

Volatile organic contaminants in HDPE milk bottles along the mechanical recycling value chain, revealing origins and contamination pathways

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ABSTRACT

To progress towards closed-loop recycling of plastic packages it is vital to understand the origin of contaminants and to develop effective mitigation strategies. High density polyethylene (HDPE) milk bottles were sampled at nine different locations along the recycling value chain and analysed with two gas chromatography – mass spectroscopy methods to study the presence of volatile organic compounds. The total approximated concentration of volatiles reduced over time from the production of the bottles up to the cross-docking facility for the separately collected lightweight packaging waste (from about 250 to 100 ppm), as alkanes and alkenes evaporate from the bottle. A clear maximum was observed in the number of identified volatiles and in the total approximated concentration in milk bottles at the sorted HDPE product. Here contaminants peaked that originate from the milk, from other packaging components (labels, caps, inlays), from other packages and from the surrounding atmosphere. When only the milk bottles were manually separated out from this sorted product and these were mechanically recycled, flakes were obtained that contain the least amount of volatile compounds, much less than the freshly produced bottles (about 25–50 ppm). The type of volatiles present was, however, markedly different. In the freshly produced bottles alkanes, alkenes and intentionally added anti-oxidant were found, whereas in the recycled flakes mostly contaminants were found that originated from: the milk, the other packaging components, other packages and the surrounding atmosphere. Seven different contamination pathways were discerned. The gathered knowledge facilitates the future development of food-grade recycled HDPE.

1. Introduction

Plastic packages offer substantial societal and economic benefits during their use and simultaneously contribute to greenhouse gas emissions and planetary pollution during their production and waste management (OECD, 2022; Schirmeister and Mulhaupt, 2022). To curb the environmental impacts of plastic packages recycling and reuse strategies are being implemented. Currently, the most common approach is mechanical recycling. Additionally, new chemical recycling facilities are erected and tested (Solis and Silveira, 2020; Quicker et al., 2022) and also various reuse systems are being tested (Moalem et al., 2023). The mechanical recycling of polyethylene terephthalate (PET) bottles into a food-grade recycled PET is well-established in Europe (Franz and Welle, 2004; Welle, 2011; Grant, 2022). The majority of the plastic packages are, however, based on polyethylene (PE) and polypropylene (PP) (Brouwer et al., 2018; Eriksen et al., 2018; Roosen et al., 2020). Only for the minute fraction of these packages that are kept in a closed loop, mechanical recycling processes have been developed, that

result in food-grade recycled plastics (EFSA panel for food contact materials, 2014 and 2015). For the vast majority of these packages, however, this is not the case. The main reason is that PE and PP based packages absorb more volatile contaminants (Dutra et al., 2014; Gerassimidou et al., 2023; Rung et al., 2023) than PET based packages due to the high diffusivity of molecules in PE and PP as compared to PET (Rung et al., 2023). Once absorbed these contaminants are also more challenging to remove by thermal desorption due to the lower melting temperatures of PE and PP compared to PET (Palkopoulou et al., 2016) and the hydrophobic nature of most contaminants which renders washing with aqueous solutions less effective (Palkopoulou et al., 2016; Demets et al., 2020; Roosen et al., 2022). Furthermore, the fourfold conservative assessment protocol of European food safety authority (EFSA) for feedstock-process combinations adds to the challenge (Franz and Welle, 2022). Finally, many PE and PP based packages have not been designed for recycling, yet (Brouwer and Thoden van Velzen, 2017; Roosen et al., 2020; Thoden van Velzen et al., 2020a; Thoden van Velzen et al., 2020b). Consequently, the vast majority of the plastic packages

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are currently open-loop recycled into non-food packages and non-packaging applications (Cimpan et al., 2021; Klotz et al., 2023).

Closed-loop recycling of PE and PP based packages can only be effectuated with knowledge of the contaminants that are present in recycled PE and PP, their origin and of mitigation strategies to reduce their presence to acceptable levels. Although, in the last decades substantial knowledge has already been gathered on these topics, these three knowledge gaps perpetuate. In large it is known that recycled plastics differ from virgin plastics in three aspects: the level of degradation of recycled plastics is larger, recycled plastics often contain particle contamination and recycled plastics usually contain higher and different levels of molecular contaminants (Vilaplana and Karlsson, 2008). The particle contaminants can relate to inorganic particles (e.g., pigments, sand, dust) (Shirzaei Sani et al., 2023), and also to polymeric contamination that results in blend formation (Gall et al., 2021; Cozzarini et al., 2023). The molecular contaminants relate to intentionally added substances (IAS) such as additives and non-intentionally added substances (NIAS) such as oligomers, absorbed volatiles, aroma compounds, etc. (Cabanés et al., 2020a,b; Horodytska et al., 2020). Not all contaminants are, however, easy to categorise, as degraded polymer fragments, oligomers, printing ink residues, adhesive residues, etc. will also be present. For example, Garofalo et al. found polar contaminants in recycled polyolefins and these are likely to be both particle contaminants and molecular contaminants (Garofalo et al., 2020, 2021).

Recycled post-consumer high density polyethylene (HDPE) produced from mono-collected British milk bottles are free from polymeric contamination (Gaduan et al., 2023), since the bottles have been designed for recycling and the recycling process effectively removes all non-targeted components. Whereas, recycled HDPE made from separately collected lightweight packaging waste (LWP) that has been sorted into sorted product "HDPE" according to the specification DKR 329 (Punkt, 2023) and has been mechanically recycled does contain polymeric contaminants (PP, low density polyethylene (LDPE), polystyrene (PS), etc.) (Thoden van Velzen et al., 2020a,b). These polymeric contaminants originate from sorting mistakes and other packaging components (labels, caps, etc.) and form blends resulting in a recycled PE with reduced mechanical properties (Luijsterburg and Goossens, 2014; Thoden van Velzen et al., 2020a,b; Gall et al., 2021; Karaagac et al., 2021). When Dutch milk bottles are manually sorted from the sorted product DKR 329 and separately recycled, the recycled PE is free from polymeric contaminants (as far as can be observed) and has mechanical properties that are close to those of virgin HDPE (Thoden van Velzen et al., 2020a,b).

Recycled post-consumer HDPE produced from only milk bottles contains several volatile organic compounds. Early gas-chromatography studies found: alkenes, alkanes, anti-oxidants, di-*t*-butylphenol (a degradation product of the anti-oxidant) and limonene (DeVlieghere et al., 1998; Welle, 2005). A more recent study by Su et al. with a more advanced solid-phase microextraction GC-MS based technology reports on recycled HDPE from milk bottles that were separately sorted by Spanish sorting facilities and found hundreds of volatile compounds (Su et al., 2021). Several of these compounds are of toxicological concern, for example: butylated-hydroxytoluene, diethyl phthalate, naphthalene, diisobutyl phthalate, dibutyl phthalate, 2-ethylhexyl-4-methoxy cinnamate, octocrylene, 1-dodecene, 1-tetradecene (Su et al., 2021). Many of these volatile compounds could be reduced by washing and decontamination, but the two NIAS that are related to sunscreen (2-ethylhexyl-4-methoxy cinnamate, octocrylene) could not and are of main concern when striving for food-grade HDPE from milk bottles (Su et al., 2021). Although the origin of these NIAS is rather obvious (sunscreen), it is unclear at which stage of the recycling value chain and along which contamination pathway these NIAS have absorbed in the HDPE resin of the milk bottles.

