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<td>Complete List of Authors:</td>
<td>Welle, Frank; Fraunhofer IVV</td>
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Investigation into the decontamination efficiency of a new post-consumer poly(ethylene terephthalate) recycling concept

Frank Welle

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Abstract

The aim of the study was to investigate and evaluate the cleaning efficiency of a new recycling concept for post-consumer poly(ethylene terephthalate) (PET). The so-called Flake To Resin (FTR®) recycling process produces PET pellets or preforms from conventionally recycled PET flakes for the application in new PET packaging in direct food contact. The investigated process can be considered as "super-clean" recycling process and was developed to introduce conventional recycled post-consumer (PCR) PET flakes up to an amount of 50% into the pellet and preform production. Within the study, the cleaning efficiency of the investigated FTR® process was determined by a challenge test. The experimental results obtained from three challenge tests with different input concentrations of the surrogates and different amounts of post-consumer PET flakes show that all applied surrogates are very efficiently removed by the investigated recycling process. The cleaning efficiencies for all surrogates are above 99.9%. In the final product of the process no surrogates could be determined above the detection limits (0.5 mg/kg) even if the initial concentrations were in the percentage range. From a migrational point the final product, which are either PET pellets or preforms, was similar to virgin PET. Only the PET typical substances acetaldehyde and ethylene glycol had slightly higher concentrations than found in a conventional PET virgin sample used as reference. The generally accepted migration limit of 10 µg/kg for the surrogates in the final products of the challenge tests is established for all kinds of foodstuffs.
Keywords

PET recycling, analytical screening, challenge test, food packaging, migration

Introduction

Since the early 1990’s several recycling process technologies for post-consumer recycled poly(ethylene terephthalate) (PCR PET) have been developed. A few years later the first PCR PET for direct food contact applications entered the market. From a consumer safety point of view the concern is that post-consumer substances or compounds from the possible misuse of PET bottles, e.g. the storage of household chemicals, might migrate from the recylcate containing PET bottle into the packed food. Therefore several studies have investigated the principles of migration from post-consumer PET into food (simulants) (Begley and Hollifield 1993, Komolprasert et al. 1997, Bayer 1997, Bayer 2002). On the other hand, studies of the sorption of compounds in the PET materials has been published (Pierce et al. 1993, Huber and Franz 1997, Franz and Welle 1999, Bayer 2002, Begley et al. 2002, Franz et al 2004b).

The key issue for any super-clean recycling process has to be the decontamination of post-consumer compounds or substances from misuse to levels down to the range of impurities of virgin PET materials. The first step is recollection of the post-consumer PET bottles. Subsequently the PET bottles were sorted and ground into PET flakes. A washing process removed surface contamination as well as dust and paper fibres from the labels. In addition using swim-sink separation techniques, the polyolefin closures were removed from the PET flakes. The final product of the sorting and washing steps are surface cleaned PET flakes, which are suitable for non-food packaging applications. If the post-consumer PET should be reused in food packaging further deep-cleansing steps should be applied. Most of the so-called super-clean recycling processes using melt phase filtration, high temperature and vacuum treatment of the flakes and pellets or surface treatment of the washed flakes. Using such treatment post-consumer contaminants from the first use of the PET packaging as well as compounds from possible misuse of PET bottles for storage of e.g. household chemicals were efficiently removed. However before
using the super-cleaned recyclates for direct food contact applications, the cleaning efficiency of
the super-clean recycling process has to be determined. Under consideration of the input
concentrations of potential compounds for the first use of the packaging materials as well as for
compounds coming from misuse of the PET bottles, the ability for the reuse of the final product for
direct food contact can be evaluated (Franz et al 1998, Franz and Welle 1999, Franz and Welle
2002). Provided that the cleaning efficiency is high enough in relation to the input concentrations
of post-consumer compounds, the final product of such super-clean processes is from a migration
point of view similar to virgin PET and can be reused in food packaging in direct food contact.

