Transport of chlorinated paraffins (CPs) from baking oven doors into the food

Jannik Sprengel [a], Stefanie Rixen [a], Oliver Kappenstein [b], Walter Vetter [a,*]

[a] *Institute of Food Chemistry, University of Hohenheim, Garbenstr. 28, D-70599 Stuttgart, Germany*
[b] *German Federal Institute for Risk Assessment, Max-Dohrn-Str. 8-10, D-10589 Berlin, Germany*

ARTICLE INFO

ABSTRACT

Chlorinated paraffins (CPs) have been repeatedly detected in the kitchen environment. Especially baking ovens were contaminated with high CP amounts on the insides of the doors. To investigate if CPs could be transferred into baked food, we spiked self-synthesized single chain C_{12}-CP and C_{15}-CP standards onto the inside door of an unused, CP-free baking oven. Experiments were performed under different conditions to assess possible CP transportation pathways. Coconut fat was used as food simulant, the exhaust air was monitored with cellulose filter paper and remaining CPs were collected via cotton wipes. In all experiments, both C_{12}- and C_{15}-CPs could be identified in both the food simulant and the cellulose samplers. Mean transfer rates into the food simulant amounted to 2.2% for C_{12}-CPs and 5.8% for C_{15}-CPs. Baking of food in CP-containing baking ovens may perceptibly increase the CP intake of consumers.

Introduction

Chlorinated paraffins (CPs) is the summarizing term for different industrial products consisting of thousands of different polychlorinated *n*-alkanes (Tomy, 2010). CPs are usually classified into three distinct subgroups according to the *n*-alkane chain range, i.e. short-chain chlorinated paraffins (SCCPs, C_{10}- to C_{13}-CPs), medium-chain chlorinated paraffins (MCCPs, C_{14}- to C_{17}-CPs) and long-chain chlorinated paraffins (LCCPs, $>C_{17}$-CPs). Although SCCPs were recently classified as persistent organic pollutants (POP) in Annex A of the Stockholm Convention (Conference of the Parties of the Stockholm Convention (COP.8), 2017), MCCPs and LCCPs are not yet regulated. However, the safety of MCCPs is increasingly disputed in recent years (Zellmer et al., 2020). In addition, these persistent high-volume production chemicals are found in environmental compartments from virtually all over the world (Tomy, 2010; van Mourik et al., 2016).

For human exposure, food (oral exposure) and the indoor environment (inhalative exposure) are the most important CP intake pathways (Fridén et al., 2011; Iino et al., 2005; Krätschmer et al., 2019). Several recent studies reported on the occurrence of CPs in the kitchen environment in which both exposure pathways are combined. E.g., CPs were detected in high concentrations in fat from kitchen hoods, accumulated in dishcloths during use, leaching into food from hand blenders and deposited on the inside of baking oven (BO) doors (Gallistl et al., 2017, 2018; Yuan et al., 2017; Bendig et al., 2013; EFSA Panel on Contaminants in the Food Chain (CONTAM) et al., 2020; Zellmer et al., 2020).

The high CP amounts (up to several mg) on the insides of BO doors which originated from the BO itself, raised the question whether CPs could be transferred into the baking goods during operation of the BO (Gallistl et al., 2018). Also, a recent study reported the loss and possible evaporation of CPs during the baking process (Perkons et al., 2019).

The goal in this study was to investigate if and to what extent CPs could be released from the inside of BO doors during the baking process, and whether they could be transferred into the food (e.g. baking good). In accordance with Article 1, 2c of Regulation (EC) No. 1935/2004 of the European Union (EU), BOs can be considered as a food contact material (FCM), because the transfer of substances from the BO inside to food can reasonably be expected during normal or foreseeable conditions of use (European Parliament, Council of the European Union, 2004). For this purpose, a BO was spiked with low amounts of CPs onto the inside of a BO door and then performed a conventional heating process. In order to exclude cross-contamination from other sources we used self-synthesized single chain CP standards whose compositions can be unequivocally distinguished from technical CP products. The baking good was simulated by placing four portions of coconut fat as food simulant and water (to create a vapor phase) into the BO, which were

---

* Corresponding author.
  E-mail address: walter.vetter@uni-hohenheim.de (W. Vetter).

analyzed after the heating process. Analyses were performed with gas chromatography coupled to electron capture negative ion mass spectrometry (GC/ECNI-MS) in selected ion monitoring (SIM) mode (Sprengel & Vetter, 2019).

Materials and methods

Standards

C_{12}-CPs (44.8%, 49.4%, 51.9%, 59.9% and 64.6% Cl), C_{15}-CPs (44.0%, 51.1%, 53.5%, 55.0% and 63.6% Cl) and C_{16}-CPs (44.6%, 50.9%, 53.1% and 58.9% Cl) were self-synthesized (Sprengel & Vetter, 2019; Sprengel et al., 2019). Perdeuterated α-hexachlorocyclohexane (α-PDHCH) was also synthesized in our working group (Vetter & Luckas, 1995). ε-Hexachlorocyclohexane (ε-HCH, 100 ng/μL) as well as PCB 158, PCB 186, PCB 194 and PCB 209 (all 10 ng/μL) were obtained from Dr. Ehrenstorfer (Augsburg, Germany).

Chemicals

Dichloromethane (DCM) and *n*-hexane (both for pesticide residue analysis grade) were ordered from Th. Geyer (Renningen, Germany), while 2,2,4-trimethylpentane (*i*-octane, for pesticide residue analysis grade) was obtained from Fluka Analytics (Seelze, Germany). Sulfuric acid (96–98%, *p.a.*) and cellulose filter paper were ordered from Carl Roth (Karlsruhe, Germany). Sodium sulfate (>99%, water free, *p.a.*) and silica gel 60 (*for column chromatography*) were produced by Sigma-Aldrich (Seelze, Germany). Native coconut fat was purchased in a retail shop and used as a food simulant in the BO experiments.

Baking oven (BO) experiments with CPs

Coconut fat was molten at 50 °C, and ~1 g each was spread inside four glass petri dishes (d = 10 cm). Additionally, 10 ng of PCB 209 was added to each petri dish. A fifth glass petri dish was filled with 30 mL of distilled water. The petri dishes were placed on an oven grate at medium height in a fixed pattern, i.e. the dish with water in the center and the four food simulants near the BO door and back as well as on the left and right side (Fig. 1). Diluted in 200 μL of *i*-octane, 215 μg of C_{12}-CPs (59.9% Cl) and 218 μg of C_{15}-CPs (53.5% Cl) were spiked onto the inside of the open BO door. The cellulose filter papers were pre-cleaned using accelerated solvent extraction (ASE) by means of a Dionex ASE 350

Back

Door

Fig. 1. Experimental setup of the petri dishes with the food simulant and distilled water inside the baking oven.

