with a possibility to also recover titanium (Wang et al., 2007; Li and Qiu, 2013).

To ensure high recyclate quality, the separation of other metals and even different aluminum alloys before remelting could be one option to avoid the addition of primary aluminum and/or alloying elements (EAA/OEA, 2006; Niero and Olsen, 2016). In fact, numerous sorting technologies and methods for removal of impurities (both pre- and postmelting) during aluminum recycling have been proposed (Gaustad et al., 2012; Jung et al., 2013; Vieira et al., 2012; Zhang et al., 2011), but only a few have so far found a wider commercial application.

Since most aluminum cans are coated, direct contact between the metal and the beverage is rather uncommon. However, Veríssimo and Gomes (2008) reported aluminum migration from cans into beverages despite the protective coating. Migration of aluminum from coated cans into beer and tea increased with time and was more severe in dented than in intact cans. Whether the migration of other metals into beverages is influenced by the content and the quality of recycled aluminum in the can, is currently unknown.

3.3.2. Steel cans

Steel food packaging is mainly comprised of cans and other containers, lids, caps, and closures. Steel cans are produced from tin-coated steel (also called tinplate) or electrolytic chromium coated steel (ECCS). For most food categories, tinplate containers are usually lacquered with organic coatings to provide a barrier between metal and food; ECCS always requires an organic coating for corrosion resistance. For light colored, acidic fruits and juices, tinplate cans do not have organic coatings as tin is more easily oxidized than the food, thus preventing darkening and flavor changes but reducing shelf life to limit tin ion migration. Steel is a permanent material that can be infinitely recycled without loss of quality. However, it can be damaged by corrosion. Although rusting is not a problem for recycling, it should nonetheless be avoided as it leads to the dispersion of iron oxides. Steel recycling saves energy, reduces the environmental damage caused by mining activities and the emissions of greenhouse gases into the atmosphere (Canmaker, 2011). Current recycling rates are limited by the collection and sorting efficiency, as well as the need to manage possible contaminations with other metals. However, Germany, Belgium and the Netherlands recycle more than 90% of their steel packaging, while other EU nations and the US are approaching more than 70% (Mohr, 2016; US EPA, 2016).

Steps in the recycling of steel and tin from cans include magnetic separation, detinning, melting, and reforming (Baeyens et al., 2010b). During detinning, a caustic chemical solution dissolves the tin which is subsequently separated by electrolysis. Meanwhile detinned steel cans are drained, rinsed, and baled into squares. After heating up >1500 °C in a furnace and adding some molten iron, the steel can be used to make new cans as well as other types of products. In contrast to the recycling of aluminum cans, metal contaminants potentially deriving from the organic coatings of

steel containers mainly separate into slag phase eliminating the need for an additional decoating step.

Steel cans are often recycled in closed loops. Impurities which are difficult to separate after melting, such as copper but also tin, can lead to downcycling of steel (Savov et al., 2003). Such quality losses are prevented by dilution with pure steel. Due to low efficiency of technical separation after melting, the best approach to managing steel contamination entails careful sorting of scrap types along with efficient detinning of tinplate cans.

Whether the quality of steel in general and the use of recycled steel in particular have an impact on metal migration from cans is unknown. However, organic coatings do not fully prevent metal migration, and storage in open cans and denting may further increase the migration of metals into food (Kassouf et al., 2013; Petropoulos et al., 2017; Raptopoulou et al., 2014; Verissimo et al., 2016). Therefore, alloying elements (e.g., manganese, chromium), contaminants from previous metallic and organic coatings (e.g., tin, zinc) as well as impurities (e.g., lead, cadmium) should be controlled during steel recycling to comply with respective standards set to ensure food safety (Cederberg et al., 2015; EC, 1881/2006).

