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Food Packaging Forum

Dossier – Plastic recycling

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1 Background

In 2009, R.C. Thompson stated that “at present our consumption of fossil fuels for plastic production is linear, from oil to waste via plastics” [1]. The steadily increasing production of plastic indeed causes severe environmental problems, which include the high energy demand during production, the consumption of fossil fuels and the accumulation of plastic waste in landfills and the natural environments [1, 2]. However, the statement ignores all the strategies and efforts undertaken to transform this one-way system into a closed-loop system.

In the context of packaging, approaches to reduce or slow-down the demand for virgin plastic have been developed and are already applied to different extents. These strategies include the **R**eduction of packaging weight and/or volume, the **R**euse of packaging and the **R**ecycling of certain polymers (**3R**). However, the development of efficient and clean recycling processes on an industrial scale is still a major challenge, although large efforts are currently undertaken to achieve these ends [3]. The source control of potentially hazardous chemicals contributes to the difficulties in achieving efficient processes.

Even though the market price of plastic waste is low, the added value created by recycling is also rather low. This is due to the downgraded quality and properties of recycled plastic in comparison to virgin materials. Furthermore, collecting, sorting and purification of waste streams are often difficult to achieve and expensive.

Processes with the aim to use recycled plastic for food packaging were first developed at the end of the 20th century [4]. However at the time, the legal situation did not permit the use of recycled plastic for food packaging, because of the risk of contaminations. Advances in technical processes and changes in legislations nowadays permit the use of recycled plastic in food contact materials (FCMs).

2 Recycling steps

2.1 Identification and sorting

The resin identification code

Thorough separation and sorting of the different materials is necessary to obtain recycled plastic of similar quality as the virgin plastic. In 1988, the Society of the Plastics Industry (SPI) devised the resin identification code (RIC) aiming at the efficient identification and separation of different plastics (Table 1) [5]. This system was not developed to inform about a product’s recyclability, but to inform consumers which types of plastic are collected for recycling. Products made from recycled materials are marked with an “R”-prefix (e.g. R-PET). In 2010, the RIC system was covered by the international standard ASTM D7611. In 2013, ASTM International issued the replacement of the three “chasing arrows”, which are often associated with recycling, by a solid equilateral triangle symbol to focus only on resin identification, not on recyclability.

Table 1. Resin identification code, old and new symbols

RIC Code	Material	Symbol Chasing arrows	Symbol Triangle
1	PET	1 PETE	1 PETE
2	HDPE	2 HDPE	2 HDPE
3	PVC	3 V	3 V
4	LDPE	4 LDPE	4 LDPE
5	PP	5 PP	5 PP
6	PS	6 PS	6 PS
7	others	7 OTHER	7 OTHER

Sorting systems

Although the RIC helps to identify the type of plastic used and may support presorting of waste by the consumer, it is not helpful when mixed waste streams have to be sorted at an industrial scale. For this purpose, manual or automated sorting systems exist that separate plastics intended for recycling from other waste. Usually, presorting efficiently segregates glass, metal and paper from the waste stream. Most of the material recovery or plastic recycling facilities apply automated sorting of the remaining plastic. Near-infrared and Fourier-transform spectroscopy is commonly used for polymer type analysis [6]. A recent research project developed a process using also mid-infrared spectroscopy at laboratory scale [7]. Optical color recognition systems allow the sorting of e.g. polyethylene terephthalate (PET) of different colors. X-Ray technology can be used for the identification of polyvinyl chloride (PVC) containers due to the high level of chlorine [8]. Optimized sorting may be achieved by applying a variety of these techniques in series. Further processes include triboelectric separation, density sorting in hydrocyclones, sorting in high-speed accelerators and separation by boiling [9-11]. Despite this high number of techniques, efficient separation is still a challenge, because it can be complicated by different shapes of the plastics, entrapped air, coatings and paints that slow-down or even impede the analysis.

2.2 Recycling processes

Depending on the final product, recycling processes are classified into four different categories. Plastic waste can be used for the production of the same type of polymers, alternative types of plastic, chemical building blocks, fuel or energy. In common language use, all these categories often fall under the term recycling although the precise use of the term only describes those processes that reform a waste material into the same product, thus closing a cycle.

Primary mechanical recycling

Primary recycling describes processes converting thermoplastic polymers into products with equivalent properties [6]. Plastic products not fulfilling product specifications and scrap produced during manufacture of plastics are generally directly recycled by re-extrusion [9]. Such closed-loop processes can only be applied for thermoplastic polymers and for plastics which have not been used or thoroughly cleaned and separated from other plastic types before recycling.