Accelerated Solvent Extractor (Thermo Fisher Scientific, Langenselbold, Germany) with conditions described elsewhere (Mandalakis et al., 2008). A stripe of the pre-cleaned cellulose filter paper was fixed on the outside of the oven, completely covering the fume extraction outlet at the front. Additional preparations are stated in the respective experiment. After preparation, the BO was operated at the stated experiment conditions. The experiments were:

- *Basic experiment (BE)*: BO operation at 180 °C for 90 min with top/bottom heat; samples were removed from the warm oven and directly extracted after cooling.
- *Variation "No Cleaning" (BE-NC)*: Same conditions as *BE*, but the BO door was neither cleaned nor spiked again with CPs after the previous *BE* experiment.
- *Variation "Over Night" (BE-ON)*: Same conditions as *BE*, but the samples were only removed/collected after cooling inside the oven for one night.
- *Variation "Circulating Air" (BE-CA)*: BO operation with circulated air instead of top/bottom heat.
- *Variation "Fat on BO door" (BE-Fat)*: Same conditions as *BE*. The CPs were spiked with ~70 mg of coconut fat onto the inside of the open BO door.
- *Variation "PCBs" (BE-PCB)*: Same conditions as *BE*, except PCB 209 was not spiked into the food simulant. Instead, 10 ng each of PCBs 158, 186, 194 and 209 were spiked together with the CPs on the BO door.
- *Variation "C_{16}-CPs" (BE-C_{16})*: Same conditions as *BE*. 21.8 μg/g of C_{16}-CPs (50.9% Cl) were spiked into the food simulant.
- *Variation "1 mg" (BE-1 mg)*: Same conditions as *BE*. Instead of 215 μg and 218 μg, 1075 μg and 1090 μg of C_{12}- and C_{15}-CPs were spiked onto the inside of the open BO door, respectively.

After finished operation, the cellulose filter paper and the petri dishes were removed from the oven. Additionally, a wipe test, using a cotton pad (d = 6 cm) obtained from a retail store, was performed on the inside of the oven door as described earlier (Gallistl et al., 2018). All experiments were performed in duplicate with a new and unused BO A, except *BE* which was carried out in triplicate. Additionally, experiments *BE*, *BE-ON* and *BE-CA* were repeated in duplicate with BO B, which was acquired in 2011. All samples were treated as described below.

Sample preparation

Wipe tests from BO doors were cold-extracted with *n*-hexane as described previously (Gallistl et al., 2018). The food simulant (coconut fat) was extracted from the petri dishes with 5 times 1 mL of *n*-hexane, while the cellulose filter papers were cut up into small pieces and cold-extracted with 3 times 25 mL of *n*-hexane. From that point on, all samples were treated in the same way. Extracted samples were concentrated to ~5 mL. Then, 10.7 ng of the recovery standard α-PDHCH and 5 mL of sulfuric acid were added to the sample extracts. After shaking vigorously for 30 s, phases were allowed to settle. For most food simulant extracts, separation could only be achieved by centrifugation (4500 rpm/2722 g, 5 min). The supernatant was transferred into a pear-shaped flask, and the residue was extracted twice again with 5 mL of *n*-hexane. The combined organic phases were evaporated to ~1 mL and subjected to column chromatography (glass column with i.d. = 1 cm, filled with a glass wool plug, 3 g of 30% deactivated silica gel and 0.5 g sodium sulfate) and polyhalogenated substances were eluted with 60 mL of *n*-hexane (Weichbrodt et al., 2000). After adding ~100 μL of *i*-octane as keeper, i.e. a small amount of a high-boiling solvent commonly used to prevent loss during evaporation (Dąbrowski, 2016; Oehme et al., 1994), the sample extracts were brought to a volume of 100 μL via rotary evaporation followed by a gentle stream of nitrogen. Before measurement, 10 ng of the internal standard ε-HCH was added to each solution.
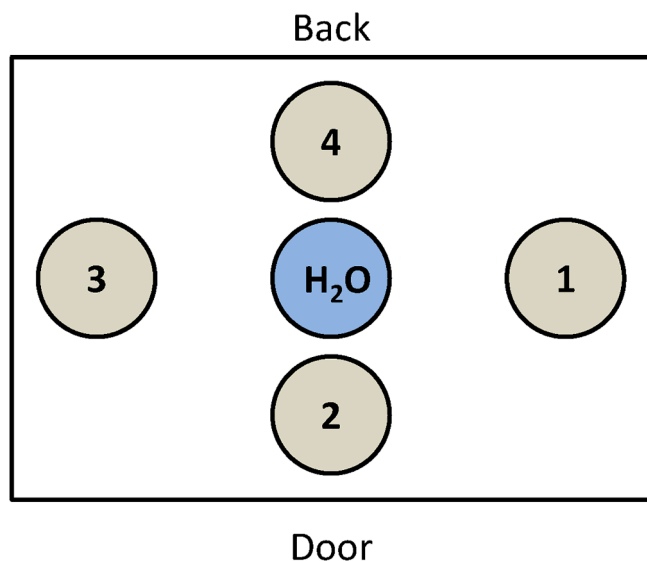
Instrumentation

Gas chromatography coupled with electron capture negative ion mass spectrometry (GC/ECNI-MS) measurements were performed on a 7890/5975C MSD system (Agilent, Waldbronn, Germany) using operation parameters described previously (Bendig et al., 2013). The GC was equipped with a 30 m × 0.25 mm i.d., 0.25 μm d_f Optima 5 MS column (Macherey-Nagel, Düren, Germany) and the MS was operated in selected ion monitoring (SIM) mode. CP quantification followed the method reported by Reth et al. (2005) with modifications described elsewhere (Sprengel & Vetter, 2019). CPs and PCBs were measured in GC/ECNI-MS-SIM mode by monitoring the two most abundant isotope peaks of the [M−Cl]⁻ fragment ion (Table S1). Calibration curves were established for self-synthesized single chain length CP mixtures, i.e. C_{12}-, C_{15}- and C_{16}-CPs of different degree of chlorination, respectively (instead of SCCP/MCCP mixtures). CP quantification by means of single chain-length CP standards is expected to negate quantification errors based on non-matching chain length composition (Sprengel & Vetter, 2019). Additional tests on the possible occurrence of chlorinated alkenes (chlorinated olefins, COs) were exemplarily performed by means of additional runs covering the [M−Cl]⁻ ions of the Cl_6-CPs and Cl_6-COs as well as Cl_7-CPs (Table S2). By comparing the retention time range of the monoisotopic peaks of the [M−Cl]⁻ fragment ions of Cl_6-CPs and Cl_6-COs with those of the Cl_7-CPs, we could exclude overlaps with [M−HCl−Cl]⁻ ions formed by CPs in addition/instead of [M−Cl]- fragment ions (Fig. S1). COs were considered to be present when the share of the resulting relative peak area of the COs in heated samples was higher than in the standards.