Furthermore, other researchers have studied the volatile organic compounds present in recycled post-consumer HDPE produced from sorted PE packages, hence a heterogeneous mixture of PE packages that

have previously been used to contain milk, juice, personal care products, home care products, detergents, etc. As expected these recycled HDPE's contain hundreds of volatile organic compounds, including IAS, polymer degradation products (alkanes and alkenes) and various NIAS (Strangl et al., 2018; Chen et al., 2020; Horodytska et al., 2020; Chen et al., 2021; Kato and Conte-Junior, 2021; Su et al., 2021; Zeng et al., 2023). Some NIAS are aroma compounds and are related to the products typically sold in PE bottles and the various degradation products are likely the result of the various grades of HDPE used for the different packages with various ages. The aroma compounds have likely originated from product residues. Unclear, however, is at which stage of the recycling value chain all these different volatile contaminants have been absorbed in the PE and to what extent these volatiles have been exchanged between bottles. This lack of knowledge thwarts the development of effective mitigation strategies.

To progress towards a more circular economy for plastic packages, besides the existing recycling processes for PET packages, also recycling processes for PE and PP packages must be developed that deliver food-grade recycled plastics. As a first step, it is crucial to understand what quality of recycled PE/PP mechanical recycling can ultimately deliver. Therefore, the most common polyolefin based package in Dutch LWP was chosen to study. This 2 L HDPE bottle for pasteurised milk (also called jug) is designed for recycling, hence minimising the risk of self-contamination. Since all non-targeted components (label, cap, inlay) can completely be removed in the recycling process, the risk of NIAS contamination from these components is minimised. Photos of these milk bottles are added to the supplementary material, figure S2. This HDPE milk bottle is a very homogeneous packaging type. Nearly 90% of this bottle is produced in one factory with the same grade of HDPE, having the same dimensions, only having different embossed details in the bottle body wall and different labels. This bottle is composed of four components: a transparent HDPE bottle body, a light blue low density polyethylene (LDPE) cap with closure ring, a white inlay made from PE-based foam and a LDPE-based label. Most brands make use of a loose shrink LDPE-label, only one brand has a self-adhesive label with a water-soluble adhesive. All these non-targeted components can be removed during the mechanical recycling process efficiently with air classification, washing, near infrared and colour sorting. The objective of this study is to understand at which stage of the recycling value chain volatile organic contaminants are absorbed into either the milk bottle or the recycled HDPE, along which contamination pathways these contaminants enter the HDPE, and to what extent conventional mechanical recycling processes can remove these contaminants. This study is not a food safety study, its aim is to analyse the development of volatiles in the HDPE milk body bodies during the recycling value chain and to gain knowledge of the underlying contamination pathways.

2. Materials and methods

2.1. Sample origin, preparation and chemicals used

In total 61 HDPE bottle bodies, 16 labels, 17 caps and 7 inlays were collected from 7 stages of the recycling value chain. Additionally, 23 samples of washed flakes made from bottle bodies with different mechanical recycling processes were collected. The various stages in the recycling chain, the amount of samples taken from the four bottle components and the replication levels are listed in Table 1. At three stages the sampling was more complex and also sub-stages were discerned. At stage G (sorting) bottle bodies sorted from separately collected LWP (G-SC) were compared with bottle bodies from mechanically recovered LWP (G-MR). Stage H relates to mechanically recycled flakes made from a concentrate of 65% pure milk bottles (and hence 35% other HDPE bottles). This concentrate was previously prepared from sorted product DKR 329 which was subjected to automatic robot-sorting at NTCP (NTCP, 2023). This sorting robot had been trained to recognise milk bottles with artificial intelligence. Two sub-stages H were

Table 1

Samples taken from the four HDPE bottle components along the nine stages of the recycling value chain, replicates are indicated with the x symbol, hence 7x2 implies 7 samples that were measured twice and 2x1+2x2 implies two samples that were measured once and two samples that were measured twice.

Stage	Description	Number of samples
A	Freshly produced, bottle bottles from the bottling company (FarmDairy, Lelystad) just after the stretch blow-moulding machine, labels, caps and inlays from the warehouse. Hence these bottles had not been filled with milk.	7x2 bodies, 4x1 caps, 4x1 labels, 2x1 inlays
B	Filled by the bottling company and emptied within 8 h	5x2 bodies, 2x2 caps, 2x1 labels, 2x1 inlays
C	Emptied by households after regular use, washed with cold tap water three times, dried at ambient temperatures and kept separate	9x2 bodies, 3+2x1 caps, 3+2x1 labels
D	Spoilage test: bottles with about 100 ml of fresh milk kept for 2 weeks at room temperature	4x2 bodies
E (+)	A: Bottles retrieved from separately collected LWP B: Clean bottle bodies exposed for 4 weeks in the summer to mixed LWP	4x2 bodies A, 4x2 bodies B
F	Milk bottles retrieved from freshly collected LWP at the cross-docking facility for LWP from ROVA in Zwolle	11x2 bodies, 2x1+2x2 caps, 2x1+2x2 labels, 4x1 inlays
G-SC	Retrieved from sorted product DKR 329 (HDPE) from two sorting facilities:	8x2 bodies SC, 9x2 bodies MR
G-MR	SC: PreZero Zwolle processing separately collected LWP MR: KSI Heerenveen processing mechanical recovered LWP	
H-M	Mechanical recycled milk bottle concentrate produced from DKR 329 with robot sorting delivering a 65% pure sorted product of HDPE milk bottles and hence 35% other PE bottles in two steps: M after milling and CW after milling and cold water wash	4x2 samples of body flakes M, 4x2 samples CW
H-CW		
I-M	Mechanical recycled milk bottles that were manually sorted from DKR 329 with a 100% purity in three steps: M after milling, CW after milling and cold water wash, HW after milling and hot water wash with lye	5x1 samples of body flakes M, 5x1 samples CW, 5x1 samples HW
I-CW		
I-HW		

+ : Samples of E-A and E-B were combined after a student t-test proved they were significantly equal with a confidence level of 95%.

distinguished: milled and unwashed flakes (H-M) and milled and cold water washed flakes (H-CW). Stage I relates to the mechanical recycling of 100% pure milk bottles that were manually sorted from sorted product DKR 329. The labels, caps, inlays were all manually removed prior to milling. Samples for sub-stage I-M were taken from the unwashed milled goods. Samples for sub-stage I-CW were milled flakes that were washed with cold tap water for 5 min and then dried overnight at 85°C. Samples for sub-stage I-HW were milled flakes that were first pre-washed with cold tap water for 5 min then washed at 85°C with a 0.01 M sodium hydroxide solution for 5 min, rinsed abundantly with cold tap water and then dried overnight at 85°C.

All samples of bodies, labels, caps and inlays, except the freshly produced bottle components from stage A, were first brushed clean to remove surface dirt, washed with cold water (without surfactants) and carefully dried at room temperature overnight. The bottle bodies were subsequently milled separately in a previously cleaned Wanner compact granulator 17.26SV with an 8 mm sieve plate. A picture of the flakes is added as figure S6. The labels, inlays and caps were cut in small pieces (<1 × 1 cm) with a clean pair of scissors. All the milled/cut samples were stored in aluminium/PE pouches.

The following chemicals were used: dichloromethane (CAS 75-09-2) of Sigma-Aldrich, analytical grade (ISO ≥99.9% GC) and 1-methylnaphthalene (CAS 90-12-0) of Merck, for synthesis grade (>95% GC).

2.2. Gas chromatography with mass spectroscopy of extracted samples

2 g of bottle body flakes were weighted on an analytical balance with 0.1 mg precision, transferred in a 25 ml glass vial with screw cap and 15 ml dichloromethane was added. The vials were slowly shaken during 72 h at room temperature as this renders near-complete extraction of volatiles, see Part V of the supplementary material. 1 ml of the extract was transferred to a glass vial, closed and placed in the autosampler. The extraction procedure for the caps, labels and inlays was similar, but with less material: only 1 g of material was used and 7.5 ml of dichloromethane.