Typically, the state-of-the-art super-clean recycling processes are separated from the virgin PET
production. Recycling lines are built up far away from the virgin PET production lines as well as
from the preforms and bottle manufacturing plants, which were using a certain amount of super-
cleaned recycled PET for the bottles. A new development in the poly(ethylene terephthalate) PET
recycling technologies is a so-called Flake To Resin (FTR®) process, which combines the virgin
PET production with the super-clean recycling in one process step. The technology is similar to the
so-called Melt-To-Resin (MTR®) processes, where PET pre-polymers are further reacted to the
final polymer in only one step without a solid stating process step. The investigated FTR® process
starts – similar to nearly every super-clean recycling process - with conventionally recycled post-
consumer PET flakes. The flakes were re-molten and mixed with PET precursors (oligomer),
which were typically used for the polymerisation process of virgin PET. In one process step the
mixture was further condensed to the final PET product and transformed into PET pellets (or PET
preforms). Due to the fact, that during the polymerisation process of the pre-polymer under high
temperature and vacuum conditions, the post-consumer PET should be also deep-cleansed
similar to other super-clean PET bottle-to-bottle recycling processes. The advantage of such a
process is, that the same production line can produce PET pellets in a flexible way either from
100% virgin pre-polymers or from a mixture of virgin PET and post-consumer PET flakes. The
amount of conventional recycled post-consumer (PCR) PET flakes can be introduced in a flexible
way into the PET pellet production process (in principle up to 100%). The PET producer is therefore able to react on the actual demands of the (recycling) market.

The cleaning efficiency of the investigated FTR recycling process was determined by carrying out a challenge test according to the principles recommended by United States FDA (FDA 1992, FDA 2006) and European Guidelines (ILSI 1998, Franz et al 2004a) in order to evaluate whether the output material is suitable for re-use in packaging materials with direct food contact.

**Experimental**

**Input Material for the Recycling Process**

The investigated FTR® process uses conventionally recycled post-consumer PET flakes as a recycling material source. Typically the input material for conventional recycling processes is post-consumer food PET containers from deposit systems as well from curbside collections. However some PET containers previously might have been used for non-food applications. The amount of the non-food PET container fraction depends on the re-collection system. A general statement about the percentage of the non-food fraction is therefore not possible. However, the amount of the non-food container fraction might be not higher than 20%. As far as we know all PET containers for food applications as well as for non-food applications are manufactured from food grade PET material thus being in compliance with 21 CFR 177.1630 as well as with 2002/72/EC.

**Recycling Process**

The investigated FTR® process starts with PET monomers which are chemically combined in a specific reactor. In this reactor the esterification and pre-polycondensation processes take place and generate the virgin pre-polymer which is used for the main process stream of the FTR® process. In a second line, conventional recycled post-consumer PET flakes are molten and partly depolymerised with ethylene glycol to a defined degree of polymerisation. The degree of polymerisation is brought into a range, which corresponds to the virgin pre-polymer in the main
The depolymerisation process of the post-consumer PET allows a fine filtration of polymer to remove solid contaminants like fibres, metals or sand. Subsequently the depolymerised post-consumer PET is blended into the virgin pre-polymer from the main process stream without a pre-drying step. This is done at an appropriate point of a continuous polycondensation process right upstream of the last reaction stage. The applied melt-phase polymerisation technology allows the increase of the viscosity to levels required for direct use in PET bottle applications. In addition to the viscosity increase and carboxyl-group regulation, post-consumer contaminants and polymerisation by-products are removed by high vacuum (approx. 100 Pa), high temperature (above 270 °C) and residence time (more than 60 min) treatment in a highly surface active DISCAGE® reactor. Following polycondensation reaction the polymer melt is filtered and converted into PET pellets by granulation. The pellets are used directly for PET preform manufacturing after having passed an acetaldehyde degassing system. The process is also capable to produce preforms directly from the melt phase without the granulation step.