Quality assurance and quality control

Procedural blanks were prepared for each experiment. Additionally, blanks of unspiked coconut fat and unused, pre-cleaned cellulose filter paper were analyzed. Traces of C_{10}- and C_{11}-CPs were found to be inherent in the coconut fat. However, since only C_{12}-, C_{15}- and C_{16}-CPs were spiked into samples, blank corrections were not required. The recovery rate of the internal standard α-PDHCH used for monitoring the sample cleanup procedure was 74 ± 13%. CP spiking experiments of the coconut fat, performed in duplicate, resulted in recoveries of 80 ± 4%, 103 ± 4% and 90 ± 5% for C_{12}-CPs (59.9% Cl), C_{15}-CPs (53.5% Cl) and C_{16}-CPs (50.9% Cl), respectively. After each single experiment, the BO door was cleaned ten times with an *n*-hexane soaked wipe, at which point CPs were no longer detectable according to preliminary experiments. The basic experiment (*BE*) was performed in triplicate and all other experiments in duplicate. Values determined in replicate experiments, especially from wipes of the BO door, sometimes deviated strongly (>20%) from each other. Most likely, these variations were caused by slight and non-controllable changes in the performance (e.g. the distribution of the standard on the BO door, airflow in the oven etc.). Still, mean values were formed for these cases as well albeit only to compare general tends. No quantitative implications were derived from the CP amounts remaining on the BO door and on the cellulose stripes due to uncertainty resulting from the varying results. However, four food simulants were placed inside the oven per experimental setup. Statistically, there was no significant influence of the position of the food simulant on the CP amount that was transferred (t-Test, p≫0.05). Therefore, the CP transfer into food was effectively determined at n = 8 for each experiment, and the values could be considered robust in this case.

Results

Study design

A new and unused BO was initially tested to release no CPs during operation by means of wipe tests according to Gallistl et al. (2018).

Instead of technical CP mixtures, we chose two self-synthesized single chain CP standards (Sprengel & Vetter, 2019; Sprengel et al., 2019) for the spiking experiments. Accordingly, cross-contamination from other sources could be easily identified due to the unique chain length distribution. Likewise, additional screenings of several samples for other chain lengths (SCCPs and MCCPs) showed that the heating process did not cause chain breakage of CPs (Table S2, Supplementary Material). Therefore, restriction to two chain lengths allowed us to measure the samples in two GC runs (instead of eight runs required for SCCPs and MCCPs) by GC/ECNI-MS.

The use of food simulants is a common practice in testing food contact materials. Hence, coconut fat was chosen as food simulant due to the lack of polymerization processes in the BO, which made quantitative sample extraction possible and the sample treatment less time-consuming. Furthermore, release of 17 to 50 g water during the baking of products has been frequently described (Wilson et al., 2002; Xue et al., 2004; Xue & Walker, 2003; Rudan & Barbano, 1998). Therefore, a petri dish with 30 mL of distilled water was placed inside the oven to create water vapor similar to that released from baked food. After the experiment, no water was left in the petri dish and it was assumed to be released as vapor. To assess a possible evaporation of the food simulant, PCB 209 was spiked as recovery standard into the coconut fat used as food simulant. After the heating process, the bulk of PCB 209 (84 ± 19%) was still detected in the food simulant. However, low amounts of PCB 209 were detected on both the BO door and in the cellulose filter papers used as air samplers, indicating a slight release from the baking good. The cellulose filter paper was attached to cover the fume extraction outlet. From this outlet, the hot air from inside the oven is released mixed with cool air from the outside (HEA – Fachgemeinschaft für effiziente Energieanwendung e.V., 2020). However, neither the material nor the sampling conditions (constant warm air stream for 90 min) allowed for a quantitative sampling and the cellulose paper acted more as an indicator of airborne transfer.

Release of spiked CPs from the BO door

Fate of CPs spiked on the BO door

Of the 215 μg of C_{12}-CPs and 218 μg of C_{15}-CPs initially spiked onto the BO door, only < 1 μg of C_{12}-CPs but 42 μg of C_{15}-CPs were recovered on the BO door after the heating experiment (Table 1). Compared to that, a wipe test performed directly after spiking and without heating resulted in recovery rates of 74% C_{12}-CPs and 89% C_{15}-CPs. This indicated a strong CP release from the BO door during heating. In agreement with that, CP patterns in the remaining share were shifted to higher chlorine content compared to the standard initially applied (Fig. 2a, e). A second BO operation without thorough cleaning and re-spiking of the oven door (*BE-NC*), further reduced the amounts of C_{12}-and C_{15}-CPs on the BO door by about one order of magnitude (Table 1). Therefore, the BO door was thoroughly cleaned between each spiking experiment (section 2.6). A thorough wipe test of the complete inner area of the oven resulted in C_{12}- and C_{15}-CPs levels ≪ 1 μg (i.e. ≪1% of the initial amount), which was found to be negligible.

In the basic experiment (*BE*), the food simulant samples were immediately removed from the hot oven, similarly to normal practice in households. At this point, CPs could have been in the gas phase and opening of the door could promote their release via hot air. This possible effect was investigated by repeating the experiment without opening the door of the hot BO after usage. Instead, the BO was allowed to cool with closed door for 12 h (*BE-ON*). In this experiment, the amount detected on the BO door was almost twice as high as in the basic experiment (Table 1). This indicated that notable amounts of CPs, especially MCCPs (close to 25% of the contamination), actually remained inside the BO after conventional usage and may be released into the kitchen air when the hot BO is opened.