The extracts in the glass vials were analysed with a Trace1300 gas chromatograph fitted with an AS3000 autosampler, a programmable temperature vaporising (PTV) injector and a ISQ7000 mass spectrometer. The applied column was a Restek rx-5SIL-MS of 25 m length, 0.2 mm internal diameter and 0.33 µm thick internal coating. The carrier gas was hydrogen with a flow rate of 2.0 ml/min. The injected volume was 1.0 µL. The PTV injector was programmed for 3 s at 50°C, increasing to 350°C with 5 °C/s, holding at 350°C for 1 min. Subsequently the

injector was cleaned at 450°C for 5 min with a split flow of 100 ml/min, followed by cooling to 50°C with a split flow of 5.0 ml/min. The column-oven was kept for 2 min at 50°C after injection, after which the temperature was increased to 350°C with a 20 °C/min rate and this final temperature was maintained for 2 min. The column was directly connected to the mass spectrometer. The transfer temperature was 320°C. The ion source temperature was 300°C. Ions were counted in the range of 35–800 amu with a 0.10 s scan time. The total ion count (TIC) was registered in relation to the retention time. Of all peaks with a peak size exceeding 1 million TIC the fragmentation patterns were analysed with the NIST library to tentatively identify the substance.

This method was calibrated with 1-methylnaphthalene as external standard, see supplementary material part I, resulting in a level of detection of 0.3 µg/L and a level of quantification of 0.9 µg/L. The response to this external standard was used to semi-quantify the TIC's of all other substances.

2.3. Headspace gas chromatography with mass spectroscopy

Roughly 1 g of bottle body flakes were weighted on an analytical balance with 0.1 mg precision and transferred into a 21 ml glass vial with a septum and a screw cap. Of samples of the labels and caps similar weights were used. Only of the inlays less material was available and roughly 0.25 g was weighed in.

The headspace composition in the vials was analysed with Trace 1300 gas chromatograph fitted with a TriPlusRSH autosampler, a CryoFocus cryogenic trap, a split/splitless (SSL) injector and a TSQ7000 mass spectrometer. The applied column was a Restek rxi-5SIL-MS of 30 m length, 0.25 mm internal diameter and 1.0 µm thick internal coating. The carrier gas was hydrogen with a flow rate of 2.17 ml/min. Prior to injection, the glass vial with the sample was heated to 120°C for 10 min and lightly shaken. Subsequently, a 1.5 ml sample of the headspace was taken with a 2.5 ml preheated glass syringe (120°C) and injected in the cryotrap, which was precooled to –120°C prior to injection. After injection, the cryotrap was kept at –120°C for 180 s and then the temperature was increased to 250°C with a heating rate of 50 °C/s, after which the cryotrap was held for 113 s at 250°C. The SSL injector was operated at 250°C for 6 min in splitless mode, after which it was changed in split mode with a flow of 22 ml/min in a 1:10 ratio. The column-oven was held for 5 min at 40°C after injection and then increased to 250°C with a heating rate of 30 °C/min after which the final temperature was maintained for 1 min. The column was directly connected to the mass spectrometer. The transfer temperature was 260°C. The ion source

temperature was 290°C. Ions were counted in the range of 35–400 amu with a 0.10 s scan time. The total ion count (TIC) was registered in relation to the retention time. Of all peaks with a peak size exceeding 1 million TIC the fragmentation patterns were analysed with the NIST library to tentatively identify the substance.

This method was calibrated with 1-methylnaphthalene as external standard, see supplementary material part II, resulting in a level of detection of 2 µg/g and a level of quantification of 6 µg/g. The response to the 1-methylnaphthalene external standard was used to semi-quantify the TIC's of all other substances.

2.4. Statistical processing

The measured data was processed to extract the number of registered volatiles per sample and the total approximated concentration per category of volatiles. Data of replicates were first averaged, then the data of all independent samples (including the replicate averages) were grouped per stage. Datasets of independent samples at the same stages of the recycling value chain were analysed statistically, by calculating mean values, standard deviations, skewness and kurtosis. Next the Kolmogorov-Smirnov test was conducted to test for the normality of the distribution. Comparisons between datasets with normal distributions were executed with heteroscedastic t-tests assuming unequal variances with a confidence level of 95%. Comparisons between datasets of which at least one was not normally distributed were conducted with the two-tailed Mann-Whitney *U* test with a confidence level of 95%.

Furthermore the data for the hundreds of volatiles per sample was categorised in groups of similar molecules, see the supplementary materials Part III for extracted volatiles and Part IV for headspace volatiles.

3. Results

The crude results with all tentatively identified compounds in the samples are listed in a data repository comprised of four databases DB1-DB4 (Teunissen et al., 2024).

3.1. Extraction GC-MS data

The number of volatiles extracted from the bottle bodies at the various stages of the recycling value chain is graphically shown in Fig. 1. The total approximated concentration of extracted volatiles is presented in Fig. 2 and the average concentration of the various types of extracted volatile compounds is listed in Table 2. At a first glance there seem to be

three phases in Figs. 1 and 2: a gradual reduction in extracted volatiles from bottle production (stage A) up to the cross-docking facility for LWP waste (stage F), followed by a peak in extracted volatiles at the sorting plant (stage G) and finally a reduction in extracted volatiles during mechanical recycling (stages H and I). Within the class of alkanes, a distinction is made in Table 2 between the even straight alkanes, of which most can be attributed to the HDPE resin itself (Bradley and Coulier, 2007; Mekap et al., 2013; Schweighuber et al., 2021), and un-even and branched alkanes might have other origins such as cross contamination or resin degradation.

From Fig. 1 it is clear that the number of extracted volatiles remains roughly stable from the production (A) up to the cross docking station (F) with on average less than 20 compounds found. Simultaneously, Fig. 2 reveals that the total approximated concentration of extracted volatiles gradually decreases, from 260 ± 60 ppm at stage A to 100 ± 40 ppm at stage F. This reduction is caused by a reduction in the concentration of alkanes, alkenes and the intentionally added anti-oxidant (Irgafos 168) as is apparent from Table 2. The reduction in alkanes and alkenes could in principle be attributed to evaporation and migration processes. The evaporation process appears dominant since this reduction is fairly constant over the recycling value chain between A and F and is not limited to the early stage when the bottle is still filled with milk (A to C). As previously reported for other PE objects with the same anti-oxidant (Garrido-Lopez et al., 2007; Moreta and Tena, 2015), the reduction of the anti-oxidant in the HDPE milk bottle was expected, as this IAS reacts with oxygen.