**Challenge-Tests**

The cleaning efficiency was determined by a challenge test by artificial contamination of PET flakes with model contaminants (surrogates). According to the generally accepted contamination protocols for challenge tests (FDA 1992, FDA 2006, ILSI 1998, Franz et al 2004a), post-consumer PET flakes should be soaked in a solution of the surrogates. Subsequently the contaminated material should be recycled with the investigated recycling process. In the special case of the investigated FTR® process, the surrogates can be injected directly into the polymer melt while introducing non-contaminated pre-polymers and conventionally recycled post-consumer PET flakes. For the contamination the solid model compounds were dissolved in the liquid chemicals without using an additional solvent to give a neat solution of the surrogates. This solution was pumped into the extruder directly to the polymer melt. This procedure allows the introduction of much higher concentrations of the surrogates in comparison to PET soaking contamination procedures as well as a flexible variation of the concentration during the challenge tests.
recyclate amount of the different challenge tests and the spiking levels of the surrogates are given in Table 1 and Table 2.

Place here Table 1

Place here Table 2

Surrogates

The cleaning efficiency of the recycling process was determined by introducing surrogates into the FTR® process. The surrogates were chosen in accordance with US FDA (FDA 1992, FDA 2006) and in agreement with EU-relevant criteria (ILSI 1998, Franz et al 2004a) such that they covered the whole spectrum of physical/chemical properties. The surrogates are in compliance with the four general categories of organic compounds: high volatile and polar, high volatile and non-polar, low volatile and polar as well as low volatile and non-polar. In addition, the surrogates used in the challenge test represent a broad variety of functional groups in order to reflect the different chemical nature and physical properties of possible real-life contaminants e.g. aliphatic and aromatic hydrocarbons, chlorinated hydrocarbons and carbonyl or ester functional groups. Also the molecular weight was important for the selection of the surrogates. Chemicals with a molecular weight up to approximately 300 g mol⁻¹ are relevant for migration from and into PET. Substances with a molecular weight >300 g mol⁻¹ have a very low migration potential in the case of low diffusivity plastics such as PET (Franz et al 2004a). The following surrogates were used for the challenge-tests: Toluene (CAS No. 108-88-3) chloroform (CAS No. 67-66-3), methyl salicylate (CAS No. 119-36-8), phenyl cyclohexane (CAS No. 827-52-1), benzophenone (CAS No. 119-61-9) and lindane (CAS No. 58-89-9). The surrogates were introduced directly into the melt-phase zone of the extruder used for remelting of the conventionally recycled PET flakes.
Determination of the Cleaning Efficiencies

During the challenge tests samples were drawn from the final product as well as from intermediate process stages. In these samples the surrogate concentrations were determined using an extraction method after swelling of the polymer matrix with help of 1,1,1,3,3,3-hexafluoro-iso-propanol (HFIP). HFIP is a well-known very aggressive solvent for PET. The extracts of the challenge test samples were analysed using gas chromatography with flame ionisation detector (FID) or electron capture detector (ECD). From each PET sample two sub-samples were drawn and extracted. The extracts were injected three times into the gas chromatograph. The cleaning efficiencies of the investigated FTR® process were determined for each surrogate from the concentration in the PET sample under consideration of the initial concentrations led into the recycling process according to Equation 1. The results were verified with another independent method (headspace gas chromatography, details see below). The headspace gas chromatograms were evaluated only on a relative basis, e.g. to verify the absence of the surrogates in some samples.

Place here Equation 1

HFIP-Extraction of the PET Material

Each PET material sample was analysed twice in the following way: 1.0 g of each PET sample was placed in a 5 ml glass vial. 1.0 ml 1,1,1,3,3,3-hexafluoro-iso-propanol (HFIP) was given to the PET material and stored for 1 d at 60 °C in order to swell the PET matrix. Then 2.0 ml iso-propanol was added for 1 d at 60 °C to extract the swollen matrix. The extract was decanted from the polymer and stored for 8 h at 4 °C. Subsequently the extracts were decanted again from the precipitate and analysed by GC/FID and GC/ECD.
**GC/FID Analysis and GC/ECD Analysis**

The extracts of the challenge test samples were analysed according to the concentrations of all surrogates except lindane by gas chromatography (FID). Chloroform was monitored by an electron capture detector (ECD). For this purpose a gas chromatograph with a simultaneous detection by an FID and ECD detector was used. Quantification was achieved by external calibration using the standard addition method. Parts of a standard solution of the surrogates in iso-propanol were added to uncontaminated PET flakes and were analysed together with the PET samples of the contamination experiments. Gas chromatograph: HP 5890II, column: SE 10 - 30 m - 0.32 mm i.d. - 0.32 µm film thickness, temperature program: 40 °C (5 min), rate 15 °C min\(^{-1}\), 240 °C (15 min), pressure: 50 kPa hydrogen, split: 10 ml min\(^{-1}\).