In addition to the basic experiment (*BE*) without air circulation, we conducted a further heating experiment using the air circulation mode

Table 1

Amounts of C$_{12}$-CPs and C$_{15}$-CPs [µg] found in food simulant (four times ~1 g of coconut fat each), the baking oven door (BO door) and an air sampler (stripe of cellulose filter paper) after performing eight spiking experiments (section 2.3). In all experiments except BE-1 mg, 215 µg of C$_{12}$-CPs and 218 µg of C$_{15}$-CPs were spiked on the BO door. In BE-1 mg, 1075 µg of C$_{12}$-CPs and 1090 µg of C$_{15}$-CPs were spiked on the BO door.

		food simulant		BO door		air sampler		sum (recovery)	
		C$_{12}$-CPs [µg]	C$_{15}$-CPs [µg]	C$_{12}$-CPs [µg]	C$_{15}$-CPs [µg]	C$_{12}$-CPs [µg]	C$_{15}$-CPs [µg]	C$_{12}$-CPs [µg]	C$_{15}$-CPs [µg]
BO-A	BE	4.6 ± 0.5	16 ± 4	0.45 ± 0.30	42 ± 21	0.54 ± 0.24	3.6 ± 0.5	5.6 (2.6%)	62 (28%)
	BE-ON	4.4 ± 0.2	15 ± 2	0.85 ± 0.21	94 ± 15	0.50 ± 0.01	3.2 ± 0.1	5.8 (2.7%)	110 (51%)
	BE-CA	3.0 ± 1.2	19 ± 4	nq	2.2 ± 1.5	0.51 ± 0.01	1.4 ± 0.1	3.5 (1.6%)	23 (10%)
	BE-NC	0.51 ± 0.1	1.2 ± 0.1	0.06 ± 0.04	2.0 ± 1.8	0.05 ± 0.01	0.30 ± 0.1	0.61 (0.2%)	3.5 (1.6%)
	BE-Fat	3.7 ± 0.3	7.3 ± 0.2	62 ± 5	231 ± 21	0.44 ± 0.21	0.99 ± 0.1	65 (31%)	240 (111%)
	BE-PCB	3.8 ± 0.3	16 ± 3	0.65 ± 0.24	56 ± 11	0.44 ± 0.12	3.1 ± 0.3	5.1 (2.4%)	76 (35%)
	BE-C$_{16}$	4.0 ± 0.3	18 ± 2	0.74 ± 0.31	85 ± 18	0.69 ± 0.09	3.9 ± 2.0	5.4 (2.5%)	110 (49%)
	BE-1 mg	72 ± 3.4	96 ± 12	120 ± 28	580 ± 16	6.5 ± 1.6	11 ± 2.1	200 (19%)	690 (63%)
BO-B	BE	2.3 ± 0.4	5.5 ± 1.2	nd	0.71 ± 0.42	1.0 ± 0.2	3.3 ± 1.1	3.3 (1.6%)	9.5 (4.4%)
	BE-ON	1.7 ± 0.1	6.1 ± 2.1	nd	1.6 ± 0.1	0.61 ± 0.04	3.2 ± 0.2	2.3 (1.0%)	11 (5.0%)
	BE-CA	0.75 ± 0.0	8.5 ± 1.6	nd	nd	0.13 ± 0.03	1.0 ± 0.3	0.88 (0.4%)	9.5 (4.4%)

nd = not detectable (<0.01 µg).
nq = not quantifiable (0.01 µg < x < 0.03 µg).

(*BE-CA*). Under *BE-CA* conditions, the CP amount detected on the BO door sharply dropped by one order of magnitude (Table 1). Namely, C$_{12}$-CPs were detectable, but below the limit of quantification while the amount of C$_{15}$-CPs dropped from 42 µg (*BE*) to 2.2 µg (*BE-CA*). Hence, the CP amounts on the BO door after use with air circulation (*BE-CA*) were similarly low as those obtained after a second heating without spiking (*BE-NC*) (Table 1).

When CPs were applied in a fat matrix on the BO door (here: 70 mg of coconut fat), the amount detected on the BO door after the heating roughly increased by two orders of magnitude in the case of C$_{12}$-CPs (*BE-Fat*, Table 1). However, the C$_{12}$-CPs pattern on the BO door showed an increase in lower chlorinated C$_{12}$-CPs, namely Cl$_6$- and Cl$_5$-homologues, contrasting the trend towards higher chlorinated homologs in the *BE* (Fig. 2d). This could be due to the formation of chlorinated alkenes (olefins) via dehydrochlorination in the fat matrix, which was observed in a previous study for CP-containing dough baked in a BO (Perkons et al., 2019).

Analysis of chlorinated alkenes (COs) in CP mixtures by low resolution GC/ECNI-MS is difficult due to the mass overlaps between COs and CPs. However, in the present case, i.e. using single chain CP standards only, *m/z* values of chlorinated alkanes and alkenes can be distinguished by means of the monoisotopic peak of the [M−Cl]⁻ fragment ions. From a related compound class, toxaphene, it was known that several congeners do also form [M−HCl−Cl]⁻ fragment ions (Vetter & Luckas, 1998), which interfere with COs bearing one Cl atom less. This type of fragmentation was also observed in the case of CPs (Krätschmer et al., 2018). However, these congeners are usually eluting at higher retention times. Recent publications also indicated that COs can be formed in hot GC injectors (>220 °C) and through in-source fragmentation (Schinkel et al., 2017a, 2017b). Consequently, the detection of *m/z* values corresponding with chlorinated alkenes in the standards must not necessarily mean that they are present. Therefore, our semiquantitative evaluation was based on changes in the abundance of the potential COs.

In wipes of the BO door after the experiment, much higher abundances were found for COs of both chain lengths. However, while a similar shift was observed for C$_{15}$-COs/C$_{15}$-CPs in the food simulant samples, the C$_{12}$-CO/C$_{12}$-CP ratio remained about the same. This indicated that the majority of the more volatile C$_{12}$-CPs (and C$_{12}$-COs) may have been released and removed from the BO in a short period, inhibiting a relevant transfer of COs into the food simulant. Not surprisingly, increased airflows (i.e. circulated air mode) resulted in lower shares of the more volatile COs in the sample (Table S4, SI). Interestingly, in *BE-Fat* the CO share was lower on the BO door (although still higher than in the single chain CP standard) but actually higher in the food simulant compared to *BE* conditions. This could indicate a preferred release of COs from the fatty matrix and therefore a more efficient transport into the food simulant.

