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Investigation into cross-contamination during cleaning efficiency testing in PET recycling

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ABSTRACT

The cleaning efficiency of a PET recycling process is typically investigated by artificial contamination of post-consumer PET flakes within a so-called challenge test. Challenging of pilot plants or industrial scale lines is done be introducing a certain amount of contaminated flakes while running the process with non-contaminated flakes of different colour. After decontamination the contaminated flakes are separated from the non-contaminated flakes and only the contaminated flakes were analysed due to their residual contamination level. The European Food Safety Authority (EFSA), however, raised the question about cross-contamination, which might reduce the overall cleaning efficiency of the recycling process. Cross-contamination is defined as the transfer of surrogate contaminants from the initially contaminated to the initially not contaminated material during a challenge test. Data for the phenomenon of cross-contamination are not available in the scientific literature. Aim of the study was to close this gap by providing experimental data for cross-contamination by use of several challenge tests. As a result cross-contamination was found only at ratios of 1:1 between contaminated and non-contaminated PET flakes. At higher ratios which were typically applied in challenge tests on pilot plant or industrial scale line cross-contamination do not play a significant role. In addition, the results show that cross-contamination is negligible for volatile compounds.

1. Introduction

The recycling of polyethylene terephthalate (PET) bottles is increasing all over the world. In Europe about 56% (1.64 Mio. t) of the PET bottles entering the market are recollected and recycled. A percentage of nearly 50% of the recycled PET ends up in new packaging applications (Welle, 2014a). Recycling processes for PET bottles are established all over Europe. In the first step, the recollected PET bottles are washed and ground to so-called PET flakes. During the washing step, also labels, polyolefin caps, glue from the labels and other non-PET materials were removed. PET recyclates which are re-used in new packaging application were treated in a further deep-cleansing process in order to clean-up the washed PET flakes to contamination levels similar to virgin PET. Although there are several different processes established, the processes are following the same decontamination principles. The deep-cleansing processes typically using high temperature treatment under vacuum or inert gas treatment and re-extrusion steps to decontaminate the flakes (Welle, 2011) in order to remove potential contaminants from the first use of the PET bottles. At the end of the PET recycling the recyclates should be – from a migrational point of view – similar to virgin PET.

Post-consumer PET recyclates in direct food contact applications are restricted by authorities, because contaminants might be introduced into the packaging materials and might be a hazard to consumers. According to the Recycling Regulation 282/2008 (EU, 2008) the recycling companies which want to re-use the recyclate in direct food contact had to submit petition for an official approval of the recycling process. The European Food Safety Authority (EFSA) had been commissioned with the scientific evaluation of submitted petitions. The main evaluation principles for PET recycling processes are (i) the control of the input materials (recollected bottles or washed PET flakes), (ii) the cleaning efficiency of the PET recycling process towards potential contaminants from the first use as well as (iii) the quality assurance of the treated post-consumer PET recylcates.

The cleaning efficiency of a PET recycling process is investigated by artificial contamination of post-consumer PET flakes within a so-called challenge test (Franz and Welle, 2003; Welle and Franz, 2007). Within such a challenge test, post-consumer PET bottle flakes were contaminated with model contaminants (surrogates) e.g. toluene, chlorobenzene, phenyl cyclohexane and benzophe-

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none (Welle and Franz, 2007). The levels of the surrogates in the artificial contaminated flakes should be in any case higher than found in the recollected bottles or washed flakes in order to establish a worst-case scenario in the challenge test. Typically the contamination levels of the surrogates in the post-consumer PET flakes during a challenge test are in the range of 300 mg/kg up to about 500 mg/kg (Franz et al., 2004a,b). The artificially contaminated flakes are subsequently recycled with the investigated PET recycling process. After each process step, recyclate samples are drawn and analysed due to the residual levels of artificial contaminants. The difference between the input concentration and the concentration in the final product of the PET recycling process is due to the cleaning efficiency of the recycling process and can be calculated thereof for each individual surrogate. As mentioned above, this cleaning efficiency is one of the main evaluation criteria for the efficiency of PET recycling to remove contaminants from the first use of the bottles.

Typically about 30 kg to 100 kg of contaminated flakes are introduced at the same time into the PET recycling process. Such high contamination levels cannot be achieved on a big scale in recollection systems but, if at all, in individual bottles only after misuse of a PET bottle by a consumer for storage of solvents or garden chemicals (Franz et al., 2004a,b). From statistical considerations regarding the frequency of return of highly contaminated (misused) bottles and the inherent high dilution effect, average contamination levels which might be present in the post-consumer PET feedstream entering the recycling technology must be significantly lower. Therefore the above described challenge test scenario with contamination levels of 300 mg/kg to 500 mg/kg can be considered as worst case for recycling of PET beverage bottles (Franz et al., 2004a,b).

Industrial scale recycling processes have typically throughputs of about 300 kg/h up to 2000 kg/h. Pilot plant scale lines are available in the range of 100 kg/h up to 300 kg/h. As mentioned above, typically the challenge tests are carried out with about 30 kg to 100 kg of contaminated PET material (Franz et al., 1998; Franz and Welle, 1999; Franz and Welle, 2002). Higher loads than 100 kg of contaminated material are not applicable due to safety reasons, because the recycling companies or machinery manufacturers are not in a position to handle such high amounts of high volatile, hazardous and flammable chemicals within their pilot plant facilities. As a consequence, challenging of pilot plants or industrial scale lines is done by introducing a certain amount of contaminated flakes while running the process with non-contaminated flakes of different colour. After decontamination the contaminated flakes can be easily separated from the non-contaminated flakes by their different colour and only the contaminated flakes were analysed due to their residual contamination level. Recycling processes which are based on re-melting of the PET flakes can be evaluated by use of integral procedure (Franz and Welle, 2002).