The number of extracted volatiles and the total approximated concentration of extracted volatiles crest at sorting (stage G). Furthermore, milk bottle bodies in sorted products made from mechanically recovered plastic waste (G-MR) contain more extracted volatiles than the milk bottle bodies in sorted products made from separately collected LWP (G-SC) as the average amounts were 33 ± 15 for G-MR and 20 ± 6 for G-SC, see Fig. 1. The median approximated total concentration of extracted volatiles is larger for G-MR (217 ppm) than for G-SC (146 ppm), and the difference in the average values is even more pronounced: 500 ± 800 ppm for G-MR versus 140 ± 60 ppm for G-SC, see Fig. 2 and table S7. This indicates differences in the distribution of these concentrations at this stage in the recycling chain between G-MR and G-SC. This is further analysed in section 3.3. This spike in extracted volatiles can both be attributed to a rise in alkanes, alkenes and in NIAS such as fatty acids (originating from milk), aromatic hydrocarbons (AHC), plasticisers, fatty acid esters, fatty acid amides and fatty acid nitriles. The presence of NIAS will be discussed further in section 3.4. This apex in extractable

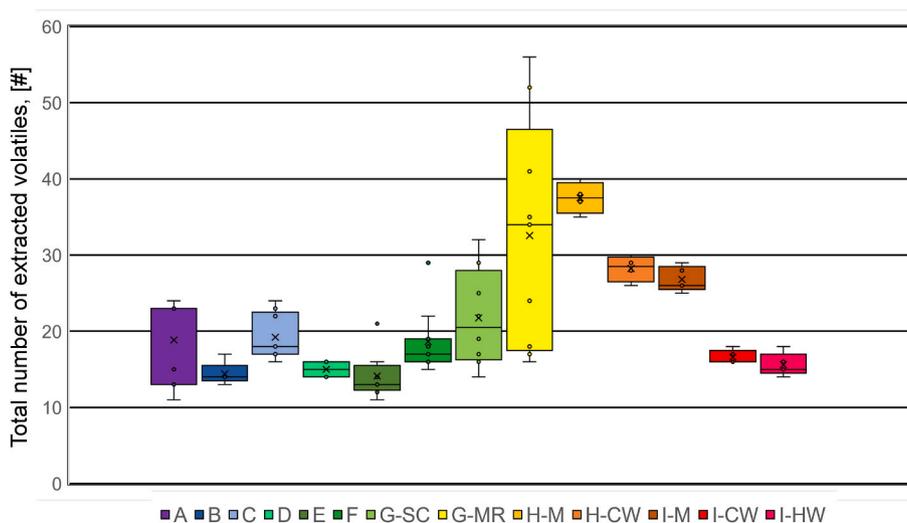


Fig. 1. Boxplot of the number of extracted volatile compounds from milk bottle bodies along the recycling value chain (stages A to I-HW), with symbol X as average and small circles for the individual data points.

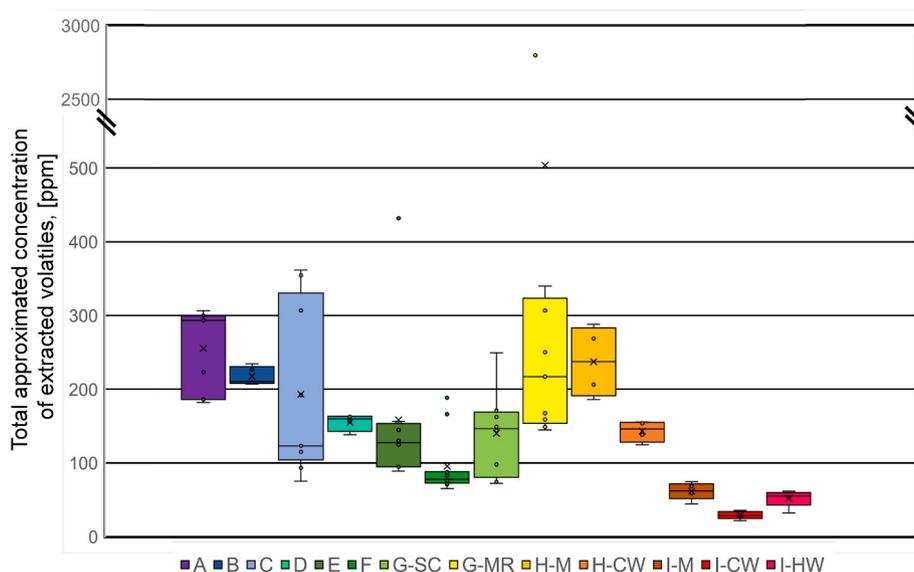


Fig. 2. Boxplot of the total approximated concentration of extracted volatiles along the recycling value chain (stages A to I-HW) in ppm, with symbol X as average and small circles for the individual data points.

Table 2

Averaged total approximated concentrations of the main type of extracted volatiles from the bottle bodies along the recycling value chain (A to I-HW), in ppm.

Type of extracted volatiles	Stage of the recycling value chain												
	A	B	C	D	E	F	G-SC	G-MR	H-M	H-CW	I-M	I-CW	I-HW
Alkanes, even & straight	63 ± 44	27 ± 4	42 ± 21	16 ± 2	30 ± 51	19 ± 5	36 ± 37	32 ± 27	100 ± 12	62 ± 7	13 ± 3	4 ± 2	7 ± 3
Alkanes, uneven & branched	0.3 ± 0.6	0.5 ± 1.1	0.7 ± 1.0	0	10 ± 8	2 ± 6	29 ± 46	19 ± 16	44 ± 30	6 ± 5	5 ± 3	0.5 ± 0.4	0
Alkenes	107 ± 46	137 ± 12	79 ± 58	99 ± 6	53 ± 29	44 ± 20	33 ± 25	129 ± 87	55 ± 10	59 ± 3	8 ± 2	3 ± 1	5 ± 3
IAS (anti-oxidant)	62 ± 52	11 ± 7	42 ± 54	27 ± 5	29 ± 35	7 ± 4	22 ± 14	28 ± 46	3 ± 2	6 ± 1	7 ± 3	10 ± 3	19 ± 5
NIAS fatty acids	0	0	0	0	15 ± 35	1 ± 4	7 ± 14	46 ± 120	8 ± 9	0	13 ± 6	0	0
NIAS degraded & used IAS	21 ± 17	41 ± 9	27 ± 19	12 ± 1	18 ± 10	21 ± 15	7 ± 4	43 ± 30	23 ± 1	6 ± 10	7 ± 2	9 ± 2	19 ± 4
NIAS AHC	0.2 ± 0.6	0	0.6 ± 0.6	0	0	1.0 ± 1.5	0.1 ± 0.3	1.0 ± 1.0	0	0	6 ± 1	2 ± 1	2 ± 1
NIAS plasticisers	1.4 ± 1.4	0	1.4 ± 1.7	1.4 ± 1.6	1.3 ± 1.9	0.4 ± 0.6	3 ± 4	6 ± 11	1.0 ± 0.2	1.0 ± 0.3	0 ± 0.1	0	0
NIAS fatty acid esters	0	0	0	0	0	0.4 ± 1.1	0	200 ± 600	1 ± 2	0	0	0	0
NIAS fatty acid amides	0	0	0	0	0	0	2 ± 5	0	0	0	0	0	0
NIAS fatty acid nitriles	0	0	0	0	0.4 ± 1.1	0	0.0	0.9 ± 1.4	0	1 ± 2	0	0	0

volatiles in bottle bodies is attributed to cross-contamination processes among the packaging components and between the milk bottle bodies and other packages/objects.