**Quantification of Lindane**

The surrogate lindane was determined by FID with the following equipment stating from the extracts. Gas chromatograph: Hewlett Packard 6890, column: DB 1 - 30 m - 0.25 µm i.d. - 0.25 µm film thickness, temperature program: 50 °C (2 min), rate 10 °C min\(^{-1}\), 340 °C (2 min), flame ionisation detection, pressure: 50 kPa hydrogen, split: 10 ml min\(^{-1}\). Quantification of lindane was achieved by external calibration using the standard addition method.

**Detection Limits**

The detection limits for the above mentioned analytical methods are given in Table 3. The detection limits were determined using stock solutions of the surrogates. From the slope of the calibration curves the detection limits were calculated for each surrogate according to DIN 32645.

Place here Table 3
Headspace Screening of the Challenge Test Samples for Verification Purposes

1.0 g of the PET samples was weighed into a headspace vial and analysed via headspace GC/FID. Gas chromatograph: Perkin Elmer AutoSystem XL, column: ZB 1 - 30 m - 0.25 mm i.d. - 0.32 µm film thickness, temperature program: 50°C (4 min), rate 20°C min⁻¹, 320°C (15 min), pressure: 50 kPa helium, split: 10 ml min⁻¹. Headspace Autosampler: Perkin Elmer HS 40 XL, oven temperature: 200°C, needle temperature: 210°C, transfer line: 210°C, equilibration time: 1 h, pressurization time: 3 min, injection time: 0.02 min, withdrawal time: 1 min. For identification, another 1.0 g sample was drawn and analysed via headspace GC/MS. Gas chromatograph: Hewlett Packard 6890, column: Optima 1 MS - 30 m - 0.25 mm i.d. - 0.25 µm film thickness, temperature program: 50°C (4 min), rate 20°C min⁻¹, 320°C (15 min), full scan, m/z 40-500. Headspace autosampler: Perkin Elmer HS 40 XL, oven temperature: 200°C, needle temperature: 210°C, transfer line: 210°C, equilibration time: 1 h, pressurization time: 3 min, injection time: 0.04 min, withdrawal time: 1 min. The mass spectra that were obtained were identified by comparison with spectra in the NIST spectral library.

Results and Discussion

Determination of the Cleaning Efficiencies

For the challenge tests the extruder was fed with the PET pre-polymer (pellets) as well as with a certain amount of post-consumer PET flakes. Three challenge tests were performed with different amounts of PCR PET flakes as well as with different spiking levels of the surrogates. PCR PET in amounts of 15%, 25% and 50% as flakes referred to the total polymer were introduced. The surrogate mixture was directly fed into the extruder for re-melting of the PCR PET flakes and PET virgin pellets so that a homogeneous mixture of PET and surrogates was introduced into the FTR® process. The artificially introduced surrogate levels calculated for the amount of PCR PET flakes are summarized in Table 4. The concentrations of the surrogates in the investigated samples determined by HFIP extraction are summarised in Table 5 to Table 7.
Screening of the Challenge Test Samples for Migration Relevant Compounds

The screening for migration relevant compounds in the PET materials and bottles was carried out using headspace gas chromatography. This method detects substances up to a molecular weight of about 300 g mol\(^{-1}\). The detection limit of the applied method is below 0.5 mg/kg (Franz et al 2004b). A representative headspace gas chromatogram of virgin PET pellets suitable for PET bottle manufacturing is shown in Figure 1 (reference material). Examples of the headspace gas chromatograms of the investigated samples are shown in Figure 2 and 3. In Figure 2 a sample of the melt after contamination is shown. Figure 3 shows the corresponding pellet sample after deep cleansing (final product of the investigated process).