By use of the above descript challenge test procedure is possible to determine the cleaning efficiency of nearly every pilot plant or industrial scale plant with a limited amount of contaminated flakes and therefore with a minimum amount of hazardous chemicals. The US FDA has accepted the procedure with contaminated green flakes while running the pilot plant line with non-contaminated clear flakes. Several companies got full FDA approval for all their PET recycling lines up to 100% of recyclate inclusive hot-fill conditions (FDA, 2015). For some applications also microwave heating conditions are approved by the US FDA, but still up to 100% of recyclate content (FDA, 2015).

The European Food Safety Authority (EFSA), on the other hand, did not completely accept the use of contaminated material in a challenge test while running the process with non-contaminated clear flakes (EFSA, 2013a,c; EFSA, 2014; EFSA, 2015a,b). The EFSA stated in their scientific opinions that there might be a cross-contamination between contaminated flakes and non-contaminated flakes, which decreases the overall cleaning efficiency determined within the challenge tests. Cross-contamination is defined as the transfer of surrogate contaminants from the initially contaminated flakes to the initially not contaminated material during the challenge test. To best of our knowledge, however, data which support the hypothesis that cross-contamination occurs are not available in the scientific literature. On the other hand, assuming that such cross-contamination exists, the amount of transfer of chemicals between contaminated and non-contaminated flakes will be dependent on the volatility of the contaminants. For highly volatile compounds like solvents the cross-contamination should be lower than for non-volatile compounds. In addition, the level of cross-contamination should be dependent on the ratio between contaminated and non-contaminated flakes.

The EFSA evaluates challenge tests, which are using contaminated flakes while running with non-contaminated flakes of a different colour, by assuming a cross-contamination rate of 10% for all surrogates independent of their volatility (EFSA, 2013a,b,c; EFSA, 2014; EFSA, 2015a,b). By use of such extremely conservative evaluation criteria, the maximum amount of 100% recyclate was in some cases not in compliance with the EFSA evaluation criteria for the minimum cleaning efficiencies of PET recycling processes (EFSA, 2011; Barthelemy et al., 2014). As a consequence, EFSA reduced the amount for some recycling lines down to levels of only 40%, depending on the residence time of the different recycling lines (EFSA, 2013a,c; EFSA, 2014; EFSA, 2015a,b). Especially the volatile compounds like toluene and chlorobenzene with its high migration potentials are influencing the EFSA decisions. The minimum cleaning efficiencies of non-volatile compounds like benzophenone or methyl stearate are still in agreement with the EFSA evaluation criteria even if 10% of cross-contamination is considered. As a consequence the assumption of cross-contamination by the EFSA can be considered as one of the main reasons for limiting the amount of PET recyclates in food contact articles in Europe.

The aim of the study was to close this gap by providing experimental data for cross-contamination by use of several challenge tests with different ratios of contaminated green flakes as well as different process conditions. For this purpose, contaminated as well as non-contaminated flakes from challenge test were analysed regarding their residual contamination levels. In order to establish general applicable data on cross-contamination, different recycling scenarios are established including hot washing and cold washing processes as well as different decontamination temperatures. From the comparison of the residual contamination levels the contribution of cross-contamination to the overall cleaning efficiencies of the PET recycling processes was evaluated.

2. Materials and methods

2.1. Study design

In the first step of the study post-consumer PET flakes were contaminated with seven model substances (surrogates) using a standard contamination protocol (see below). The surrogates represent the four general categories of contaminants like (i) volatile and polar, (ii) volatile and non-polar, (iii) non-volatile and polar and (iv) non-volatile and non-polar. After contamination, the PET flakes were divided into three sub-batches. The first batch was further processed without any washing process. The two other sub-batches were processed with two different washing process conditions. One batch was treated with a hot wash process for 10 min at 75 °C. The other batch was washed only at a temperature of 50 °C ("cold wash"). Both washing processes are followed by a rinsing step with cold water as well as by hot air drying (140 °C) in order to remove

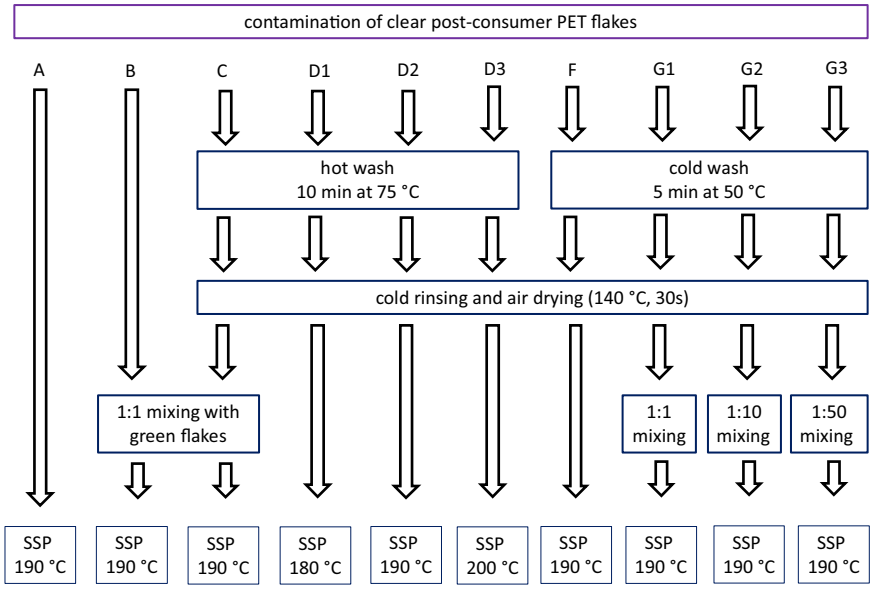


Fig. 1. Scheme of the process steps for the individual decontamination trials.

surface water on the flakes. The flakes samples were subsequently treated by use of a solid-state poly-condensation (SSP) process. Within some trials, the contaminated flakes were mixed with non-contaminated flakes in order to determine the cross-contamination during the cleaning efficiency of the recycling process. The whole study design with all trials and process conditions is shown in Fig. 1.