For the C$_{15}$-CPs, the withholding effect of the fat matrix was even more pronounced: the initial amount of C$_{15}$-CPs was almost quantitatively conserved by the matrix on the BO door. Additionally, the chlorine content was lower for the C$_{15}$-CPs used, which may decrease susceptibility to dehydrochlorination. Consequently, the C$_{15}$-CP homolog composition on the BO door much more resembled the one of the standard which was in contrast to the basic experiment (*BE*, Fig. 2h). Such a fatty matrix has been frequently detected on the insides of BO doors after regular use in households (Gallistl et al., 2018).

All in all, amounts of C$_{15}$-CPs detected on the BO door after heating were higher than those of C$_{12}$-CPs, apparently due to their lower volatility, which can also be seen in the later GC elution range of C$_{15}$-CPs (here: 21–31 min) compared to C$_{12}$-CPs (here: 19–27 min) (Fig. S4). Generally, in the basic experiment, only a low share (0.45 µg SCCPs (<1%), 42 µg MCCPs (<20%)) could be recovered on the BO door. In addition, air circulation reduced the detectable CP amount and the presence of fat matrix increased the CP amount.
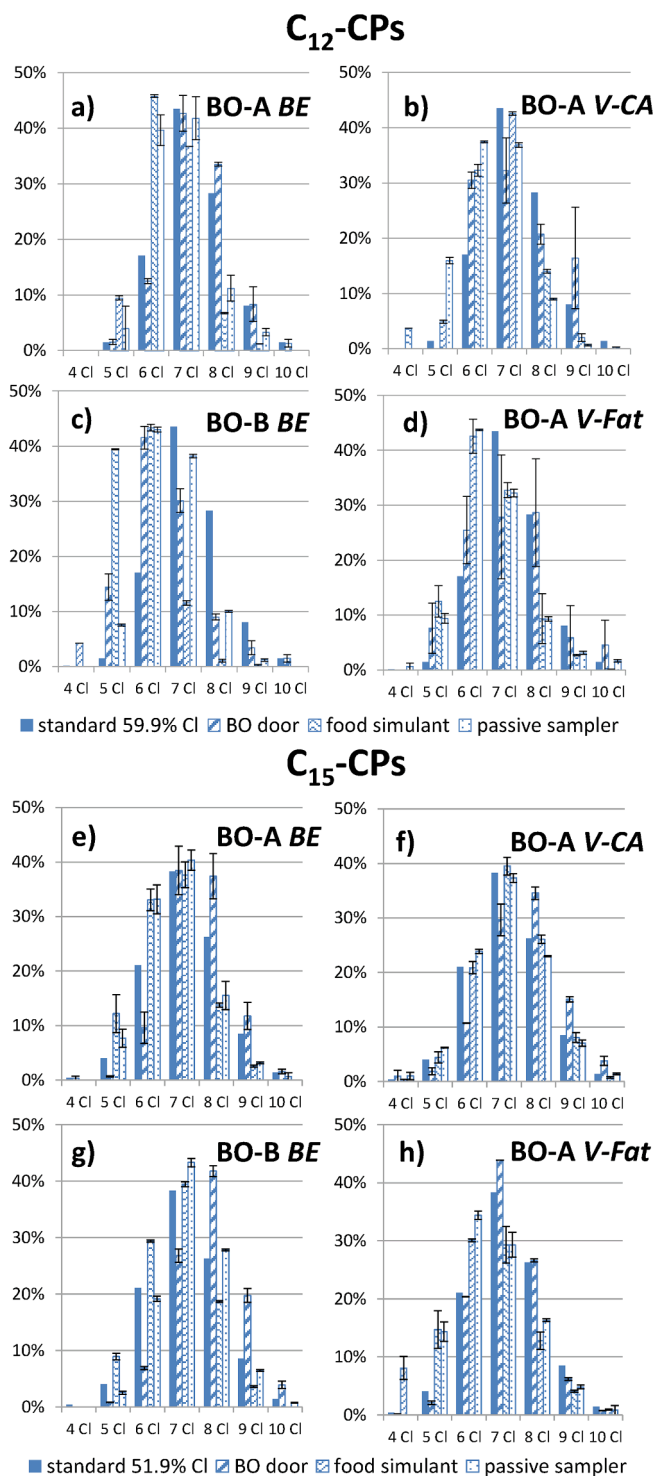
Evidence for CP release from the BO door into the kitchen environment

Both C$_{12}$- and C$_{15}$-CPs were detected in the sub-µg to low µg range on the cellulose filter paper (Table 1). Although this kind of sampler was not suitable for quantitative evaluation (see experimental part), the qualitative detection clearly demonstrated that CPs were partially released from the BO during heating. Such release was not surprising, because it is a common experience that the odor of the baking good can be perceived during the use of the BO, which verifies the release of volatile compounds. While cooling overnight (*BE-ON*) had no impact on the detected amount, lower CP amounts on the air sampler when applying a fat matrix confirmed a lower CP release due to the withholding effect. However, lower CP amounts were also recovered from the cellulose filter paper in circulating air (*BE-CA*) mode although higher CP amounts were expected. This attested the limitations of this sampler use for quantitative conclusions. However, more hermetic sampling approaches would have changed the airflows in the BO, which was omitted in order to keep conditions as realistic as possible.

Detection of CPs in the food simulant (coconut fat)

In the *BE*, 4.6 µg C$_{12}$-CPs and even 16 µg C$_{15}$-CPs were detected in the food simulant (coconut fat) after the heating experiment (shared amount in four 1 g portions with comparable separate CP amounts) (Table 1, Fig. 3). These amounts corresponded with transfer rates (TR) of 2.1% C$_{12}$-CPs and 7.3% C$_{15}$-CPs (Table 2). Compared to the standards applied, the CP pattern of both chain lengths was shifted to lower chlorinated (and thus more volatile) homologs (Fig. 2a,e). After a second use