During mechanical recycling stages (H and I) the amount and concentration of extracted volatiles reduce again. The level of reduction varies strongly with the purity of the recycling feedstock. In case a PE feedstock of 65% milk bottles is used the number of extractables and the approximated total concentration is higher than in case 100% pure milk bottle body feedstock is used; stage H versus I, respectively. After milling the flakes made from 65% pure milk bottle feedstock contain 38 ± 2 extractable volatiles with a total average concentration of 240 ± 50 ppm (H-M), whereas the flakes made from 100% pure milk bottle bodies only contain 27 ± 2 extractable volatiles with a total concentration of 62 ± 11 ppm (I-M). For the cold water washed flakes made from 65% pure feedstock (H-CW) the amount of extractable volatiles and their total average concentration are both reduced as compared to the 65% pure feedstock (H-M). In these H-CW flakes 28 ± 2 extractable volatiles were

found with an average total concentration of 143 ± 15 ppm (H-CW). Cold-water washing the flakes made from pure milk bottles (I-CW) reduced the number of the extracted volatiles to 17 ± 1 with a total average concentration of 29 ± 5 ppm (I-CW), see database 1 (Teunissen et al., 2024). The large difference between the samples H-CW and I-CW indicate that cross-contamination occurs when the feedstock contains a mixture of bottles, and this cross-contamination can be avoided by using a pure milk bottle feedstock. Remarkably, the number of extractable volatiles in these flakes (I-CW) is lower than the number in the freshly produced milk bottles (A), which can be attributed to a large reduction in alkanes and alkenes and small increase in AHC, see Table 2. In case the pure milk bottle body flakes are washed with hot water instead of cold water, the number of extractable volatiles remains constant at about 16 and the total average concentration rises from 29 ± 5 ppm (I-CW) to 52 ± 12 ppm (I-HW). A part of this increase can be related to higher levels of even alkanes and even 1-alkenes, but also by raised levels of the intentionally added antioxidant Irgafos 168. The rise in the

level of antioxidant is most likely a methodical artefact, as the thermal treatment (hot water washing) has temporarily raised the low diffusion coefficient of the large antioxidant molecule in the thick flakes making it more accessible for extraction. The raised levels of even alkanes and even 1-alkenes could indicate that the HDPE gets more degraded by the hot water washing with lye, as has been suggested for LDPE based on tensile properties (Boz Noyan et al., 2022).

3.2. Headspace GC-MS data

The development of the volatiles measured with headspace GC-MS along the recycling value chain is relatively similar to the development of the extracted volatiles, see figure S3 for the number of headspace compounds and figure S4 for the approximated total headspace concentration. Again, there is gradual decrease in the total concentration of volatiles between stages A and F, a steep incline at the sorted product (stage G) and a drop to minimal levels for the recycled flakes made from 100% milk bottles (stage I). The average approximated concentrations of headspace volatiles is shown in Table 3 per category of compounds. The NIAS that the headspace GC-MS reveals are more volatile than those determined with extraction GC-MS and include aroma compounds (limonene, eucalyptol, isopulegol, myrtenol), aldehydes, various alcohols and one AHC, see table S4. From Table 3 it is clear that the massive increase in headspace volatiles at the sorting stage is due to the uptake of alkanes and aroma compounds due to cross contamination with other packages/objects.

3.3. Statistical assessment

The statistical assessment of the datasets of the total approximated concentrations of extracted volatiles (from A to I-HW) is summarised in table S7. Most datasets are normally distributed, with the exception of G-MR. The prevalence of the normal distribution for most datasets was expected since in most datasets the systemic contaminants (contaminants that originate from the resin itself, IAS, and previous use) dominate. Only in datasets where incidental contaminants (occasionally occurring cross-contaminants) are relevant, the distributions are non-parametric and skewed to the right with occasionally thick tails.

The total approximated concentration of extracted volatiles in HDPE milk bottle bodies/flakes doesn't differ significantly for most single steps in the recycling value chain up to stage G-SC (sorting) with 95% confidence, as is listed in Table 5. The concentration of extracted volatiles in the bottles at the consumer (C) remains fairly constant in the LWP (E), and in the milled goods made from 65% pure milk bottles (H-M), as the losses of some extractable volatiles are roughly compensated by the uptake of others. Only if larger steps in the recycling value chain are considered, such as from stage A to F or from C to F, the concentration is significantly different. After sorting (stages H and I) the concentration in

the bottle flakes is significantly different from previous stages, with only stage H-M as exception.

Furthermore, the analysis also clearly shows that the recycled flakes (I-HW) contain less extracted volatiles than the original bottles (A) and also in comparison to the bottles at the households (C). Additionally, the median concentration of extracted volatiles in milk bottles from mechanically recovered sorted products (G-MR) is significantly larger than from separately collected sorted products (G-SC).

The statistical assessment of the datasets of the total approximated concentration of headspace volatiles is summarised in table S8. All these datasets are normally distributed, with the exception of the dataset F. Most datasets between stage A and G-SC do not differ significantly with 95% confidence in pairwise comparisons of the total approximated headspace concentration, see table S9. The only exception is between stage B and C where a large stepwise reduction in headspace volatiles is noted. The datasets for the two sorted products G-SC and G-MR do differ significantly and also all further datasets up to I-CW do differ significantly in pairwise comparisons.

3.4. Development of volatile contaminants along the recycling value chain

The development of the detection rates of groups of volatile contaminants over the recycling value chain reveals when these groups of NIAS absorb in the bottle bodies (Table S5 and S6) and hence render clues on the involved contamination pathways. Other inklings are given by the share of even straight alkanes of all extracted alkanes in figure S5. This will be discussed for each stage in the recycling value chain.

Stage A. Unfortunately, the results for the freshly produced bottles (Stage A) are less reliable, as the bottles were stored for several weeks in boxes together with freshly printed labels, caps and inlays prior to the analysis. Hence the plasticiser (dibutyl decanedioic acid, CAS 109-43-3) that was found in these freshly produced bottles is likely to originate from exchange with the printed labels as this plasticiser is present in these labels, see database DS1 and DS2 (Teunissen et al., 2024).

Stage B. The bottles that were filled with milk at the production plant and directly emptied (stage B) do not yet show milk related compounds (fatty acids, aldehydes, limonene), because the exposure time was too short and the milk was still fresh.

Stage C. The bottles retrieved from the households after use (stage C) contain much more types of NIAS. Again the plasticiser is found that originates from the labels. Apparently after the shelf-life of the milk (14 days), the migration of the dibutyl-decanedioic acid plasticiser has progressed sufficiently to be well detectable in the bottle. Furthermore, aldehydes, fragrant and AHC molecules become detectable at this stage. The aldehyde detected at this stage is E-2-hexadecenal, which is a cellular metabolite (Semenkova et al., 2023). The detected fragrant molecules are mostly eucalyptol, isopulegol and to a lesser extent also limonene. Eucalyptol is widely used as food flavour (Pandey and Kim,

Table 3

Averaged total approximated concentrations of the various types of headspace volatiles emitted by bottle bodies along the recycling value chain (A to I-HW), measured with headspace GC-MS in ppm.

Type of volatile compound	Stage of the recycling value chain													
	A	B	C	D	E	F	G-SC	G-MR	H-M	H-CW	I-M	I-CW	I-HW	
Alkanes	18 ± 10	16.0 ± 0.5	8 ± 3	11 ± 3	9 ± 2	8 ± 2	8 ± 2	24 ± 8	42 ± 5	35 ± 2	4.0 ± 0.1	5.9 ± 0.4	5.9 ± 0.8	
Alkenes	7 ± 5	10.3 ± 0.5	1.7 ± 1.4	4 ± 1	3 ± 2	4 ± 5	2 ± 1	5 ± 2	7 ± 1	6 ± 1	0.5 ± 0.1	1.3 ± 0.1	1.5 ± 0.1	
NIAS Aldehydes	0	0	0.3 ± 0.3	0.4 ± 0.7	0.1 ± 0.1	0.6 ± 0.3	0.4 ± 0.2	3 ± 2	2 ± 1	2.2 ± 0.1	0.3 ± 0.1	0.4 ± 0.1	0.4 ± 0.1	
NIAS Aroma compounds	0.1 ± 0.2	0	0.3 ± 0.2	0	0	0.5 ± 0.4	0.2 ± 0.1	6 ± 6	12 ± 1	12 ± 1	0.1 ± 0.1	0.2 ± 0.1	0.2 ± 0.1	
NIAS Alcohols	0.5 ± 0.4	0.7 ± 0.1	0.7 ± 0.8	0.4 ± 0.2	0.2 ± 0.2	0.4 ± 0.1	0.4 ± 0.2	0.6 ± 0.5	1.0 ± 0.2	1.0 ± 0.4	0.3 ± 0.1	0.7 ± 0.1	0.8 ± 0.1	
NIAS AHC	0	0	0	0	0	0	0.1 ± 0.1	0.1 ± 0.1	0.3 ± 0.2	0.1 ± 0.1	0.6 ± 0.1	0	0	