The study design allows to compare the different recycling trials regarding the amount of cross-contamination as well as the influence of process conditions on the cleaning efficiencies of the applied recycling steps. For example, comparison of the results of trials A and B allows to investigate the influence of mixing of contaminated with non-contaminated flakes (cross-contamination). The trials B, C and G1 to G3 show the influence of the washing step on the cross-contamination. The results of trials A, D2 and F can be used to investigate the influence of the washing temperature on the cleaning efficiency of the subsequent recycling processs whereas trials D1 to D3 investigate the influence decontamination temperature on the cleaning efficiency of hot wahed flakes. The influence on the percentage of mixing the contaminated flakes with non-contaminated flakes are investigated in trials G1 to G3.

All trials were performed with six kinetic points. For this purpose samples were drawn at 25 min, 55 min, 85 min, 115 min, 175 minand 355 min after starting the decontamination process (SSP). Each kinetic point was analysed in triplicate and the standard deviation of the concentrations of these sub-samples are given in the Tables.

2.2. Surrogates used for the challenge test

The cleaning efficiency of PET recycling processes is usually determined by a so-called challenge test by artificial contamination of PET flakes. Post-consumer PET flakes were contaminated with surrogates instead of real contaminants (Welle and Franz, 2007). The surrogates applied in this study are given in Table 1.

2.3. Contamination of clear PET flakes

50 kg of hot washed, clear post-consumer flakes were purchased from an European recycling company (Polyrecycling, Weinfelden Switzerland). The flakes were split into two barrels of 25 kg each. A solution of the following chemicals were given to each of the two barrels: Toluene (25 ml), chlorobenzene (25 ml), chloroform (25 ml), methyl salicylate (25 ml), phenyl cyclohexane (25 ml), benzophenone (25 g), and methyl stearate (25 g). Subsequently the barrels are sealed and stored for 7 days at 50 °C with daily agitation. The flakes were subsequently rinsed with 10% ethanol in order to remove contaminants on the surface of the flakes. All applied chemicals were supplied by Sigma-Aldrich, Steinheim Germany.

2.4. Process conditions and sample material

The process conditions of the different recycling trials applied within this study are given in Fig. 1. The following process conditions were applied: (a) hot washing: The contaminated flakes were treated for 10 min at 75 °C with detergents followed by cold rinsing and surface drying at 140 °C for 30 s. (b) Cold washing: 5 min at 50 °C without detergents followed by cold rinsing and surface drying at 140 °C for 30 s. (c) solid state reaction (SSP): The PET flakes were treated at 190 °C under inert gas stream for residence times up to about 6 h. Also SSP trials at 180 °C as well as 200 °C were performed in order to investigate the influence of the decontamination temperature. All solid stating trials are performed in laboratory scale with 370 g of contaminated flakes per recycling trial. For each of the above mentioned process conditions, one recycling trial was performed. After each residence time one sample was drawn and analysed twice.

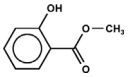
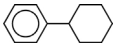
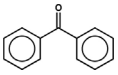
2.5. HFIP-extraction of the PET material

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, Sigma_Aldrich, Steinheim, Germany) 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.

2.6. GC/FID analysis and GC/ECD analysis

The HFIP extracts were analysed by gas chromatography with an FID detector for surrogates except chloroform, which was monitored by an ECD detector. For this purpose a gas chromatograph

Table 1
Model contaminants for the challenge test.

Surrogate	M_W[a]	Structure	Functional Group	Physical properties
Toluene	92.1		aromatic hydrocarbon	volatile, non-polar
Chlorobenzene	112.6		halogenated aromatic hydrocarbon	volatile, medium-polar, aggressive to PET
Chloroform	119.4	$CHCl_3$	halogenated hydrocarbon	volatile, polar, aggressive towards PET
Methyl salicylate	152.2		aromatic ester	non-volatile, polar
Phenyl cyclohexane	160.3		aromatic hydrocarbon	non-volatile, non-polar
Benzophenone	182.2		aromatic ketone	non-volatile, polar
Methyl stearate	298.5	$CH_3(CH_2)_{16}COOCH_3$	aliphatic ester	non-volatile, polar

[a] Molecular weight in g mol.

Table 2
Analytical detection limit of the surrogates in PET samples.

surrogate	detection limit [mg/kg]
toluene	0.1
chloroform	0.2
chlorobenzene	0.1
methyl salicylate	0.1
phenyl cyclohexane	0.1
benzophenone	0.1
methyl stearate	0.2

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}.

The detection limits were determined according to DIN 32645. The results are given in Table 2.

3. Results and discussion

The recycling trials within this study were performed in laboratory scale. In a previous study (Welle, 2014b) it could be shown, that the diffusion process in an individual contaminated flake or pellet is very similar as for the whole bulk of contaminated flakes. The efficiency of the decontamination process is mainly depend on the applied temperatures. It could be shown that the diffusion processes of the contaminants in the artificial contaminated flakes are following the Fickian laws of diffusion. Therefore, the cleaning efficiency determined in a couple of PET flakes or pellets is very similar to the cleaning efficiency of the whole recycled batch as long as flake or pellet size is similar. As a consequence the results from laboratory or pilot plants studies are also similar to cleaning efficiency data from industrial scale PET recycling lines.

Within this study, ten challenge tests with different process steps and process parameters were performed (Fig. 1). For this purpose clear washed PET flakes were contaminated according to the standard protocol published in the scientific literature (Franz et al., 2004a,b). One part of the challenge tests were carried out with only contaminated PET material, which means that only contaminated green flakes were treated under the recycling conditions. Cross-contamination cannot occur in this case, because all flakes have the same contamination level. These challenge tests can be considered as reference tests. The cleaning efficiencies of these tests should be the lowest because the highest loads of chemicals are entering the PET recycling process. Another set of challenge tests were performed with a certain amount of contaminated green flakes while running the process together with non-contaminated clear flakes. These tests can be considered as the typical procedure for challenging industrial scale PET recycling lines. The comparison of the results of the challenge tests allows conclusions on the amount of cross-contamination.