C$_{12}$-CPs

standard 59.9% Cl ▨ **BO door** ▥ **food simulant** ▢ **passive sampler**

C$_{15}$-CPs

standard 51.9% Cl ▨ **BO door** ▥ **food simulant** ▢ **passive sampler**

Fig. 2. Homolog patterns of C$_{12}$-CPs and C$_{15}$-CPs found in the food simulant, wipes of the BO door and the cellulose filter paper used as air sampler in four experiments performed on two baking oven (BO-A and BO-B). C$_{12}$-CPs: a) BO-A (*BE*), b) BO-A (*BE-CA*), c) BO-B (*BE*) and d) BO-A (*BE-Fat*); C$_{15}$-CPs: e) BO-A (*BE*), f) BO-A (*BE-CA*), g) BO-B (*BE*) and h) BO-A (*BE-Fat*). Error bars indicate the mean variation of the homolog distribution. CP homolog patterns of all conducted experiments are found in the Supplementary Material (Fig. S3, S4).

without cleaning and re-spiking the BO but new food simulant (*BE-NC*), still 0.51 µg C$_{12}$-CPs and 1.2 µg C$_{15}$-CPs were detected in the food simulant (Table 1). This indicated that ~10% of the initial CP amount had remained in the BO after the heating process. This share is in very good agreement with the findings on the BO door (compare *BE* with *BE-NC*).
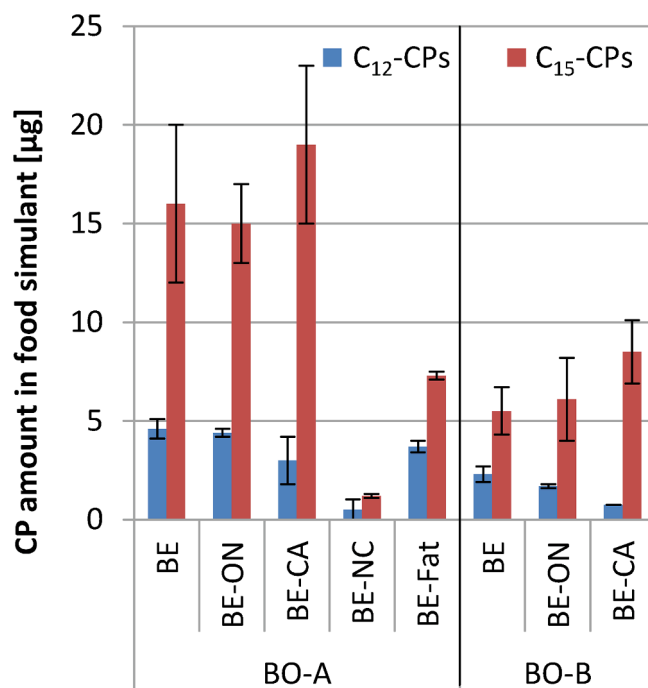
Fig. 3. C$_{12}$-CP and C$_{15}$-CP amounts [µg] transferred from ~200 µg on a baking oven door into a food simulant (coconut fat) in five experiments (BE, BE-ON, BE-CA, BE-NC and BE-Fat, section 2.3) and two different baking ovens (BO-A and BO-B).

Table 2
Transfer rates (TR) of spiked CPs into the food simulant (sum of four times ~1 g of coconut fat) for eight experiments and two different baking ovens. Mean TR was calculated from experiments marked in bold (BE-ON, BE-PCB and BE-C$_{16}$ were not included since the circumstances for CP transport into the baking good were virtually the same as in BE). Based on that rate, CP transfer of a BO door with mean (190 µg SCCPs + 2.7 mg MCCPs) and maximum (270 µg SCCPs + 9.5 mg MCCPs) CP contamination (according to Gallist *et al.* (Gallistl et al., 2018)) as well as CP intake (two meals for two people from the BO per week) were calculated.

	BO-A		BO-B	
	C$_{12}$-CPs	C$_{15}$-CPs	C$_{12}$-CPs	C$_{12}$-CPs
BE	**2.1%**	**7.3%**	**1.1%**	**2.5%**
BE-ON	2.0%	6.9%	0.79%	2.8%
BE-CA	**1.4%**	**8.7%**	**0.35%**	**3.9%**
BE-NC[a]	(0.24%)	(0.55%)	–	–
BE-Fat	**1.7%**	**3.3%**	**–**	**–**
BE-PCB	1.8%	7.3%	–	–
BE-C$_{16}$	1.9%	8.3%	–	–
BE-1 mg	**6.7%**	**8.8%**	**–**	**–**
mean	**2.2%**	**5.8%**		
CP transfer [µg] mean (max)	4.2 (5.9)	160 (550)		
daily CP intake [µg] mean (max)	0.60 (0.85)	22 (79)		

[a] TR based on the CP amount spiked in the previous BE experiment; the "real" CP amount on the door was unknown.

This experiment also indicated that the amount transferred into the food simulant was dependent on the amount present on the inside of the BO door (see below).

In addition, neither cooling overnight (*BE-ON*) nor air circulation (*BE-CA*) affected the transfer of CPs into the food simulant. Only when CPs were applied in a fat matrix on the inside of the BO door (withholding effect, *BE-Fat*), lower amounts of CPs were released and a lesser share of CPs, namely ~50% of the C$_{15}$-CPs and ~80% of the C$_{12}$-CPs of the basic experiment were observed in the food simulant (Table 1). With

regard to the CP pattern, the distribution for C_{12}-CPs changed only marginally (Fig. 2d), while the main C_{15}-CP homolog group in the baking good was shifted from Cl_7- in *BE* to Cl_6-C_{15}-CPs in *BE-Fat* (Fig. 2h).

CP transfer from the food simulant (coconut fat) into the baking oven (BO)

In experiment *BE-C_{16}*, the food simulant was additionally spiked with C_{16}-CPs. After the baking process a small share of C_{16}-CPs ($\sim$8%) was detected on the BO door while >90% of the spiked amount remained in the food simulant (Table 3). This result was in accordance with a recent baking study with one oven, in which the CP content (especially shorter chained CPs) decreased by $\sim$ 24% from dough to baked goods (Perkons et al., 2019). The reported loss of higher chlorinated homologs in the food simulant, which probably originated from dehydrochlorination processes (Perkons et al., 2019) was also observed in the present study. In addition, a preferential release of lower chlorinated CPs was verified by a notable shift from the predominant Cl_7-C_{16}-CPs in the standard to Cl_6-homologs on the BO door, and even Cl_5-homologs in the air sampler (Fig. S5).