2011), isopulegol in personal care products (Bhatia et al., 2008), which suggests cross-contamination in the households. As the households did not abuse these packages and kept them apart, this indicates that these volatile fragrant molecules have exchanged through the domestic atmosphere. The only detected AHC which was present in roughly half of these bottles was 4-methyl-1,2-dihydronaphthalene, which must originate from the households as it starts to be observable from stage C on and is absent in the other packaging components and the milk. As with the fragrant molecules a likely contamination pathway of this AHC is cross contamination through the household atmosphere.

Stage D. Although, the bottles that contained rotting milk (stage D) clearly smelled when opened, only few volatiles could be detected by the GC-MS technologies, such as nonanal and the label-related plasticiser. Nonanal is a known degradation product from milk (Yue et al., 2015; Li et al., 2019). Apparently the malodorous compounds of rotting milk such as small amines and sulphur compounds are beyond the detection limits of the analytical methods. The absence of fatty acids suggests that micro-organisms have depleted this resource completely.

Stage E. The milk bottles that either were exposed to LWP for 4 weeks or retrieved from collected LWP (Stage E) contained the same plasticiser from the label material, but also nonanal and fatty acids (myristic acid, palmitic acid, oleic acid) that can be related to milk decay (Yue et al., 2015). Remarkably also a fatty acid nitrile (Z-docos-9-enitrile, CAS 73170-89-5) was found in the milk bottle, although at a low detection rate of 6%. As this NIAS was also found in the caps at relatively high concentrations and in the inlays at mediocre concentrations, the migration of this NIAS from the caps is the most likely contamination pathway. Possibly this nitrile is formed from the corresponding amide that is used as an anti-slip agent in the cap, see DB2 (Teunissen et al., 2024). As the thermal conversion of fatty acid amides in fatty acid nitriles by the elimination of water has previously been described (Zwierzykowski et al., 1981). Moreover, related fatty acid nitriles have recently also been found in recycled PP labels (Schlossnikl et al., 2024). Although the exchange of NIAS between the packaging components is apparent, the exchange of NIAS between different packages and objects in the LWP is still fairly limited at this stage.

Stage F. Milk bottles that were retrieved from the cross-docking station of LWP collection (Stage F) contained various NIAS; the label related plasticiser, milk-related aldehydes (nonanal, hexanal) and fatty acids, but also some NIAS that indicate cross-contamination with product residues of different packages. In all bottles limonene was detected and although this normally occurs in milk at low concentrations (Yue et al., 2015), here the concentrations are elevated, see DB1 (Teunissen et al., 2024), which suggests cross-contamination with for instance juices and citrus based detergents (Bacanli et al., 2018). In roughly half of the samples 4-methyl-1,2-dihydronaphthalene as AHC was detected, which suggests cross contamination with other packages. And finally in less than 20% of the milk bottles isopropyl myristate was found, a fatty acid ester used in personal care products (Specialchem, 2024) also suggesting cross contamination with other types of packages.

Stage G. Milk bottles that were retrieved from the sorted product DKR 329 (HDPE rigid) produced at two sorting facilities (Stage G) contained much more NIAS than bottles from the preceding stages. A steep increase is observed for all NIAS categories and in bottles retrieved from both type of sorted products; made from separately collected LWP (G-SC) and made from mechanically recovered LWP (G-MR). However, the amount of extracted volatiles in G-MR is significantly larger than for G-SC (see section 3.3).

The most detected aldehyde was nonanal and the most detected fatty acid was oleic acid. All detected aldehydes and fatty acids are likely to have originated from milk. The total approximated concentration of both NIAS types peaks at this stage in the recycling stage.

The most detected plasticiser is still the dibutyl-decanedioic acid that originates from the labels, see DB2 (Teunissen et al., 2024), but also

others are found with detection rates of about 25% or less: diethyl-phthalate, di-n-octyl-phthalate and bis(2-ethylhexyl)-phthalate. These must arise from cross-contamination with other packages. The most detected alcohol is 5-(hydroxymethyl)undecane that appears to originate from the bottles themselves. But in half of the samples bis(2-hydroxyethyl)adipate is found, which occurs in the labels, caps and inlays, see DB4 (Teunissen et al., 2024) and hence cross-contamination between the packaging components is the likely pathway.

The presence of other NIAS also indicates that cross-contamination between product residues of other packages and the milk bottles occurred during sorting. For instance the most detected fragrant was eucalyptol (used in food & detergents), the most detected AHC was 4-methyl-1,2-dihydronaphthalene and there are also relatively many un-even and branched alkanes present, see figure S5. Fatty alkyl esters (isopropyl myristate, octyl palmitate and octyl stearate) that are used in personal care products (Draelos, 2011) were detected in only a few milk bottles, but when detected their concentrations are usually substantial. Similarly, Z-13-docoseneamide was found in a few bottles at relatively high concentrations, as this anti-slip agent is present in the caps, migration from the cap to bottle body is most likely pathway.

In roughly a quarter of the retrieved milk bottles from the sorted products two uncommon fatty acid nitriles were found (Z-hexadec-9-enitrile and Z-docos-9-enitrile). These were also detected in the caps from stage A and B, although the concentration at stage B was clearly larger, see DS2 (Teunissen et al., 2024). The most likely pathway is migration between packaging components.

Stage H and I. The intention of mechanical recycling is to produce valuable secondary resources with reduced levels of volatile contaminants as they compromise its odour and safety. For the samples made from 65% milk bottles (H), this is hardly the case. For the samples made from 100% milk bottles (I) the amount of volatiles and the total approximated concentration clearly do reduce, but this mostly relate to a reduction in alkanes and alkenes. With respect to the NIAS present in these flakes, clear reductions for only the fatty acids are observed. Surprisingly, even an increase in AHC's in samples I-M (100% pure milk bottles) and not in samples H-M (65% milk bottles) was noticed, and since different mills were used this is likely to be caused by a contamination with lubricant from the cutting mill. Furthermore, these were quite different AHC's than previously detected in the recycling value chain, namely: pentyl-benzene, hexyl-benzene and heptyl-benzene. The main difference between the recycled PE flakes made with 65% pure milk bottles (H) and those made with 100% pure milk bottles (I) is that in general the former samples contain more NIAS than the latter, this is especially obvious for plasticisers, fragrant molecules and aldehydes. Most NIAS that are present in the milled flakes (H-M or I-M) cannot be washed out (H-CW or I-CW), the concentration of aldehydes, fragrant molecules, alcohols remain nearly constant. The only exception are the fatty acids as these can efficiently be removed by washing. Also hot washing of the flakes from 100% milk bottles (I-HW) does hardly reduce the concentration of most NIAS in comparison to cold water washed flakes (I-CW). The limited removal efficiency of contaminants from PE bottle flakes with various washing media has previously been reported by Roosen et al. they corroborate that carboxylic acids are removed fairly good with water whereas alkanes, aromatics and aldehydes are hardly removed (Roosen et al., 2022).