The concentrations of the contaminated clear flakes are given in Table 3. The concentrations given in Table 3 can be considered as initial concentrations for all individual challenge test trials A to G. Fig. 1 summarizes the different challenge tests and the applied process conditions. The results for the individual decontamination trials are given in Tables 4–13.

3.1. The comparison of the decontamination processes shows the following results

3.1.1. Comparison of the results of trials A, D2 and F

The comparison of the recycling trials D2 and F shows the influence of the washing temperature on the efficiency of the subsequent decontamination process at a temperature of 190 °C. 190 °C is a typical temperature used for decontamination in a solid-state PET recycling process (SSP). The residual concentrations of the investigated surrogates in the first SSP sample after a residence time of 25 min are significantly lower after hot washing of the contaminated PET flakes, especially for the high molecular weight surrogates like benzophenone and methyl stearate. This indicate, that the surrogates near the surface of the PET flakes more efficiently reduced during hot washing of the contaminated PET flakes compared to cold washing. However, after a decontamination time of 55 min this effect has been vanished and the residual concentration in the samples after hot washing and cold washing, respectively, are similar. The comparison of the results of trial A (non-washed flakes) with trial D2 (hot washed flakes) shows the same effect. The residual concentrations in non-washed flakes in trial A are significantly higher than found in trial D2 with hot washed flakes. Non-washed flakes had therefore a higher concentration of the surrogates near the surface of the PET flakes, which can be faster decontaminated. However, after a residence time of 115 min this effect is nearly vanished and the residual concentrations of the investigated surrogates in both recycling trials are similar.

Table 3
Concentrations of the surrogates in the investigated flakes samples (contaminated samples).

Sample	Concentration [mg/kg]						
	Toluene	Chloroform	Chlorobenzene	Phenyl cyclohexane	Methyl salicylate	Benzo-phenone	Methyl stearate
contaminated flakes	444.0 ± 2.9	409.0 ± 2.0	750.3 ± 3.4	844.9 ± 5.5	985.0 ± 6.8	808.8 ± 3.9	941.4 ± 4.2
contaminated flakes	474.3 ± 5.6	418.8 ± 3.2	816.4 ± 9.0	925.6 ± 8.7	1028.7 ± 9.5	843.9 ± 5.7	1063.6 ± 6.0
contaminated flakes	463.4 ± 1.3	425.0 ± 1.4	796.7 ± 1.3	857.7 ± 2.7	975.7 ± 0.8	803.0 ± 3.7	1078.0 ± 1.5
contaminated flakes	472.9 ± 3.9	424.3 ± 2.5	811.5 ± 5.2	810.1 ± 7.1	956.3 ± 7.2	748.2 ± 5.1	898.8 ± 6.1
mean contaminated flakes (standard deviation)	463.7 ± 14.0 (3.0%)	419.3 ± 7.4 (1.8%)	793.7 ± 30.1 (3.8%)	859.6 ± 48.4 (5.6%)	986.4 ± 30.6 (3.1%)	801.0 ± 39.6 (4.9%)	995.5 ± 88.9 (8.9%)
contaminated flakes, rinsed	390.9 ± 1.4	401.7 ± 1.3	669.5 ± 1.7	617.6 ± 2.0	821.9 ± 3.8	683.3 ± 0.7	728.0 ± 3.9
contaminated flakes, rinsed	378.6 ± 0.8	395.7 ± 0.4	645.2 ± 1.6	615.4 ± 1.6	834.6 ± 2.2	721.3 ± 2.0	898.0 ± 3.3
contaminated flakes, rinsed	413.9 ± 2.4	407.2 ± 0.8	717.0 ± 3.2	674.4 ± 1.8	933.7 ± 3.4	803.4 ± 1.2	966.2 ± 2.6
contaminated flakes, rinsed	434.7 ± 2.2	413.9 ± 1.6	744.6 ± 4.0	671.4 ± 2.5	948.1 ± 3.0	789.0 ± 1.1	821.5 ± 2.1
mean contaminated flakes after rinsing (standard deviation)	404.5 ± 24.9 (6.1%)	404.6 ± 7.8 (1.9%)	694.1 ± 45.0 (6.5%)	644.7 ± 32.6 (5.1%)	884.6 ± 65.5 (8.7%)	749.3 ± 56.7 (7.6%)	853.4 ± 102.4 (12.0%)

3.1.2. Comparison of the results of trials D1 to D3

The comparison of the recycling trials D1 to D3 shows the influence of temperature on the decontamination process (SSP). As expected, lower decontamination temperatures lead to slightly higher residual concentrations in the investigated contaminated flake samples or vice versa to lower cleaning efficiencies. However, there is overall only a little effect of temperature on the cleaning efficiency of the decontamination process.

3.1.3. Comparison of the results of trials G1 to G3

The comparison between the recycling trials G1 to G3 shows the influence of the ratio between contaminated and non-contaminated flakes. The residual concentrations of the surrogates in the contaminated clear flakes in trials G1 and G3 are very similar. The mixing ratio (1:1 trial in G1 and 1:50 in trial G3) do not have a significant influence on the cleaning efficiency of the recycling process. Trial G2 shows slightly higher residual concentrations for the high volatile substances (toluene to methyl salicylate) in comparison to trials G1 and G3. Methyl stearate on the other hand was determined in slightly lower concentration compared to the other two trials within these series.