Secondary mechanical recycling

Secondary recycling generally leads to products of lower mechanical properties. Thermoplastic plastics composed of only one polymer may be mechanically recycled after use (e.g. PE, PP, PS, PET, and PVC) [9]. Efficient sorting and washing procedures that remove contaminations are prerequisites for this process. Although polymerization reactions are theoretically reversible, side reactions may occur e.g. under heating or UV irradiation. This may lead to a reduction of the polymer's molecular weight and its mechanical properties. Suitable drying and vacuum degassing procedures [12] and the addition of more stabilizers during recycling [13] can counteract this problem. However, this type of recycling cannot be maintained indefinitely as the plastic degrades over its lifetime.

Tertiary and quaternary chemical recycling

Tertiary recycling is usually defined as chemical recycling, where polymers are degraded into smaller molecules by chemical or also biological processes. The degradation products can serve as starting materials for new polymerization reactions or other chemical processes (e.g. the production of fuel). PET is one example of economically efficient chemical recycling, because de-polymerization can occur under relatively mild conditions and the reaction products can be re-used for the production of PET. Attempts at the recycling of polyolefins were also made in the recent years, but here industrial processes were much more difficult to establish [6, 14]. Quaternary technologies generally do not aim for the production of new materials, but rather for recovering energy from plastics through incineration.

3 Recycled materials

3.1 PET Recycling

General information

PET is a plastic material which is easily recyclable by secondary mechanical recycling as it is fairly inert and contains a limited range of additives. These characteristics result in low diffusion rates out of and into the polymer matrix [15]. Furthermore, PET has become the most used packaging material for water and soft-drinks worldwide [16, 17]. Public concerns over the environment, as well as recyclability and availability of collected PET bottles, promoted the development of recycling processes in many countries during the last two decades. In the beginning, PET recyclates were mainly used in the production of polyester fibers. But with supply of recycled PET being higher than the demand for these fibers, bottle-to-bottle recycling processes were developed. Together with the establishment of a regulatory framework, high recycling capacities for food-contact grade PET were established in many countries [18, 19]. Thereby recycled PET is often used in combination with virgin PET.

Recycling processes

Post-consumer PET packaging, especially bottles, are collected and transferred to materials recovery facilities, where the PET is separated from other materials, e.g. lids and labels, usually sorted by color and pressed into bales. In recycling facilities post-consumer PET is washed and ground into flakes. Detergents and 2-3% sodium hydroxide solutions are used as washing additives to remove dirt, labels, glue, and food leftovers from the surface of the polymer [12]. The flakes obtained after conventional recycling processes are typically not sufficiently clean for reuse in FCMs. Thus, additional steps for the decontamination of post-consumer PET are necessary. They usually include high temperature treatment, vacuum or inert gas treatment and surface treatment with non-hazardous chemicals to obtain so-called super-clean PET [12].

Risk assessment

Several contamination routes were identified as sources of concern during PET recycling (Figure 1) [15]. Contaminating chemicals can enter the recycling stream after misuse of PET containers by the consumer (e.g. by storing garden or household chemicals), when non-food contact PET or other plastics are not separated before recycling, when PET degradation products or process chemicals are not sufficiently removed, and when food components sorbed into the plastic during first use by a process called flavor scalping.

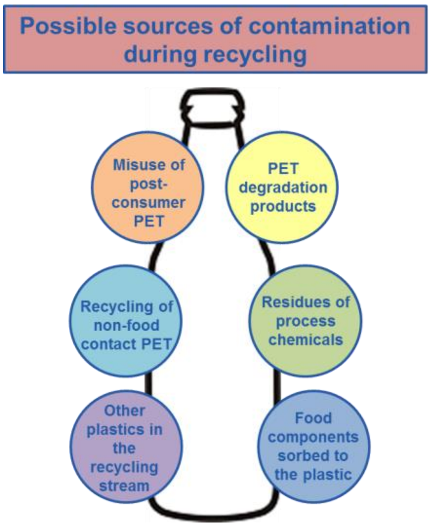

Figure 1. Contamination sources during PET recycling

It is generally agreed that comprehensive chemical testing of every batch of incoming recovered PET is technically impossible; neither can all production batches of recycled PET be tested [15]. As alternatives so-called "challenge tests" were developed to evaluate the efficiencies of recycling processes in removing contaminants and to be able to assess the compliance of recycled PET with legal requirements. In these tests, it is measured whether a recycling process can reduce any chemical contamination below a set limit. For this purpose, plastic is deliberately contaminated with defined concentrations of surrogate contaminants having different chemical characteristics and then fed into the recycling process. The remaining concentrations of contaminants are compared with the initial chemical load and cleaning efficiencies are calculated from these values. Challenge tests are currently employed by the U.S. Food and Drug Administration (FDA) and European Food Safety Authority (EFSA) to evaluate PET recycling processes [20-22].