In the virgin PET reference sample (figure 1) only PET typical compounds like acetaldehyde (retention time \(R_t = 1.7\) min), 2-methyl-1,3-dioxolane (\(R_t = 2.4\) min) and ethylene glycol (\(R_t = 2.7\) min) could be determined. Acetaldehyde is a thermal degradation product of poly(ethylene terephthalate) whereas ethylene glycol is one of the residual monomers. 2-Methyl-1,3-dioxolane is the condensation product of acetaldehyde and ethylene glycol. These compounds could be determined in every PET pellet sample, independent whether post-consumer PET is introduced or not. In Figure 2 several additional substance peaks could be determined. The artificially introduced surrogates were identified by mass spectrometry as well as by comparison of the retention time of neat standard substances: chloroform (\(R_t = 2.2\) min), toluene (\(R_t = 3.6\) min), chlorobenzene (\(R_t = 4.4\) min), methyl salicylate (\(R_t = 9.5\) min), phenyl cyclohexane (\(R_t = 10.7\) min), benzophenone (\(R_t = 12.8\) min) and lindane (\(R_t = 13.6\) min). The other substance peaks are by-products or impurities of the applied surrogates. The surrogates could only be determined in the samples drawn from the
melt before the melt-phase reactor used for decontamination and IV increase (Figure 2). In this process stage, the contaminated melt is mixed with the melt from the pre-polymers. Therefore the concentration of the spiked surrogates is reduced by the introduction of the virgin pre-polymers (e.g. by 50% if 50% virgin pre-polymer is used). In the pellet samples after decontamination (Figure 3) surrogate concentrations below the analytical detection limits were determined. This result indicates the high decontamination effect of the applied process. Only the PET degradation product acetaldehyde has higher concentrations than found in the virgin PET samples used as reference.

The experimentally determined concentrations in the final product are far below the maximum concentrations given by Franz et al (2004a). Based on migration models (Begley and Hollifield 1993, Baner et al 1996 Piringer and Baner 2000, Piringer and Hinrichs 2001, Begley et al. 2005) Franz et al calculated the maximum residual concentration $c_0$ of contaminants (surrogates) in the bottle wall for a 1 l bottle with 600 cm$^2$ surface area (“EU cube”) which correspond to the migration limit of 10 $\mu$g/kg in PCR PET bottled foodstuff (FDA 1992, ILSI 1998, BgVV 2000). Toluene for example would allow a maximum concentration of 4.5 mg/kg of storage of 10 d at 40 °C for olive oil as food simulant. The higher molecular weight compound benzophenone allows a maximum concentration of 8.6 mg/kg in the bottle wall. Therefore the investigated process fulfills the requirements on the migration limits for the applied surrogates under worst case contamination conditions.

Conclusions

The experimental results obtained from the challenge-tests show that all applied surrogate compounds are efficiently removed by the investigated FTR recycling process. It could be shown that the melt phase polycondensation reactor plays a crucial role in the decontamination of the post-consumer recyclates (PCR). The cleaning efficiencies for the applied surrogates are above or far above 99.9%. The high cleaning efficiencies are due to the high diffusion rates of compounds in
the molten PET. These cleaning efficiencies are similar or higher in comparison to other PET
super-clean recycling technologies based on decontamination of PET flakes or pellets (Franz and
Welle 2003).

For methyl salicylate, the low concentrations determined in the PET melt are most probably due to
a trans-esterfication reaction which decreases the concentration of the surrogate by a chemical
reaction and not by evaporation from the polymer melt. Methyl salicylate could therefore not be
used as an indicator for the surrogates removal potential of the investigated recycling process.

This result indicates that methyl salicylate is not a useful surrogate for recycling processes which
are based on a decontamination step in the melt phase. In solid stating processes, however,
methyl salicylate seems to be a good surrogate for the simulation of non-food PET contaminants
(Franz et al 2004a). In principle, the possible side reaction of this surrogate shows, that no general
procedure exists for a challenge test, which could be applied to any recycling PET process.