4. Discussion

4.1. Significance of the results

This study presents semi-quantified concentrations of volatiles in HDPE milk bottles and recycled flakes made thereof. Some of the reported concentrations in the databases are below the level of detection and are therefore non-reliable. This study was not intended to measure

accurate concentrations of all NIAS in HDPE at ppt levels. The aim of this study was to describe the development of the volatiles in the HDPE milk bottle along the value chain and since consistently the same analytical procedure has been followed with all samples, the relative differences between different datasets can be studied and these differences are significant (section 3.3).

4.2. Contamination pathways for volatiles in HDPE milk bottles

The results of this study suggest that there are five different main contamination processes and pathways relevant for HDPE milk bottles, namely: 1) degradation of IAS (in this case the anti-oxidant) and of HDPE itself to form degradation products, 2) migration of product residues (milk) and degradation products of these product residues into the bottle, 3) migration of IAS of other packaging components such as labels, caps and inlays to bottle bodies, 4) migration from different packages/objects to milk bottles (either as vapours or as liquids), 5) migration of volatile contaminants from the local environment to the bottle body. In Table 4 the contaminants found are classified according to these contamination pathways. Fig. 3 depicts the development of the volatile contaminants in the milk bottle along the recycling value chain and clarifies at which stage these contaminants absorb in the bottle.

There are two additional contamination pathways that lay beyond the scope of this study. The sixth pathway is consumer abuse, meaning that civilians use empty packages deliberately to store or mix hazardous chemicals prior to discarding it. Empirical research on consumer abuse is scarce, as it involves repetitive, laborious measurements. The only study we are aware of (from 2005) approximates an abuse incidence rate of 0.03–0.04% (Franz and Welle, 2022).

The seventh contamination pathway is the thermal conversion of the recycled flakes into pellets and subsequently in final articles (blown bottles, injection moulded objects, etc.). Impurities present in the flakes can thermally convert into NIAS, as has previously been described for the degradation of PVC in recycled PET to benzene (Thoden van Velzen et al., 2020a,b). Recently, Mayrhofer et al. reported the generation of mutagenic compounds during the thermal conversion of some types of recycled PE, PP and PS flakes (Mayrhofer et al., 2023) and Schlossnikl et al. reported the generation of various Cramer class II and III compounds after recycling PP labels (Schlossnikl et al., 2024).

Hitherto, the European commission distinguished only two contamination pathways in the directives for the production of food-grade recycled plastics; systematic contaminants from previous use and incidental contaminants by consumer misuse (European Commission, 2008, 2022). This study has shown that there are not two but seven contamination pathways and although we underscore the same

Table 4
Classification of volatile contaminants over the five contamination pathways and the stages involved.

Contamination pathway	Stages involved	Type of contaminants involved
1 Degradation of HDPE and IAS (anti-oxidant)	A-I	Even n-alkanes, alkenes, oxidised Irgafos 168, various phenols with t-butyl side groups
2 Migration of product residues and degradation products thereof	B-I	Fatty acids, aldehydes, limonene
3 Migration of IAS of the other packaging components to the bottle body	B-I	Dibutyl-decanedioic acid, (Z)-Hexadec-9-enenitrile and (Z)-Docos-9-enenitrile
4 Migration from different packages to milk bottles	E-I	Uneven and branched alkanes, fragrant molecules, other plasticisers, fatty acid esters, fatty acid amides, AHC
5 Migration of volatile compounds from the surrounding atmosphere to the milk bottle	C-I	AHC, fragrant molecules

conclusion, namely that decontamination after mechanical recycling is required, the more detailed analysis of contamination pathways does allow for more dedicated and direct mitigation strategies.

Other researchers have previously reported on a single contamination pathway for recycled plastics. Recently, Roosen et al. observed that the cross-contamination of volatile substances between flexible packages in a sorted bale hardly occurs (Roosen et al., 2023). As discarded flexible packages hardly contain liquid product residues, this is expected. Conversely, for HDPE milk bottles this is the most important contamination pathway. This reconfirms that the relevance of contamination pathways is strongly dependant of the type of packaging, its product residues and the position along the recycling value chain.

This is further underlined by the observation that only hydrophobic volatiles were found to migrate to the HDPE bottle bodies. For instance, the hydrophobic fatty acid nitriles were found to migrate from the labels to the bottle body, whereas the migration of the more hydrophilic ink-components (2,4-dimethylaniline and N,N'-diacetyl-ethylene-diamine) could not be detected.

The seven different pathways in this study describe the origin of the contaminants and facilitate the development of dedicated mitigation strategies.

4.3. Implications for the quest for mechanically recycled food-grade HDPE

The recycling of HDPE bottles to food-grade quality recycled HDPE is challenging for technical (Dutra et al., 2011; Gerassimidou et al., 2023; Rung et al., 2023) and legal reasons (Franz and Welle, 2022). This study revealed that the following aspects are important to limit the contamination of milk bottles and hence to facilitate the production of food-grade HDPE in the future. First of all, systemic migration of NIAS from the labels and caps to the bottle bodies needs to be limited. This implies that also the other packaging components need to be redesigned for food-grade recycling to minimise the migration between packaging components. This insight has major ramifications for future design-for-food-grade-recycling guidelines, as these will not only have to be defined on the material level of the packaging components but on the molecular level.

Secondly, the incidental contamination with product residues, IAS, etc. from other packages and objects needs to be minimised. This contamination occurs predominantly during sorting (stage G) and to a lesser extent during collection with LWP and cross-docking (stage E and F). Moreover this type of incidental contamination was more pronounced in the sorted product made from mechanically recovered plastics (G-MR) than in the sorted product made from the separately collected plastics (G-SC), see section 4.4. The most likely location in the sorting facility where cross-contamination between packages occurs is at the bale-press at the end of the sorting process. Here the sorted packages are compressed to blocks of roughly a cubic metre. Liquid product residues are squeezed out of these bales, especially when the sorted products are mostly comprised of either bottles (PET bottles, HDPE) or beverage cartons. During baling these sorted products, the liquid product residues drip out of the compressed bales into the gutter. This implies that this type of contamination can most effectively be reduced by sorting the HDPE milk bottle to a separate sorted product. Hence, bale pressing of mixed PE packages needs to be avoided.

This can be operationalised in two separate manners: implementing a mono-collection system for these bottles or keeping these bottles in the LWP and sort them out selectively. Mono-collection can be executed with separate drop-off containers (Switzerland PET recycling, 2023), or a deposit-refund system (Calabrese et al., 2021) or a mixture of collection methods as in Australia (Madden et al., 2023). Selective sorting can be executed with fluorescent markers (Woidasky et al., 2020; Larder and Hatton, 2023) and digital watermarks (De Tandt et al., 2021; ten Klooster and Thoden van Velzen, 2022). It is important to ascertain the absence of non-milk bottles when this new sorted product is

Table 5

Statistical comparison of the total approximated extracted volatiles from milk bottle bodies between the stages of the recycling value chain, showing if datasets are significantly different (SD) or not (NSD). White cells: *t*-Test. Grey cells: Mann-Whitney-U test.