Regarding cross-contamination, trial G1 (1:1 mixing with non-contaminated flakes) shows the highest concentrations of the surrogates in the non-contaminated flakes. For benzophenone cross-contamination of 14% up to 22% was measured. The percentage of the methyl stearate within the non-contaminated flakes increased from about 5% after 25 min up to 25% at 115 min. These results clearly indicate, that cross-contamination can be determined within these samples. For the high volatile surrogates like chloroform, chlorobenzene, phenyl cyclohexane and methyl salicylate the cross-contamination is significantly lower. For chloroform, toluene and chlorobenzene cross-contamination could not be determined at all. The concentrations of the solvents were below the analytical detection limits in the non-contaminated flakes. Only in the samples drawn after 25 min, about 2% to 3% cross-contamination could be determined for phenyl cyclohexane and methyl salicylate. Increasing the mixing ratio from 1:1 up to 1:10 reduces significantly the cross-contamination. In trial G2 cross-contamination is only determined for benzophenone and methyl stearate in levels of about 1% to 2%. At residence times of >85 min cross-contamination was not detectable in these recycling trials. Following this trend, at a mixing ratio of 1:50 cross-contamination could not be determined at all.

3.1.4. Comparison of the results of trials B, C and G1

The comparison of the recycling trials B, C and G1 shows the influence of the washing step on the cross-contamination. Trials B, C and G1 are mixed with non-contaminated flakes before solid stating in a mixing ratio of 1:1. As mentioned above, the 1:1 mixing ratio shows the highest level of cross-contamination. Trial B was performed without a washing step. Trial C and G1 were performed with a hot wash and cold wash step, respectively, before decontamination in the SSP reactor. As a result, the highest level of cross-contamination was determined for trial B (non-washed flakes). For benzophenone cross-contamination was measured between 15% and about 29%. Cross-contamination for methyl stearate was slightly lower in the range of 4% up to about 20%. For phenyl cyclohexane and methyl stearate cross-contamination was determined only at very short residence times of 25 min with levels of 1.1% and 3.3%, respectively. Both recycling trials with washed flakes show a lower percentage of cross-contamination. For benzophenone between 7% and 18% of the concentrations determined in the contaminated flakes were found in the non-contaminated flakes. For methyl stearate the level of cross-contamination is between 4% and about 10%. It is interesting to note, that the cross-contamination of phenyl cyclohexane and methyl salicylate after a

Table 4
Results of the quantification of the applied surrogates in Trial A (contaminated flakes followed by SSP 190 °C).

Sample	Concentration [mg/kg]						
	Toluene	Chloroform	Chlorobenzene	Phenyl cyclohexane	Methyl salicylate	Benzo-phenone	Methyl stearate
SSP 25 min, green flakes	2.7 ± 0.1	8.8 ± 0.1	6.4 ± 0.1	5.0 ± 0.1	10.4 ± 0.1	87.5 ± 0.6	105.4 ± 0.2
SSP 55 min, green flakes	0.4 ± 0.1	1.8 ± 0.1	1.4 ± 0.1	5.0 ± 0.1	3.6 ± 0.1	29.3 ± 0.5	36.2 ± 0.9
SSP 85 min, green flakes	0.3 ± 0.1	1.6 ± 0.1	1.3 ± 0.1	4.6 ± 0.1	3.4 ± 0.1	12.9 ± 0.1	14.2 ± 0.1
SSP 115 min, green flakes	0.5 ± 0.1	0.8 ± 0.1	0.6 ± 0.1	1.9 ± 0.1	0.9 ± 0.1	5.0 ± 0.1	4.5 ± 0.1
SSP 175 min, green flakes	<0.1	0.2 ± 0.1	0.2 ± 0.1	1.2 ± 0.1	0.3 ± 0.1	2.8 ± 0.1	2.5 ± 0.1
SSP 355 min, green flakes	0.8 ± 0.1	<0.2	<0.1	0.7 ± 0.1	<0.1	0.9 ± 0.1	0.6 ± 0.1

Table 5
Results of the quantification of the applied surrogates in Trial B (contaminated flakes, mixed with non-contaminated green flakes (1:1) followed by SSP 190 °C).

Sample	Concentration [mg/kg] (percentage cross-contamination)						
	Toluene	Chloroform	Chlorobenzene	Phenyl cyclohexane	Methyl salicylate	Benzo-phenone	Methyl stearate
SSP 25 min, green flakes	1.4 ± 0.1	6.8 ± 0.1	4.8 ± 0.1	9.4 ± 0.1	9.2 ± 0.1	47.2 ± 0.6	96.0 ± 1.6
SSP 55 min, greenflakes	<0.1	1.2 ± 0.1	0.8 ± 0.1	2.4 ± 0.1	1.6 ± 0.1	10.3 ± 0.2	18.6 ± 0.2
SSP 85 min, greenflakes	0.5 ± 0.1	0.7 ± 0.1	0.6 ± 0.1	2.2 ± 0.1	1.3 ± 0.1	4.9 ± 0.1	8.7 ± 0.2
SSP 115 min, greenflakes	0.2 ± 0.1	0.9 ± 0.1	0.8 ± 0.1	3.1 ± 0.1	1.7 ± 0.1	5.3 ± 0.1	5.5 ± 0.1
SSP 175 min, greenflakes	<0.1	0.2 ± 0.1	0.2 ± 0.1	1.7 ± 0.1	<0.1	2.8 ± 0.1	4.6 ± 0.1
SSP 355 min, greenflakes	<0.1	<0.2	<0.1	<0.1	<0.1	<0.1	0.3 ± 0.1
SSP 25 min, clear flakes	<0.1	<0.2	<0.1	0.1 ± 0.1 (1.1%)	0.3 ± 0.1 (3.3%)	9.9 ± 0.1 (21.0%)	9.6 ± 0.1 (9.0%)
SSP 55 min, clear flakes	<0.1	<0.2	<0.1	<0.1	<0.1	2.4 ± 0.1 (23.3%)	2.6 ± 0.1 (14.0%)
SSP 85 min, clear flakes	<0.1	<0.2	<0.1	<0.1	<0.1	1.4 ± 0.1 (28.6%)	1.7 ± 0.1 (19.5%)
SSP 115 min, clear flakes	<0.1	<0.2	<0.1	<0.1	<0.1	0.8 ± 0.1 (15.1%)	1.0 ± 0.1 (18.2%)
SSP 175 min, clear flakes	<0.1	<0.2	<0.1	<0.1	<0.1	<0.1	0.2 ± 0.1 (4.3%)
SSP 355 min, clear flakes	<0.1	<0.2	<0.1	<0.1	<0.1	<0.1	<0.1

Table 6
Results of the quantification of the applied surrogates in Trial C (contaminated flakes, hot washed and rinsed, mixed with non-contaminated green flakes (1:1) followed by SSP 190 °C).