In 2014, several scientific employees of European authorities published peer-reviewed recommendations for the development of a general scheme for the safety evaluation of mechanical PET recycling processes [15]. In this study, the evaluation of such processes by challenge tests is further supported. Additionally, the Threshold of Toxicological Concern (TTC) approach is recommended for setting contaminant levels in food below which a risk for human health can be considered negligible. Thresholds for contaminant concentrations in drinking water bottled in recycled PET were determined to be 0.017, 0.028 and 0.15 µg/kg food for infants, toddlers and adults, respectively. These numbers are based on the threshold of 0.15 µg/person/day (d) for substances with structural alerts for genotoxicity and on exposure data. Furthermore, it is assumed that under foreseeable conditions high-potency genotoxic compounds which are excluded from the TTC approach enter the recycling stream only sporadically and react to harmless chemicals under recycling conditions.

3.2 Further materials

Besides PET recycling, many other recycling routes were investigated with respect to their technical and also economic feasibility in the last years. Here, we list some recent illustrative examples of processes. However, this list is not comprehensive.

Polyolefins

In general polyolefins belong to the recyclable polymers. Regarding food packaging, the number of registered recycling processes for polyolefins is much lower than for PET [18, 23].

- In 2012, Marino Xanthos published a review paper on the recycling of polypropylene (PP) [24]. He states that recycling of PP food containers to food-contact grade packaging is currently not feasible due to the high quality standards, but many efforts are undertaken to achieve them. The author further states that the quality of PP declines with each round of recycling, because PP is especially sensitive towards photooxidative and thermooxidative degradation during use and recycling. This degradation can be overcome by the addition of blends of processing stabilizers.
- Achilias and colleagues investigated the recycling of PE, PP and HDPE by dissolution/reprecipitation technique and pyrolysis [14]. The first method requires high amounts of solvent, but the recovery and quality of the resulting polymer is high. Pyrolysis, on the other hand, produces mainly oil and gaseous fractions that can be used as new building blocks in the petrochemical industry.

Separation of multilayer materials into their constituents

A big part of the plastic used as FCM is integrated into multilayer materials that can also contain non-plastic components such as paper or aluminum.

- A recycling process for composite packaging materials like beverage cartons was investigated by Zhang et al. [25]. The researchers extracted the pulp in a precedent step, which was not described in detail, and optimized the separation of aluminum and three different types of polyethylene (PE) by solvent extraction with benzene-ethanol-water. The authors do not mention whether their recycled materials are also intended for use in food contact.
- A study by Favaro and colleagues showed the recyclability of multi-layer material composed of low-density polyethylene (LDPE), aluminum and PET in lab scale [26]. It was delaminated in acetone and subsequently PET was depolymerized into diethyl terephthalate and ethylene glycol.
- In 2013, Barlow and Morgan assessed the environmental impact of polymer film packaging for food and focused also on the recycling issue [2]. The authors pointed out technical difficulties prohibiting efficient film recycling from the domestic waste stream (e.g. multi-layer structures), inefficient identification of the material and contaminations arising from food.

(Nano-)Composites

The production of (nano-)composites, or blends is one way to improve the decreasing mechanical properties of recycled plastics.