Stage of the recycling value chain	Stage of the recycling value chain													
	A	B	C	D	E	F	G-SC	G-MR	H-M	H-CW	I-M	I-CW	I-HW	
A		NSD	NSD	SD	NSD	SD	SD	NSD	NSD	SD	SD	SD	SD	
B			NSD											
C				NSD	NSD	SD	NSD	NSD	NSD		SD		SD	
D					NSD									
E						NSD	NSD	SD	NSD		SD		SD	
F							NSD	SD	SD		SD		SD	
G-SC								SD	SD	NSD	SD		SD	
G-MR									NSD	NSD	SD		SD	
H-M										SD	SD			
H-CW											SD	SD		
I-M												SD		
I-CW													SD	
I-HW														

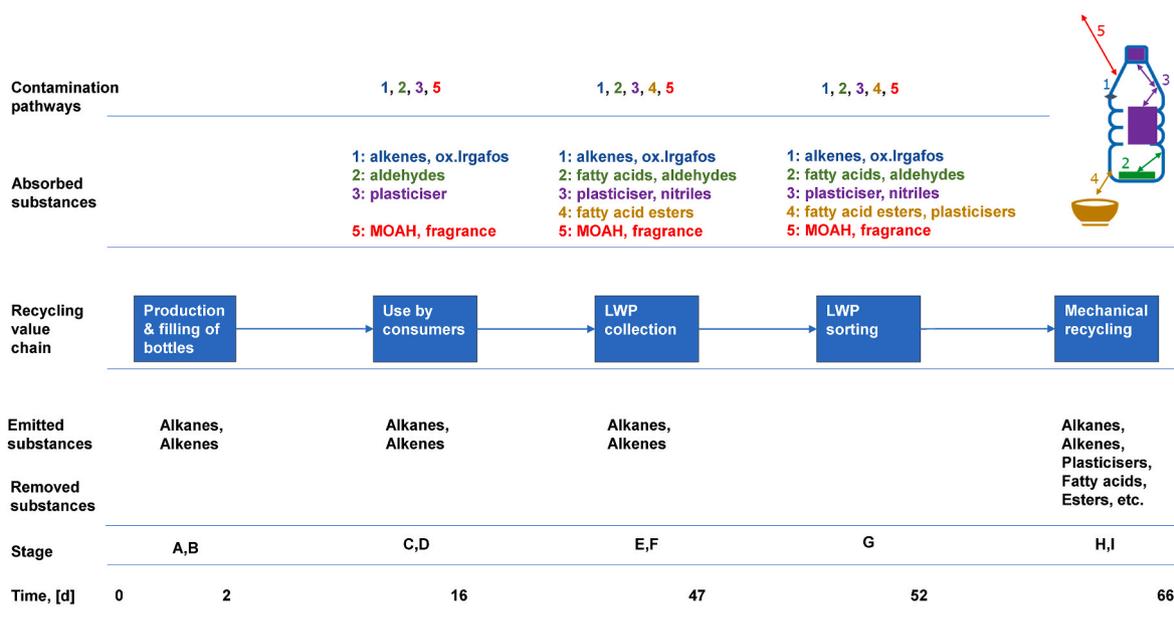


Fig. 3. Uptake and release of volatile contaminants in HDPE milk bottle bodies via five contamination pathways during the recycling value chain from stage A (bottle production up to mechanical recycling with a hot-wash (I-HW)).

bale-pressed, to exclude the exchange of non-milk-product residues such as personal care products and juices. By confirming that this sorted product contains only milk bottles, also the cross-contamination with non-milk product residues during the recycling stage is prevented.

In case the incumbents of the Dutch dairy industry want to proceed to make a high quality recycled HDPE from these milk bottles and also want to maintain the current LWP collection system, then a selective mechanical sorting technique, such as for instance marking, will have to be implemented and these bottles will need to be mechanically recycled separately. When an appropriate decontamination technology has been developed and has been placed on the list of “suitable technologies” as described in 2022/1616/EC (European Commission, 2022), a recycling company could develop a recycling process and apply for approval to make food-grade recycled HDPE in the coming years (European Commission, 2022).

4.4. Difference between separate collected versus mechanical recovered

Sorted milk bottles originating from mechanical recovery operations are more contaminated than sorted milk bottles that were separately collected, as is apparent in Figs. 1 and 2, S.3 and S.4 as the difference between G-SC and G-MR. More volatile compounds were identified in the mechanical recovered milk bottles than in the separately collected milk bottles (on average 33 ± 15 versus 20 ± 6 for extracted volatiles) and although the total approximated concentration overlaps partially, the statistical analysis reveals a significant difference between the milk bottles (section 3.3). A striking difference is formed by the outlier, indicating that the chance of incidental contaminants from other packages and objects is slightly larger in mechanical recovered milk bottles than in separately collected milk bottles, but in case this incidental contamination happens the impact of the total concentration is

pronounced. Bale pressing of the plastic concentrate at the recovery facility is the likely location where this cross contamination occurs. This can be avoided by feeding the plastic concentrate from the recovery facility directly to a neighbouring sorting facility. Previous public literature on this topic is scarce. A Spanish article reports that mechanically recovered recycled film smells differently from separately collected recycled film (Cabanes et al., 2020a; Cabanes et al., 2020b) and a Dutch study reports that the differences between recycled plastics from both origins are subtle (Thoden van Velzen et al., 2021).

5. Conclusion

The volatile contaminants in HDPE milk bottles were studied by sampling these bottles at nine different stages of the recycling value chain. The number of volatile contaminants registered and the total approximated concentration decrease between the production of the bottle and the cross-docking facility for lightweight packaging waste. The number of contaminants and the total approximated concentration peak at the sorted facility. In case the milk bottles are removed from the PE sorted product and they are mechanically recycled, a high quality of recycled HDPE can be obtained, which contains less contaminants than the freshly produced milk bottle. The composition of these contaminants is, however, different. In the freshly produced milk bottle only degradation products of HDPE and the antioxidant can be found, whereas in the mechanically recycled HDPE also traces of volatile contaminants are found that originate from the milk, the other packaging components, other packages and the surrounding atmosphere. Additionally, contaminants can originate from consumer abuse and thermal processing. This study expounds the seven different contamination pathways that can be distinguished for mechanically recycled plastic packages and engenders options to reduce these contaminants. To progress towards closed loop food-safe recycling of HDPE milk bottles two improvements need to be made. The other packaging components (cap, label, inlay) need to be redesigned-for-recycling on a molecular level to minimise migration of contaminants to the bottle. And secondly, the milk bottles need to be sorted into a separate sorted product prior to bale pressing to minimise contamination with product residues from different packages. As next steps in research, the exchange of volatiles in more complex packages that contain prints and adhesives could be studied and food safety risk assessments can be performed.

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CRedit authorship contribution statement

Eggo U. Thoden van Velzen: Writing – review & editing, Writing – original draft, Formal analysis, Data curation, Conceptualization. **Yarek Workala:** Investigation. **Wouter Teunissen:** Methodology, Investigation. **Ingeborg Smeding:** Writing – review & editing, Supervision, Project administration, Funding acquisition, Conceptualization.

Declaration of competing interest

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

Data availability

Data is saved in Mendeley Data repository

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.jclepro.2024.142571>.

Abbreviations

AHC	Aromatic hydrocarbons
DKR	Deutsche Gesellschaft für Kreislaufwirtschaft und Rohstoffe
EFSA	European food safety authority
GC-MS	Gas chromatography combined with mass spectroscopy
HDPE	High density polyethylene
IAS	Intentionally added substances
LDPE	Low density polyethylene
LOD	Level of detection
LOQ	Level of quantification
LWP	Lightweight packaging waste
MR	Mechanical recovery
NIAS	Non-intentionally added substances
NSD	Not significantly different
NTCP	National test centre for circular polymers
PE	Polyethylene
PET	Polyethylene terephthalate
PP	Polypropylene
PS	Polystyrene
PTV	Programmable temperature vaporising
SC	Separate collection
SD	Significantly different
SSL	Split/splitless
TIC	Total ion count

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