Moreover, the applied surrogates and the input concentration should be chosen in relation to the
investigated recycling process as well as in relation to the concentration of post-consumer
contaminants in the input material.

In the final product of the investigated process, which are either PET pellets or preforms, no
surrogates could be determined either at standardized contamination levels or freely selected
several thousands mg/kg. Such high initial concentrations represent an extremely high
contamination level of the PET which most probably could not be established in real life. Post-
consumer compounds as well as compounds from misuse of PET containers are on average
present in the lower mg/kg range (Franz and Welle 2003, Franz et al 2004b). This already follows
from purely statistical considerations regarding the frequency of return of highly contaminated
bottles and the correspondingly very high dilution factor.

The high cleaning efficiency was verified using a second, independent screening method. The
results of both methods are in good agreement. Except the PET typical substances like
acetaldehyde and ethylene glycol, the final products of the challenge test were - from a migrational
point of view - comparable to virgin PET.
Under the assumption that the measured residual surrogate concentrations in the final product would be the same as in the bottle produced thereof, the maximum concentration in the bottle wall corresponding with the generally accepted migration limit of 10 µg/kg are established for all kinds of foodstuffs. The investigated FTR® recycling process is therefore in a position to produce pellets and preforms with up to 50% recycled PET which fully meets European (ILSI 1998, BgVV 2000) as well as US regulations (FDA 1992) and is therefore capable to recycle post-consumer PET material for the sensitive applications of direct contact packaging for foodstuff. Therefore, the applied melt phase decontamination of post-consumer PET provides an interesting process for the effective decontamination of post-consumer PET.

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Figure 1: Headspace gas chromatogram of virgin PET (reference sample)
Figure 2: Headspace gas chromatogram of a melt sample before reactor (25% flakes, Cocktail A at 5 ml min⁻¹)
Figure 3: Headspace gas chromatogram of the deep-cleansed final product (25% flakes, Cocktail A at 5 ml min⁻¹)
Equation 1: cleaning efficiency \( = \left( 1 - \frac{\text{sample conc.}}{\text{initial conc.}} \right) \times 100\% \)

Table 1: Parameters used for the challenge tests

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<tr>
<th>Trial (recyclate content)</th>
<th>Amount</th>
<th>Surrogate cocktail (dosage)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>PET Prepolymer</td>
<td>PCR PET</td>
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<tr>
<td>Trial 1 (25%)</td>
<td>75 kg h(^{-1}) (1250 g min(^{-1}))</td>
<td>25 kg h(^{-1}) (417 g min(^{-1}))</td>
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<tr>
<td>Trial 2 (50%)</td>
<td>50 kg h(^{-1}) (833 g min(^{-1}))</td>
<td>50 kg h(^{-1}) (833 g min(^{-1}))</td>
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<tr>
<td>Trial 3 (15%)</td>
<td>85 kg h(^{-1}) (1417 g min(^{-1}))</td>
<td>15 kg h(^{-1}) (250 g min(^{-1}))</td>
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Table 2: Amounts of surrogates used for the spiking cocktails

<table>
<thead>
<tr>
<th>Surrogate</th>
<th>Amount [g]</th>
<th>Cocktail A</th>
<th>Cocktail B</th>
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</thead>
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<tr>
<td>Toluene</td>
<td>226.5</td>
<td>151.0</td>
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<tr>
<td>Chloroform</td>
<td>357.0</td>
<td>238.0</td>
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<tr>
<td>Chlorobenzene</td>
<td>86.3</td>
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<tr>
<td>Phenyl cyclohexane</td>
<td>22.5</td>
<td>15.0</td>
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<tr>
<td>Methyl salicylate</td>
<td>69.0</td>
<td>46.0</td>
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<tr>
<td>Benzophenone</td>
<td>60.8</td>
<td>40.5</td>
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</tr>
<tr>
<td>Lindane</td>
<td>53.3</td>
<td>35.5</td>
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<tr>
<td>Ethylene glycol[a]</td>
<td>60.0</td>
<td>583.5</td>
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</tbody>
</table>