Summed-up recovery of spiked CPs after operation of the BO

The total CP amount detected after the heating on cellulose filter, food simulant (coconut fat) and the BO door was only $\sim$10% or less of the CP amount initially spiked onto the inside of the BO door. Moreover, a thorough wipe test of the complete inner area of the oven resulted only in traces of C_{12}- and C_{15}-CPs levels (see above). This produced evidence that the vast majority of the CPs had left the BO via either the (non-hermetic) BO door or the internal ventilation system. Deposition of released CPs in the ventilation duct of the BO was considered unlikely, as no lipophilic compartments, which could serve as sinks could be observed within. Only when applied in a fatty matrix (*BE-Fat*) the summed-up recovery was higher (>3% of the C_{12}-CPs and $\sim$50% of the heavier C_{15}-CPs) because of the reduced release from the BO door (Table 1).

Comparison of the transfer of CPs and PCBs from the BO door

Recovery rates of four PCBs (PCB 158, PCB 186, PCB 194 and PCB 209, each 100 ng), spiked onto the inside of the BO door (*BE-PCB*) correlated with the degree of chlorination (Table 3). Namely, recovered amounts of Cl_6- and Cl_7-congeners PCB 158/PCB 186 from the BO door were similarly low (0.39%/0.12%) as those of C_{12}-CPs (Table 1). This single-chain C_{12}-CP standard was rich in Cl_7-congeners and therefore of similar molecular weight. Recovery rates of the Cl_8- and Cl_{10}-biphenyl (PCB 196/PCB 209) were one order of magnitude higher (2.4% and 8.4%) than those of the more volatile PCBs, i.e. between those of C_{12}- and C_{15}-CPs (section 3.2.1). In addition, the share of PCBs detected in the food simulant (2.5–10%, Table 3) was comparable to the transfer rates of the CPs. All four PCB congeners were also detected in the

Table 3
Distribution of PCBs 158, 186, 196 and 209 as well as C_{16}-CPs (50.9% Cl) in the BO after spiking on the BO door (PCBs, 100 ng each) and the food simulant (C_{16}-CPs, 81.5 µg), respectively.

sample				
	food simulant	BO door	air sampler	sum
PCB 158	8.9 ng	0.39 ng	0.27 ng	9.6 ng
PCB 186	2.5 ng	0.12 ng	0.10 ng	2.7 ng
PCB 196	10 ng	2.4 ng	0.72 ng	13 ng
PCB 209	6.4 ng	8.4 ng	0.90 ng	16 ng
C_{16}-CPs (50.9% Cl)	74 µg (91%)	6.3 µg ($\sim$8%)	detected	80 µg (99%)

cellulose filter paper samples at amounts <1 ng. Therefore, the distribution processes of PCBs and CPs were comparable in the BO and volatility was more important than the compound class.

Use of a second BO model

Three experiments were repeated with a second BO from a different producer (BO–B). In all experiments, the amounts of CPs on the BO door were more than one order of magnitude lower (Table 1), and a lower overall share was recovered when summing up the amounts in the individual samples. In contrast to BO-A, the highest share of CPs in BO-B was detected in the food simulant (coconut fat), with amounts reaching about 30–50% of the CP levels detected in the main baking oven BO-A (Table 1). This indicated that the BO design and the built-in technology had a strong impact on the release of CPs. This could be due to different ways air currents are guided inside and out of the oven, and the tightness of the BO door. However, the deposited amount in the food simulant remained in the same order of magnitude between both ovens, indicating that the transfer into the fat could be similar for different BO models.

Spiking of five-fold higher CP amounts

Initial CP amounts spiked on the inside of the BO door were selected comparably low, last but not least due to concerns that noticeable shares of CPs once spiked into the BO may cause memory effects in subsequent runs, which turned out not being the case. Since several experiments pointed to a concentration-dependent release and transfer of CPs we chose a five-fold higher spiking level in this experiment, i.e. $\sim$1 mg per chain length (C_{12}- and C_{15}-CPs). This amount was still below the highest CP amounts detected on the insides of BO doors in German households (Gallistl et al., 2018). Using *BE* conditions, higher shares of CPs remained on the BO door ($\sim$8% C_{12}- and 58% C_{15}-CPs vs. <1% and $\sim$20%, respectively, at the 200 µg level), and CP amounts were higher on the cellulose air sampler (Table 1). Most importantly, however, the CP amounts in the food simulant were also higher and now in the range of 72 µg C_{12}-CPs and 96 µg C_{15}-CPs (Table 1). This corresponded with $\sim$3-fold and $\sim$1.5 fold higher transfer rates of C_{12}-CPs and C_{15}-CPs, respectively, into the food simulant compared to the basic experiment (*BE*) (Table 2). In either case (C_{12}- and C_{15}-CPs), the CP patterns in food simulant and passive sampler were similarly shifted to lower chlorinated homologues than in the *BE*. However, the homologue patterns on the BO door were similar to the ones of the CP-standards that was spiked (Fig. S2, S3, Supplementary Material). This was in agreement with the lower CP share that was released.

Discussion

Overall, the following general observations could be made:

a) high shares of CPs were released from the BO door, especially when no fat was present/applied
b) high amounts of CPs could not be detected in any oven parts, mainly due to release from the oven
c) the release increased with the volatility of the CPs: C_{12}-CPs > C_{15}-CPs and, e.g. Cl_5-CPs > Cl_7-CPs
d) similar behaviour of CPs and PCBs of similar volatility was observed
e) the CP amount in the baking good increased with the amount spiked

Irrespective of the questions that could not be solved (fate of the undetected share of CPs) and some differences between the individual experiments, all experiments verified the transfer of CPs from the BO into the food simulant (Fig. 2). When $\sim$ 200 µg CPs were freshly spiked on the BO door (*BE, BE-ON, BE-CA, BE-PCB, BE-C16* and *BE-Fat*), $\sim$0.5–4.6 µg SCCPs (mean 2.6 µg or 1.2%) and $\sim$ 1.2–19 µg MCCPs (mean 9.8 µg or 4.5%) were detected in the baking good. After a second

heating without a new CP spike the CP amounts were ~ 10 fold lower (*BE-NC*) while those with a higher CP spike (*BE-1 mg*) resulted in slightly higher transfer rates of 6.7% (72 µg) of C_{12}- and 8.8% (96 µg) of C_{15}-CPs. Approximately, the transfer rates of the CPs from the BO door linearly increased with the amount supplied. In addition, the concentration ranges applied in this study were comparable with those detected on the inside of BO doors (ages 1–20 y) sampled by wiping tests in different households (Gallistl et al., 2018). Furthermore, Gallistl *et al.* produced evidence that the CPs detected on the inside of BO doors originated from the BOs themselves. Hence, the spiked CP concentrations used in this study and the observed transfer rates into the baking goods seemed to be well suited to estimate the CP intake via release from the BO.