- In 2012, Lopez et al. investigated the recyclability of three bioplastics based on polylactide, thermoplastic starch and an aliphatic polyester, which is fossil-fuel based, but biodegradable [27]. The scientists recycled the pure materials and cellulose-enforced polymers and compared their performances after several cycles. The recyclability of the pure materials was best for the aliphatic polyester, followed by polylactide and then thermoplastic starch. The incorporation of cellulosic fibers slightly enhanced the recyclability of thermoplastic starch, but it made the polylactide less stable and impossible to recycle. The fibers affected the stability of the polyester only slightly, but did not affect its recyclability.
- In 2014, Mitchell and colleagues described a process in which disposable cups composed of laminated cellulose were shredded into flakes and mixed with PP to produce plastic-paper composites [28]. The authors suggest using this material for products needing enhanced stiffness such as pallets, pipes and bins, but they do not comment on the recyclability of the new material or their application as FCMs.
- In 2013, Yasser Zare published a detailed review of nanocomposites made from recycled polymers. In the article, he analyzes processes for the production of nanocomposites based on e.g. PET, PP, HDPE, PVC and polystyrene (PS) [29]. The recycled plastics were mixed with nanoparticles such as nanoclay, calcium carbonate nanoparticles, carbon nanotubes and graphene. Whether these new materials could be suitable for the production of FCMs and whether they could be recycled several times was not further stated.

4 Market data

4.1 PET collection and recycling rates

In many countries high efforts were undertaken to increase the recycling rates of PET. In 2012, the National Association for PET Container Resources (NAPCOR) reported that in the U.S. 30.8% of the sold PET bottles were re-collected and 21.1% were efficiently recycled (Figure 2B) [30]. PETCORE Europe reported a collection rate of more than 52% for post-consumer PET bottles in 2012 in the EU corresponding to an actual volume of 1'640'000 tonnes of PET (Figure 2A) [31]. In the same year the recycling rate in Switzerland reached 81% corresponding to 37'571 tons of PET according to PET Recycling Switzerland [32]. In Japan, the PET recycling rates exceeded 80% in 2007 and have remained at this high level since then [33]. In 2012, 582'896 tons of PET were collected in Japan.

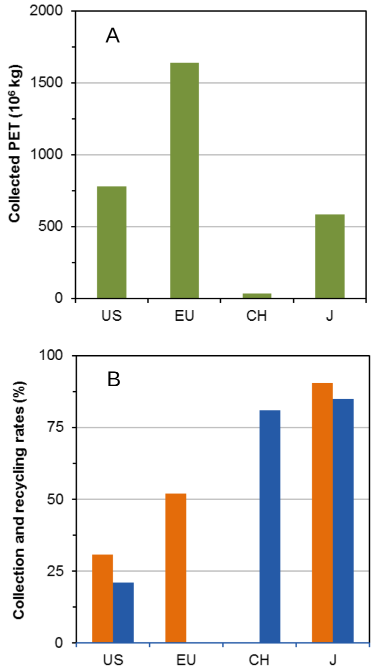

Figure 2. (A) Total volume of collected PET (green) and (B) PET collection (orange) and recycling (blue) rates in the U.S., EU, Switzerland and Japan, respectively, in 2012.

In 2012, the biggest European end-market for recycled PET was the fibers market, which was closely followed by the sheet and bottle markets [34].

4.2 Prices

The news provider *Plastics News* publishes regular updates of the prices for many different post-consumer resins. In general, transparent recycled materials are more expensive than colored ones and pellets are more expensive than flakes. A historical overview of the price development of PET and HDPE flakes is given in Figure 3 (data derived from [35]). These data refer to small batches; the prices for bigger batches were not published on this site, but can be expected to be lower.

The information provider *letsrecycle.com* published the price development of recycled plastic bottles composed of PET and HDPE for the year 2014 (Figure 4) [36]. These prices are significantly lower than the prices found on *Plastic News* (Figure 3), which might be due to e.g. different qualities of the materials included or the scale of the batches. In 2014 recycled PET was reported to be even slightly more expensive than virgin PET due to overproduction of virgin PET in the U.S. [37].

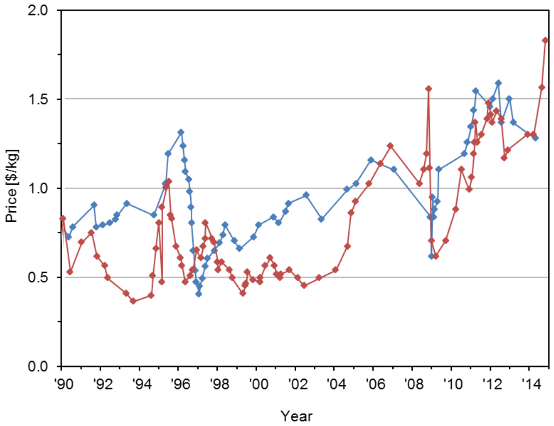

Figure 3. Historical development of resin prices for clear flakes of post-consumer PET bottles (blue) and natural flakes of post-consumer HDPE (red) (Figure adapted from [35]).