Sample	Concentration [mg/kg] (percentage cross-contamination)						
	Toluene	Chloroform	Chlorobenzene	Phenyl cyclohexane	Methyl salicylate	Benzo-phenone	Methyl stearate
SSP 25 min, green flakes	2.3 ± 0.1	7.5 ± 0.1	6.0 ± 0.1	9.9 ± 0.1	11.7 ± 0.1	30.8 ± 0.3	37.2 ± 0.4
SSP 55 min, green flakes	0.2 ± 0.1	1.8 ± 0.1	1.2 ± 0.1	3.6 ± 0.1	3.1 ± 0.1	10.5 ± 0.1	16.2 ± 0.2
SSP 85 min, green flakes	<0.1	0.6 ± 0.1	0.5 ± 0.1	1.7 ± 0.1	0.9 ± 0.1	3.9 ± 0.1	5.2 ± 0.2
SSP 115 min, green flakes	<0.1	<0.2	<0.1	0.6 ± 0.1	<0.1	1.9 ± 0.1	5.7 ± 0.1
SSP 175 min, green flakes	<0.1	0.4 ± 0.1	0.4 ± 0.1	1.9 ± 0.1	0.5 ± 0.1	3.4 ± 0.1	4.4 ± 0.1
SSP 355 min, green flakes	<0.1	<0.2	<0.1	<0.1	<0.1	<0.1	0.2 ± 0.2
SSP 25 min, clear flakes	<0.1	<0.2	<0.1	3.0 ± 0.1 (30.3%)	3.0 ± 0.1 (25.6%)	5.6 ± 0.1 (18.2%)	2.9 ± 0.1 (7.8%)
SSP 55 min, clear flakes	<0.1	<0.2	<0.1	<0.1	<0.1	0.7 ± 0.1 (6.7%)	0.7 ± 0.1 (4.3%)
SSP 85 min, clear flakes	<0.1	<0.2	<0.1	<0.1	<0.1	0.5 ± 0.1 (12.8%)	0.5 ± 0.1 (9.6%)
SSP 115 min, clear flakes	<0.1	<0.2	<0.1	<0.1	<0.1	0.2 ± 0.1 (10.5%)	0.2 ± 0.1 (3.5%)
SSP 175 min, clear flakes	<0.1	<0.2	<0.1	<0.1	<0.1	<0.1	<0.1
SSP 355 min, clear flakes	<0.1	<0.2	<0.1	<0.1	<0.1	<0.1	<0.1

Table 7
Results of the quantification of the applied surrogates in Trial D1 (contaminated flakes, hot washed and rinsed, followed by SSP 180 °C).

Sample	Concentration [mg/kg]						
	Toluene	Chloroform	Chlorobenzene	Phenyl cyclohexane	Methyl salicylate	Benzo-phenone	Methyl stearate
SSP 25 min, green flakes	3.3 ± 0.1	11.3 ± 0.1	9.2 ± 0.1	2.9 ± 0.1	11.0 ± 0.1	45.9 ± 0.6	89.2 ± 8.4
SSP 55 min, green flakes	<0.1	2.6 ± 0.1	1.8 ± 0.1	2.9 ± 0.1	3.2 ± 0.1	11.4 ± 0.1	14.7 ± 0.3
SSP 85 min, green flakes	<0.1	1.2 ± 0.1	1.0 ± 0.1	3.3 ± 0.1	2.4 ± 0.1	7.6 ± 0.1	8.3 ± 0.3
SSP 115 min, green flakes	<0.1	0.6 ± 0.1	0.4 ± 0.1	2.0 ± 0.1	1.2 ± 0.1	4.7 ± 0.1	7.9 ± 0.3
SSP 175 min, green flakes	<0.1	0.6 ± 0.1	0.4 ± 0.1	2.8 ± 0.1	0.9 ± 0.1	4.0 ± 0.1	3.9 ± 0.1
SSP 355 min, green flakes	<0.1	<0.2	<0.1	0.3 ± 0.1	<0.1	0.6 ± 0.1	0.8 ± 0.1

Table 8
Results of the quantification of the applied surrogates in Trial D2 (contaminated flakes, hot washed and rinsed, followed by SSP 190 °C).

Sample	Concentration [mg/kg]						
	Toluene	Chloroform	Chlorobenzene	Phenyl cyclohexane	Methyl salicylate	Benzo-phenone	Methyl stearate
SSP 25 min, green flakes	1.8 ± 0.1	6.4 ± 0.1	4.8 ± 0.1	4.9 ± 0.1	7.4 ± 0.2	29.4 ± 0.7	35.9 ± 1.5
SSP 55 min, green flakes	<0.1	1.1 ± 0.1	0.6 ± 0.1	2.1 ± 0.1	1.6 ± 0.1	10.2 ± 0.2	20.7 ± 0.7
SSP 85 min, green flakes	<0.1	0.3 ± 0.1	<0.1	1.5 ± 0.1	0.6 ± 0.1	5.3 ± 0.1	9.7 ± 0.2
SSP 115 min, green flakes	<0.1	0.7 ± 0.1	0.5 ± 0.1	4.2 ± 0.1	1.5 ± 0.1	6.0 ± 0.1	5.0 ± 0.1
SSP 175 min, green flakes	<0.1	<0.2	<0.1	0.3 ± 0.1	<0.1	1.0 ± 0.1	2.0 ± 0.1
SSP 355 min, green flakes	<0.1	<0.2	<0.1	0.2 ± 0.1	<0.1	0.2 ± 0.1	0.2 ± 0.1

Table 9
Results of the quantification of the applied surrogates in Trial D3 (contaminated flakes, hot washed and rinsed, followed by SSP 200 °C).