3.4. Multimaterial multilayers

Packaging consisting of several layers (i.e., multilayers) is widely used in the food and beverage industry. Such packaging is either made of different materials such as paperboard, aluminum, and plastic (i.e., multimaterial multilayers) or exclusively consists of plastic (i.e., plastic multilayers, for more details see 3.1). Multimaterial multilayers include beverage cartons which can have varying compositions, but are typically comprised of about 75% of paperboard, 20–21% of plastic (usually PE), and up to 5% of aluminum foil (Agamuthu and Visvanathan, 2014). PE layers are applied to prevent leakage, while aluminum protects drinks from light and oxygen and is therefore most often used in products intended to provide a relatively long shelf-life. Laminated films made of aluminum and plastic are another example for multimaterial multilayers. They are typically used in the form of pouches, tubes, and bags for the storage of a diverse range of foods and beverages (WRAP, 2011). Industrial processes for recycling laminated packaging are not yet broadly available (WRAP, 2011). Thus, we focus on beverage cartons as an example of a widely used packaging material for which recycling procedures have been developed and implemented.

Continuous growth is projected worldwide for the recycling of beverage cartons (Agamuthu and Visvanathan, 2014). The historical increase in beverage carton recycling in the EU, for example, has been from about 6000 tons (<1%) in 1992 to 100,000 tons (about 12%) in 1995, to 430,000 tons (about 47%) in 2016 (ACE, 2017). All three components of a typical beverage carton are on their own recyclable, but there is a risk of cross-contamination. Thus, paperboard needs to be separated from the polymeric and aluminum content. In dedicated facilities, the collected and sorted beverage cartons undergo hydropulping, a process involving mixing with water and mechanical treatment. The carton breaks down into a fiber soup which can be separated from the plastic and aluminum layers, as well as contaminants. The fiber obtained in this way can then be mixed with virgin material and used to make new paper products, including food packaging for dry products that are not in direct contact with food, and other paper products such as corrugated boxes, carton boxes, towels and tissue, stationery, and paper bags. After further separation, PE and aluminum may be used for energy recovery and recycling, respectively (Charlier and Sjöberg, 1995). Alternatively, both materials can be applied as a composite to produce, for example, roofing tiles, construction panels, or

garden furniture (Agamuthu and Visvanathan, 2014). Since recycled fractions of beverage cartons are not used in primary food packaging, potentially elevated migration levels are currently not an issue. However, the examples show that complete recycling in the strict sense is currently not feasible for beverage cartons.

3.5. Glass

Glass has been used in contact with food for thousands of years (e.g., tableware, jars). Glass is an inorganic permanent material, usually made up of silicon dioxide as main structural component, as well as alkaline and earth-alkaline metal oxides. Because of its molecular structure, glass is an inert material with high barrier properties, hence even small molecules cannot pass through glass. Neither does glass absorb molecules from the food it contacts, nor the environment. Migration from glass by diffusion-controlled processes from the glass bulk material is hindered (Müller-Simon, 2010). However, acidic foodstuffs can lead to an ion exchange between cations from the glass' inner surface and foodstuffs, resulting in migration; under basic conditions, hydrolysis of Si–O bonds can occur (Müller-Simon, 2010).

Soda-lime glass is the most prevalent type of glass and commonly used for beverage and food containers. Recycling of container glass across Europe reached 74% in 2014, with 11.6 million metric tons collected and some countries exceeding recycling rates of 90% (FEVE, 2016). Recycling of glass packaging saves raw materials, minimizes the energy needed for production of glass-packaging and reduces carbon dioxide emissions (Beerckens et al., 2011). For most efficient recycling, container glass needs to be collected separately by color (Beerckens et al., 2011; Beerckens and van Santen, 2008). Glass is then crushed into glass fragments, so called cullet, and sorted further by removing metals, plastics, paper, ceramics, stones, and porcelain (Beerckens et al., 2011). Next, up to 90% of cullet is mixed with virgin raw materials, consisting of silicon dioxide, soda ash (Na_2CO_3), and lime stone (CaCO_3), as well as additional ingredients in smaller quantities (e.g., coloring agents). The mixture is then melted at temperatures of 1350–1400 °C and formed into new glass products. Importantly, glasses from applications other than packaging should not be included in container glass recycling, because they have a different chemical composition that can hamper efficient melting or affect quality, for example by introducing hazardous elements. Examples are fluorescent light bulbs containing mercury, or glass TV screens and crystal glass which can both result in increased lead contents (Angeli et al., 2016; Ross, 2011). Further, heat-resistant glass used in kitchen and laboratory glassware is commonly made of borosilicate glass that disrupts the recycling of soda-lime glass.