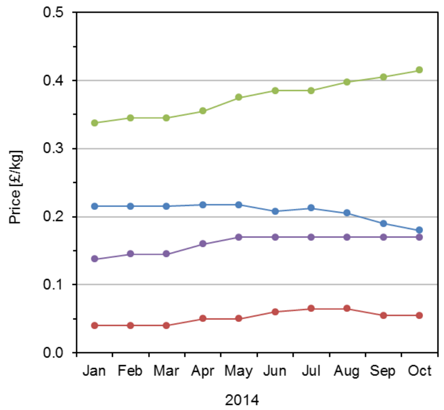

Figure 4. Prices for recycled plastic bottles composed of clear and light blue PET (blue), colored PET (red), natural HDPE (green) and mixed color HDPE (violet) (Figure adapted from [36]).

5 Regulatory Background

5.1 European Union

In the EU, recycled plastic materials and articles intended to come into contact with foods are regulated under Commission Regulation (EC) No 282/2008, commonly referred to as Recycling Regulation [38]. Article 4 of the regulation specifies that all articles and plastic materials used for recycling must have been produced in accordance with Community legislation on plastic FCMs and articles. Further, contaminations have to be reduced to a content that does not pose a danger to human health according to article 3 of Regulation (EC) No 1935/2004 [39].

Before recycling processes can be authorized by the European Commission, an application for authorization has to be submitted to EFSA. Based on EFSA's scientific opinion, the Commission then decides on the authorization. In 2008, guidelines for the technical and administrative requirements for the safety evaluation of such a recycling process were published [21]. More than 80% of the present applications describe processes for the recycling of PET. Due to the high number of applications for PET recycling processes an opinion on the criteria for their safety evaluations was published separately in 2011 [20].

EFSA assigns a process recycling number with the format RECYCxxx to each application. Currently, the recycling processes with valid applications are registered as part of the initial authorization process according to article 13 of Commission Regulation (EC) No 282/2008 [19]. The status of each application can be retrieved from EFSA's register of questions by entering the process recycling number or the corresponding EFSA question number into the keyword field. According to these numbers, 127 recycling process have been registered and partially evaluated by EFSA. None of the evaluated recycling processes has been authorized by the European Commission to this point (October 30, 2014) [23]. In future a register of authorized regulation processes shall be established under article 9 of the recycling regulation.

5.2 United States

The use of recycled plastic in the manufacturing of food contact articles is evaluated on a case-by-case basis by the FDA [40]. In 2006, the FDA published a guidance document for industry with non-binding recommendations needed for evaluating the suitability of a plastic recycling process [22]. FDA's safety concerns include possible contaminations arising from post-consumer materials (e.g. cleaning agents, household chemicals, paints), the incorporation of plastic materials not fulfilling the requirements for food-contact use and the use of unsuitable adjuvants, which might not comply with the regulations.

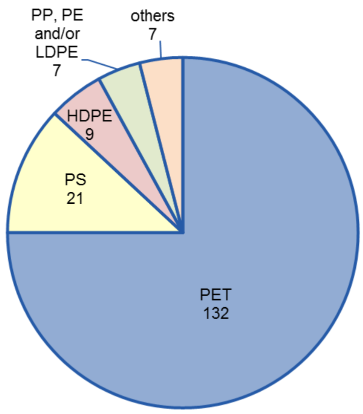

Figure 5. FDA-evaluated processes for post-consumer recycled (PCR) plastics for food-contact articles [18].

Between 1990 and 2014, 176 processes for producing post-consumer recycled (PCR) plastic were judged suitable by the FDA [18]. On average seven recycling processes were evaluated annually, but the numbers show a slight upward trend. 75% of these processes describe the recycling of PET (Figure 5). 12, 5 and 4% of the processes deal with the recycling of PS, high-density polyethylene (HDPE), and PP, PE and/or LDPE, respectively.

5.3 Japan

In 1997 the Japanese Ministry of the Environment enforced the Container and Packaging Recycling Law with the aim to reduce the waste of glass, PET and paper [41]. Three years later the law was amended to include plastic containers and packages other than PET. In Tokyo, the Council for PET Bottle Recycling was established in 1993. It promotes the mechanical recycling of PET bottles.