Sample	Concentration [mg/kg]						
	Toluene	Chloroform	Chlorobenzene	Phenyl cyclohexane	Methyl salicylate	Benzo-phenone	Methyl stearate
SSP 25 min, green flakes	0.9 ± 0.1	4.2 ± 0.1	3.0 ± 0.1	3.3 ± 0.1	4.4 ± 0.1	24.8 ± 0.1	31.8 ± 0.1
SSP 55 min, green flakes	<0.1	0.4 ± 0.1	<0.1	1.4 ± 0.1	0.6 ± 0.1	6.7 ± 0.1	15.4 ± 0.2
SSP 85 min, green flakes	<0.1	<0.2	<0.1	0.4 ± 0.1	0.1 ± 0.1	1.4 ± 0.1	1.4 ± 0.1
SSP 115 min, green flakes	<0.1	<0.2	<0.1	0.4 ± 0.1	0.3 ± 0.1	1.3 ± 0.1	2.6 ± 0.1
SSP 175 min, green flakes	<0.1	<0.2	<0.1	0.1 ± 0.1	<0.1	0.2 ± 0.1	0.3 ± 0.1
SSP 355 min, green flakes	<0.1	<0.2	<0.1	0.2 ± 0.1	<0.1	0.3 ± 0.1	<0.2

Table 10
Results of the quantification of the applied surrogates in Trial F (contaminated flakes, cold washed and rinsed, followed by SSP 190 °C).

Sample	Concentration [mg/kg]						
	Toluene	Chloroform	Chlorobenzene	Phenyl cyclohexane	Methyl salicylate	Benzo-phenone	Methyl stearate
SSP 25 min, green flakes	2.7 ± 0.1	7.3 ± 0.1	5.9 ± 0.1	8.5 ± 0.2	10.0 ± 0.1	35.9 ± 0.5	52.0 ± 1.2
SSP 55 min, green flakes	<0.1	0.8 ± 0.1	0.4 ± 0.1	1.7 ± 0.1	1.3 ± 0.1	10.0 ± 0.1	25.6 ± 0.4
SSP 85 min, green flakes	<0.1	0.7 ± 0.1	0.5 ± 0.1	3.0 ± 0.1	1.3 ± 0.1	6.0 ± 0.1	7.8 ± 0.1
SSP 115 min, green flakes	<0.1	0.7 ± 0.1	0.5 ± 0.1	4.1 ± 0.1	1.2 ± 0.1	6.5 ± 0.1	4.7 ± 0.1
SSP 175 min, green flakes	<0.1	0.7 ± 0.1	0.5 ± 0.1	2.6 ± 0.1	0.6 ± 0.1	3.5 ± 0.1	1.9 ± 0.1
SSP 355 min, green flakes	<0.1	<0.2	<0.1	<0.1	<0.1	0.1 ± 0.1	0.2 ± 0.1

Table 11
Results of the quantification of the applied surrogates in Trial G1 (contaminated flakes, cold washed and rinsed, mixed with green flakes (1:1) followed by SSP 190 °C).

Sample	Concentration [mg/kg] (percentage cross-contamination)						
	Toluene	Chloroform	Chlorobenzene	Phenyl cyclohexane	Methyl salicylate	Benzo-phenone	Methyl stearate
SSP 25 min, green flakes	n.d.	8.5 ± 0.5	7.1 ± 0.3	6.7 ± 0.2	6.9 ± 0.1	32.2 ± 0.6	66.8 ± 1.6
SSP 55 min, green flakes	n.d.	1.1 ± 0.1	0.7 ± 0.1	0.7 ± 0.1	0.6 ± 0.1	6.4 ± 0.1	11.8 ± 0.1
SSP 85 min, green flakes	n.d.	0.5 ± 0.1	0.3 ± 0.1	1.9 ± 0.1	0.8 ± 0.1	5.2 ± 0.1	7.1 ± 0.1
SSP 115 min, green flakes	n.d.	0.2 ± 0.1	0.1 ± 0.1	0.9 ± 0.1	0.3 ± 0.1	1.7 ± 0.1	1.6 ± 0.1
SSP 25 min, clear flakes	n.d.	<0.2	<0.1	0.1 ± 0.1 (1.5%)	0.2 ± 0.1 (2.9%)	4.2 ± 0.1 (13.0%)	3.6 ± 0.1 (5.4%)
SSP 55 min, clear flakes	n.d.	<0.2	<0.1	<0.1	<0.1	1.4 ± 0.1 (21.9%)	1.0 ± 0.1 (8.5%)
SSP 85 min, clear flakes	n.d.	<0.2	<0.1	<0.1	<0.1	0.7 ± 0.1 (13.5%)	0.8 ± 0.1 (11.3%)
SSP 115 min, clear flakes	n.d.	<0.2	<0.1	<0.1	<0.1	0.3 ± 0.1 (17.6%)	0.4 ± 0.1 (25.0%)

Table 12
Results of the quantification of the applied surrogates in Trial G2 (contaminated flakes, cold washed and rinsed, mixed with green flakes (1:10) followed by SSP 190 °C).