Sands used for glass manufacture contain lead, a geogenic trace element, at typical levels of around 12 ppm (Hough, 2010), but lead concentrations in sandy soils can even range above 25 ppm or higher, due to air pollution and lead's legacy use as anti-knocking element in gasoline, mining, or spreading of sewage sludge (Archer and Hodgson, 1987). As a consequence, lead is also present in container glass and may migrate into foods via surface cation exchange by a diffusion mechanism (Angeli et al., 2016). Levels ranging up to 0.417 µg lead/L in glass bottled drinking water have been found after 6 months storage at room temperature, due to migration from the glass packaging (Shotyk and Krachler, 2007). Migration of lead from crystal glass (with a lead content of 28% by weight) may result in higher concentrations (Angeli et al., 2016). Cullet sorting is a common practice for removing lead from glass recycling. Hereby, X-ray fluorescence or UV fluorescence analysis is used to detect cullets with an increased lead content, allowing to automatically sort out such fragments (Ceola et al., 2016). Another possible remediation method for removing lead and other trace

heavy metal ions from glass surfaces may be the treatment with citric or acetic acid (JRC, 2015). Additional trace elements known to be hazardous and present in container glass are for example chromium VI and cadmium. Both are naturally present in the earth's crust and therefore their presence in container glass is to be expected at trace levels, however, they can also be intentionally added as colorants (Ross, 2011).

Glass packaging also lends itself to reuse, because it is inert and efficient to sanitize. Deposit schemes for glass beverage bottles exist worldwide. Thereby, customers pay a small deposit on a glass bottle when purchasing, and get refunded after returning the empty bottle. Refillable glass packaging can be reused up to 50 times, depending on the market requirements (R3 Consulting Group and Morawski, 2009).

4. Discussion

In the existing economic system, food packaging is generally designed for single-use and discarded after relatively short periods of time. To avoid negative impacts on the environment the fate of packaging waste needs to be controlled (Marsh and Bugusu, 2007). Reduction of material input, one major goal of the circular economy, has been proposed for reducing the total amount of food packaging (Allwood, 2014; Winans et al., 2017). Accordingly, for many types of food packaging, the weight per unit has strongly decreased over the last years. Such measures save resources and costs, but may affect the mechanical properties of the packaging. Furthermore, recycling processes may be impaired, as difficulties in the separation of ultra-light PET bottles have been demonstrated (Amazonas et al., 2017; Bevnet, 2017). Similarly, multilayer plastic packaging typically requires less material to achieve the same functionality as a mono-material but its recyclability is very limited (Ragaert et al., 2017). Thus, material reduction can come at the cost of recyclability, leading to immediate, but not repeated savings of resources and waste prevention. Reuse of packaging is recommended to accomplish the goals of the circular economy (EC, 2015; European Parliament, 2017), but for food packaging, reuse is commercially only feasible for refillable and cleanable containers (e.g., glass bottles, stainless-steel containers). Recycling has been implemented for different packaging materials in many countries to reduce the quantity of waste and its related environmental impact (Ghisellini et al., 2016). While higher quantitative recycling targets for different types of packaging waste are under discussion in the EU, specifications on the quality and the further use of the recycled materials are not available yet (EC, 2015; European Parliament, 2017, 2018; Lee et al., 2014). Hence, it remains open whether the recycled material shall be used for its original application or if it can only be used for other purposes due to lower functionality, potentially hindering repeated recycling. It needs to be clarified if this practice of downcycling is in line with the goals of the circular economy.

In terms of quality and safety, recycling of permanent materials, such as metal and glass, is generally considered suitable for food packaging, because the material properties do not change and the heat required for remelting destroys microorganisms and organic compounds (Marsh and Bugusu, 2007). However, repeated recycling of aluminum, steel, and glass may lead to the accumulation of unwanted metal ions in the material, e.g., when metal scrap is not sorted carefully or container glass is contaminated with crystal glass. Metal ion migration from glass surfaces can be a problem if high levels are present in the material. In food and beverage cans, coatings generally prevent metal ion migration and further interactions between the metal packaging and the food. In contrast, recycling of non-permanent FCMs, which is already limited to a few cycles and/or requires the addition of virgin materials, causes