6 Contaminations

6.1 Polymeric substances

The efficient separation of different polymers is a necessity to obtain recycled plastics of high quality. Recycling processes can become inoperable, if the main polymer is contaminated with other polymers. One prominent example is the contamination of a PET recycling stream with polylactide (PLA) bottles. If PLA enters the recycling stream by mistake, the whole material may become unusable [42]. The two materials cannot easily be distinguished by the consumer and an automated separation technology is very costly. On the other hand, the producers of polyfuranoate terephthalate (PEF), a biobased polymer that is envisaged to replace PET within the next years, has been tested to be compatible with PET during recycling [43].

6.2 Chemical contamination

As illustrated in Figure 1, several other sources of possible chemical contaminations exist. Here, some examples are listed describing the introduction of unwanted chemicals into recycled materials.

- The influence of plastic and paper recycling on the exposure to phthalates was investigated recently by Lee et al. [44]. The study showed that increased use of recycled food packaging can cause elevated childhood exposure to the anti-androgenic phthalates dibutylphthalate (DBP, CAS 84-74-2) and diisobutylphthalate (DiBP, CAS 84-69-5).
- In 2009, Cheng and colleagues published a study on the contamination of recycled PET with antimony [45]. The migration of antimony into the food strongly depended on the further treatment and storage conditions. Rinsing the bottles before filling generally reduced the antimony concentrations, whereas microwaving or heating significantly increased the levels.
- The levels of chromium, antimony, lead, nickel and chromium in 200 samples of post-consumer PET films and containers were determined by Whitt et al. [46]. 29 samples were contaminated with mixtures of heavy metals at concentrations below the California's Toxics in Packaging Prevention Act. No virgin PET samples were tested in this study.
- In 2013, Samsonek and Puype tested black FCMs purchased at the European market for the presence of brominated flame retardants [47]. Approximately 40% of the samples contained these brominated compounds, although they are not authorized for the use in FCM plastic and some are even banned under the Stockholm convention. The flame retardants probably originated from electric and electronic equipment waste that was fed into the recycling stream.

7 Environmental and health issues

Different aspects of plastic recycling may affect the environment and human health. Studies show that especially in developing countries, occupational health and safety may not be sufficiently ensured in the plastic recycling industry. Although recycling reduces the amount of waste, these processes can generate emissions that have an impact on the direct environment of the factory.

- Yorifuji and colleagues measured volatile organic compounds (VOCs) e.g. toluene, benzene, ethyl acetate and different alkanes in the vicinity of a plastic recycling plant and correlated them with mucocutaneous and respiratory symptoms of the surrounding residents [48]. The results implied possible associations of exposure to open-air VOCs with health effects including sore throat, eye itch, eye discharge, eczema and sputum.
- A preliminary, but demonstrative report on the possible environmental and health effects caused by the plastic reprocessing industry in Bangalore, India was published in 1997 [49]. In 2011, the use of recycled plastic for food packaging was prohibited by law [50].
- In 2011, a publication focused on the health conditions of workers employed at the plastic recycling industry in Dhaka, Bangladesh [51]. The occupational hazards included formation of dust and fumes, noise, unsafe machines, exposure to hazardous chemicals, heat, no ventilation and missing protective gear. Although these problems might also occur in other sectors of industry in many developing nations, it might be of special relevance due to the export of plastic waste from Europe and the U.S. to many Asian countries.
- In 2005, Björklund and Finnveden reviewed LCA studies comparing materials recycling to incineration and landfill [52]. For non-renewable materials including plastics they concluded that energy use and global warming potential are generally lower for recycling than for landfill or incineration.
- Another LCA study focused on the further recyclability of packaging produced from recycled material [53]. A recycled PET tray was compared to a multilayer tray and demonstrated environmental advantages mainly due to its end-of-life scenario.

Abbreviations

DBP	Dibutylphthalate
DiBP	Diisobutylphthalate
EFSA	European Food Safety Authority
FCM	Food Contact Material
FDA	U.S. Food and Drug Administration
HDPE	High-Density Polyethylene
LDPE	Low-Density Polyethylene
NAPCOR	National Association for PET Container Resources
PCR	Post-Consumer Recycled
PE	Polyethylene
PET	Polyethylene terephthalate
PP	Polypropylene
PS	Polystyrene
PVC	Polyvinylchloride
RIC	Resin Identification Code
SPI	Society of the Plastics Industry
TTC	Threshold of Toxicological Concern
VOC	Volatile Organic Compounds

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