[a]Ethylene glycol was used to reduce the IV during processing. Ethylene glycol was not used as a surrogate.
Table 3: Analytical detection limits of the surrogates (HFIP Method)

<table>
<thead>
<tr>
<th>surrogate</th>
<th>detection limit [ppm]</th>
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<tr>
<td>Toluene</td>
<td>2.7</td>
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<tr>
<td>Chloroform</td>
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<tr>
<td>Chlorobenzene</td>
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<td>Phenyl cyclohexane</td>
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<tr>
<td>Benzophenone</td>
<td>0.2</td>
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<tr>
<td>Lindane</td>
<td>0.8</td>
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Table 4: Initial concentrations of the surrogates in relation to the amount of PCR PET flakes

<table>
<thead>
<tr>
<th>Surrogate</th>
<th>Initial concentration [ppm]</th>
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</thead>
<tbody>
<tr>
<td></td>
<td>Trial 1</td>
</tr>
<tr>
<td></td>
<td>Cocktail A, 5 ml min(^{-1}), 25% PCR PET</td>
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<tr>
<td>Toluene</td>
<td>3295</td>
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<td>Chloroform</td>
<td>5194</td>
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<tr>
<td>Chlorobenzene</td>
<td>1255</td>
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<tr>
<td>Phenyl cyclohexane</td>
<td>327</td>
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<tr>
<td>Methyl salicylate</td>
<td>1004</td>
</tr>
<tr>
<td>Benzophenone</td>
<td>885</td>
</tr>
<tr>
<td>Lindane</td>
<td>775</td>
</tr>
<tr>
<td>Ethylene glycol(^{[a]})</td>
<td>873</td>
</tr>
</tbody>
</table>

\(^{[a]}\)Ethylene glycol was used to reduce the IV during processing. Ethylene glycol was not used as a surrogate.
Table 5: Concentrations (determined using the HFIP extraction method) of the surrogates in the investigated PET samples of Trial 1 (Cocktail A at 5 ml min\(^{-1}\), 25% PCR flakes)

<table>
<thead>
<tr>
<th></th>
<th>Concentration [ppm]</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Toluene</td>
</tr>
<tr>
<td>before contamination</td>
<td>&lt;2.7</td>
</tr>
<tr>
<td>calculated contamination conc.</td>
<td>3295</td>
</tr>
<tr>
<td>Before deep-cleansing</td>
<td>852 ±23</td>
</tr>
<tr>
<td>After deep-cleansing (final product)</td>
<td>&lt;2.7</td>
</tr>
</tbody>
</table>
Table 6: Concentrations (determined using the HFIP extraction method) of the surrogates in the investigated PET samples of Trial 2
(Cocktail A at 10 ml min\(^{-1}\), 50% PCR flakes)

<table>
<thead>
<tr>
<th></th>
<th>Concentration [ppm]</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Toluene</td>
</tr>
<tr>
<td>calculated contamination conc.</td>
<td>3295</td>
</tr>
<tr>
<td>Before deep-cleansing</td>
<td>1999 ±28</td>
</tr>
<tr>
<td>After deep-cleansing (final product)</td>
<td>&lt;2.7</td>
</tr>
</tbody>
</table>
Table 7: Concentrations (determined using the HFIP extraction method) of the surrogates in the investigated PET samples of Trial 3 (Cocktail B at 10 ml min\(^{-1}\), 15% PCR Flakes)

<table>
<thead>
<tr>
<th></th>
<th>Concentration [ppm]</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Toluene</td>
</tr>
<tr>
<td>calculated</td>
<td></td>
</tr>
<tr>
<td>contamination conc.</td>
<td>5812</td>
</tr>
<tr>
<td>Before deep-cleansing</td>
<td>826 ±1</td>
</tr>
<tr>
<td>After deep-cleansing (final product)</td>
<td>&lt;2.7</td>
</tr>
</tbody>
</table>