higher safety concerns, because contaminants from previous uses are not *per se* removed or destroyed during processing, and new substances of concern can be formed or introduced. This makes the risk assessment of recycled, non-permanent FCMs even more challenging than evaluating the corresponding virgin materials. Novel approaches support risk assessment of recycled FCMs by addressing the chemical exposure to unknown substances and mixtures with biological activity: *In vitro* bioassays support the toxicity assessment of chemical mixtures migrating from recycled FCMs (Bengtström et al., 2016; Groh and Muncke, 2017; Rosenmai et al., 2017), while semi-quantitative analytical methods allow the screening and prioritization of substances based on their estimated exposure levels (Pieke et al., 2017). Another technique for analyzing the biological effects of single substances in unknown mixtures combines *in vitro* bioassays with planar chromatography and may allow for the simplified identification of a chemical's biological effect and its structure (Weiss et al., 2017). These approaches identify substances migrating from FCMs and/or their effects, but they do not solve the problem of increased levels of potentially hazardous substances in recycled, non-permanent materials. Therefore, specific restrictions may facilitate the use of recycled non-permanent materials by limiting the potential exposure to contaminants (e.g., by allowing certain uses only behind a functional barrier or for certain food types). Considering food packaging in a larger context, it would be ideal to combine the assessments of human health and environmental effects. Tools such as life-cycle assessment (LCA) have been applied to quantitatively compare the environmental impact of different types of food packaging (Morris, 2005; Vignali, 2016). However, LCA traditionally does not consider human exposure to chemicals related to product use (Fantke et al., 2016). To overcome this limitation, different models have been developed to be included in LCA for estimating chemical exposure from, e.g., food packaging (Ernststoff et al., 2017), cosmetics (Ernststoff et al., 2016), and consumer products (Huang et al., 2017). Such tools could compare the human health impact of chemical migration from recycled packaging versus virgin materials. Nevertheless, focusing on chemical safety within the circular economy will add a new level of complexity during assessment and may identify additional conflicting goals, e.g., between reduced environmental impacts and protection of human health.

In the long term, chemical safety of recycled food packaging could be improved by phasing out and carefully substituting hazardous substances in all materials that enter the recycling stream. Modeling has revealed that such a measure would efficiently reduce BPA, DEHP, and MOHs in recycled paper and board (Pivnenko et al., 2016b). However, a period of 10–30 years is estimated for these contaminants to reach insignificant levels. Improved decontamination strategies during recycling may decrease the levels of contamination, but lead to lower material yields (Pivnenko et al., 2016b). The mandatory use of mostly food-grade materials for recycling, as it is already required for plastic food packaging, could further improve the safety of recycled paper and board. In any case the cleaning efficiency of recycling processes to eliminate typical and random contaminants needs to be thoroughly characterized. In the short term, different activities can immediately contribute to a more efficient recycling system and sustainable use of food packaging: For example, packaging items may be (re)designed in view of their recyclability and reusability, current and future packaging trends (e.g., nanocomposites, active and intelligent materials) may be evaluated focusing on their safety and recyclability, and consumers may be further educated to support the goals of the circular economy (Wikström et al., 2017).

The concepts of reduction and reuse could also be rethought, while keeping the major functions (e.g., food preservation, transport) and the safety of packaging in mind. Reduction cannot only be

achieved by thinner, yet more complex materials, but also by simply decreasing the packaging to volume ratio (e.g., bigger portion sizes for durable foods), or by changing food production and food distribution systems entirely, with a focus on local production and local consumption of seasonal foods (Coelho et al., 2018). Zerowaste shops offer many types of food without packaging and require customers to bring their own, ideally reusable, packaging (Beitzen-Heineke et al., 2017). Reuse could also be strongly promoted by deposit schemes using few standardized containers that can be returned and refilled at many different sites, thus reducing transport distances. Packaging redesign can significantly contribute to reduction, reuse, or recycling, if the end of life is already considered during packaging development (Lofthouse et al., 2017). Sustainable food packaging in the circular economy may only be achieved by combining such efforts, considering any conflicting goals and involving all stakeholders, including food and packaging manufacturers, recyclers, decision makers, civil society, and consumers.

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