Sample	Concentration [mg/kg] (percentage cross-contamination)						
	Toluene	Chloroform	Chlorobenzene	Phenyl cyclohexane	Methyl salicylate	Benzo-phenone	Methyl stearate
SSP 25 min, green flakes	n.d.	10.5 ± 0.2	9.8 ± 0.1	11.7 ± 0.2	12.2 ± 0.2	34.4 ± 0.5	52.6 ± 0.7
SSP 55 min, green flakes	n.d.	1.5 ± 0.1	1.1 ± 0.1	4.3 ± 0.1	3.0 ± 0.1	10.3 ± 0.1	14.5 ± 0.1
SSP 85 min, green flakes	n.d.	0.7 ± 0.1	0.6 ± 0.1	3.4 ± 0.1	1.8 ± 0.1	6.6 ± 0.1	6.5 ± 0.1
SSP 115 min, green flakes	n.d.	0.3 ± 0.1	0.2 ± 0.1	1.2 ± 0.1	0.4 ± 0.1	2.5 ± 0.1	4.0 ± 0.1
SSP 25 min, clear flakes	n.d.	<0.2	<0.1	<0.1	<0.1	0.7 ± 0.1 (2.0%)	0.5 ± 0.1 (1.0%)
SSP 55 min, clear flakes	n.d.	<0.2	<0.1	<0.1	<0.1	<0.1	0.2 ± 0.1 (1.4%)
SSP 85 min, clear flakes	n.d.	<0.2	<0.1	<0.1	<0.1	<0.1	<0.1
SSP 115 min, clear flakes	n.d.	<0.2	<0.1	<0.1	<0.1	<0.1	<0.1

Table 13
Results of the quantification of the applied surrogates in Trial G3 (contaminated flakes, cold washed and rinsed, mixed with green flakes (1:50) followed by SSP 190 °C).

Sample	Concentration [mg/kg]						
	Toluene	Chloroform	Chlorobenzene	Phenyl cyclohexane	Methyl salicylate	Benzo-phenone	Methyl stearate
SSP 25 min, green flakes	n.d.	8.2 ± 0.1	7.0 ± 0.1	6.8 ± 0.1	6.9 ± 0.1	28.5 ± 0.2	71.4 ± 0.1
SSP 55 min, green flakes	n.d.	1.4 ± 0.1	1.1 ± 0.1	3.4 ± 0.1	2.4 ± 0.1	8.1 ± 0.1	16.9 ± 0.1
SSP 85 min, green flakes	n.d.	0.3 ± 0.1	0.2 ± 0.1	0.8 ± 0.1	0.4 ± 0.1	2.2 ± 0.1	5.0 ± 0.1
SSP 115 min, green flakes	n.d.	0.4 ± 0.1	0.3 ± 0.1	1.2 ± 0.1	0.4 ± 0.1	2.2 ± 0.1	3.2 ± 0.1
SSP 25 min, clear flakes	n.d.	<0.2	<0.1	<0.1	<0.1	<0.1	<0.1
SSP 55 min, clear flakes	n.d.	<0.2	<0.1	<0.1	<0.1	<0.1	<0.1
SSP 85 min, clear flakes	n.d.	<0.2	<0.1	<0.1	<0.1	<0.1	<0.1
SSP 115 min, clear flakes	n.d.	<0.2	<0.1	<0.1	<0.1	<0.1	<0.1

residence time of 25 min is 30% and 26%, respectively. The reason for that high levels of cross-contamination is unclear. After 55 min the level is below the detection limit for both substances. The cross-contamination levels the recycling trials G1 (cold wash) is between trial B (non-washed flakes) and trial C (hot washed flakes).

Comparison of the results of trials A and B: The comparison of these two challenge test shows the influence of mixing of contaminated with non-contaminated flakes. These results show, that 1:1 mixing has only a slight effect on the decontamination of the contaminated flakes. The residual concentrations in recycling trial A are slightly higher as found in recycling trial B (1:1 mixing with non-contaminated flakes). After a decontamination time of 115 min this concentration effect has been vanished and both recycling trials show similar cleaning efficiencies.

4. Conclusions

Within the study the decontamination efficiencies of the model contaminants have been determined for ten different PET recycling process conditions in laboratory scale. In addition, the amount of cross-contamination between artificially contaminated and non-contaminated flakes has been determined. From the results and the comparison between the ten individual challenge tests of this study the following conclusions could be drawn:

- Higher decontamination temperatures result in higher cleaning efficiencies.
- Hot washing has a higher cleaning efficiency than cold wash processes. However, the washing temperature has a minor influence on the overall cleaning efficiency of the PET recycling process.
- Mixing of contaminated with non-contaminated flakes has no influence on the decontamination efficiency of the contaminated flakes.
- Cross-contamination is found at a mixing ratio of 1:1 especially for the low volatile surrogates like benzophenone and methyl stearate. Moving towards higher mixing level (1:10 or 1:50) cross-contamination do not play a significant role within the evaluation of the cleaning efficiency. Also volatile surrogates like chloroform, chlorobenzene, phenyl cyclohexane and methyl salicylate do not show significant cross-contamination.
- The highest amount of cross-contamination was found if the contaminated flakes were introduced into the decontamination process without a washing process. The lowest cross-contamination was found if the contaminated flakes are treated in a hot washing process before decontamination in the SSP.

It is important to note, that cross-contamination of the volatile surrogates like toluene and chlorobenzene lead to the reduced maximum recyclates amount within the cleaning efficiency evaluation. For low volatile surrogates like benzophenone and methyl stearate the cleaning efficiency was still sufficient even if a cross-contamination of 10% was assumed in the food law compliance evaluation, e.g. considering the EFSA evaluation criteria. The results of this study, however, clearly show that the volatile compounds do not have a significant contribution on cross-contamination. On the other hand, a significant cross-contamination was found at ratios of 1:1 between contaminated and non-contaminated PET flakes especially for non-volatile model compounds. At higher ratios, which were typically applied in challenge tests on pilot plant or industrial scale lines, cross-contamination do not play a significant role. Challenging a pilot or industrial scale plant with a limited amount of contaminated PET while running the plant with non-contaminated PET of a different colour will be a good option for cleaning efficiency evaluation. In addition, the results show that cross-contamination is negligible for volatile compounds. Therefore, it can be concluded that 10% of cross-contamination for each surrogate applied by EFSA in their opinion letters is extremely conservative.

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