Preliminary estimate of the CP intake from food prepared in the BO

As shown in the spiking experiments, CPs on the inside of a BO door can be released into the baking good. When a fatty matrix was present, as is often the case in household ovens (Gallistl et al., 2018), a one-time CP contamination could be preserved and lead to multiple releases. This would result in a continued CP exposition of the consumer. The daily CP intake $DI(CP_x)$ through baking good contamination was estimated by multiplying the mean transfer rate TR_x (2.2% for C_{12}-CPs and 5.8% for C_{15}-CPs; mean values from TRs of experiments *BE, BE-CA, BE-Fat* and *BE-1 mg* from both BO-A and BO-B, Table 2) with the amount of CPs on the BO door (m_{CP} in [µg]) according to equations (1) and (2):

$$m_{CP}^T = m_{CP} * TR_x \tag{1}$$

$$DI(CP_x) = m_{CP}^T * \frac{n_t}{p} \tag{2}$$

with n_t being the weekly usage frequency of the BO (x/7 days), and *p* is the amount of people living in the household. The BO was expected to be used twice per week ($n_t = 2$; 100 uses per year, Verbraucherzentrale Rheinland-Pfalz, 2014). For Germany, the representative "mean" household consists of two people ($p = 2$, Statistische Ämter des Bundes und der Länder, 2018), i.e. each person eats 50% of the meal and takes up 50% of the CP load. In addition, results obtained with C_{12}-CPs were considered representative for SCCPs and those of C_{15}-CPs for MCCPs. Further, it was assumed that the food simulant was representative for baking goods. A further assumption was that a normal CP-containing BO contained about 1 g CPs or more in its inner parts and that by each use, (similarly high) CP amounts in the µg–mg range could be released and found on the BO door.

According to equations (1) and (2) and using the mean CP amounts found on BO doors (i.e. 190 µg SCCPs and 2.9 mg MCCPs, Table S5), uptake of SCCPs via transfer into the baking good was about 0.6 µg/day (Table 2). This amount was ~ 50% lower than the general amount taken up via food (1.4 µg) according to Yuan et al. (2017). Noteworthy, the mean estimated uptake of MCCPs via transfer into the baking good of 22 µg was ~7-fold higher than the reported uptake via food (i.e. 3.0 µg MCCPs, Yuan et al., 2017). Consequently, baking processes in CP-containing BOs could considerably contribute to the daily intake of CPs. Moreover, even higher amounts than the ones applied in this study have been previously detected in in household BOs (i.e. 270 µg SCCPs and 9.5 mg MCCPs, Gallistl et al., 2018). For these maximum amounts and assuming the same TR, the daily intake via transfer into the baking good would be as high as 0.85 µg SCCPs and 79 µg MCCPs, respectively (Table 2). Therefore, the BO should be considered as a relevant source of CP exposure in future uptake studies. Since BO fall within the definition of food contact materials, it is recommended to refrain from using CPs in BOs as long as there is no final health-related risk assessment. The study of Gallistl et al. (2018), indicated that ~50% of BOs were free of CPs. Hence, it appears technically possible to produce BO without CPs.

CRediT authorship contribution statement

Jannik Sprengel: Investigation, Formal analysis, Visualization, Writing - original draft. **Stefanie Rixen:** Investigation, Writing - review & editing. **Oliver Kappenstein:** Conceptualization, Resources, Funding acquisition, Writing - review & editing. **Walter Vetter:** Conceptualization, Writing - original draft, Writing - review & editing, Supervision.

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.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.fochx.2021.100122.

References

Bendig, P., Hägele, F., & Vetter, W. (2013). Widespread occurrence of polyhalogenated compounds in fat from kitchen hoods. *Analytical and Bioanalytical Chemistry, 405*(23), 7485–7496.

Conference of the Parties of the Stockholm Convention (COP.8) (2017). Decision SC-8/11: Listing of short-chain chlorinated paraffins. Geneva, 65. http://chm.pops.int/Portals/0/download. aspx?d=UNEP-POPS-COP.8-SC-8-11.English.pdf. Accessed 12.08.2020.

Dąbrowski, Ł. (2016). Review of use of keepers in solvent evaporation procedure during the environmental sample analysis of some organic pollutants. *Trends in Analytical Chemistry, 80*, 507–516.

EFSA Panel on Contaminants in the Food Chain (CONTAM), Schrenk, D., Bignami, M., Bodin, L., Chipman, J. K., del Mazo, J., Grasl-Kraupp, B., Hogstrand, C., Hoogenboom, L., Leblanc, J.-C., Nebbia, C. S., Ntzani, E., Petersen, A., Sand, S., Schwerdtle, T., Vleminckx, C., Wallace, H., Brüschweiler, B., Leonards, P., Rose, M., Binaglia, M., Horváth, Z., Ramos Bordajandi, L., & Nielsen, E. (2020). Risk assessment of chlorinated paraffins in feed and food. EFSA J., 18.

European Parliament, Council of the European Union (2004). Regulation (EC) No 1935/2004 of the European Parliament and of the Council of 27 October 2004 on materials and articles intended to come into contact with food and repealing Directives 80/590/EEC and 89/109/EEC: Regulation (EC) No 1935/2004.

Fridén, U. E., McLachlan, M. S., & Berger, U. (2011). Chlorinated paraffins in indoor air and dust: Concentrations, congener patterns, and human exposure. *Environment International, 37*(7), 1169–1174.

Gallistl, C., Lok, B., Schlienz, A., & Vetter, W. (2017). Polyhalogenated compounds (chlorinated paraffins, novel and classic flame retardants, POPs) in dishcloths after their regular use in households. *Science of the Total Environment, 595*, 303–314.

Gallistl, C., Sprengel, J., & Vetter, W. (2018). High levels of medium-chain chlorinated paraffins and polybrominated diphenyl ethers on the inside of several household baking oven doors. *Science of the Total Environment, 615*, 1019–1027.

HEA – Fachgemeinschaft für effiziente Energieanwendung e.V. (2020). Elektroherde: Aufbau und Technik. https://www.hea.de/fachwissen/herde-backoefen/elektroherde-aufbau-und-funktion. Accessed 28.07.2020.

Iino, F., Takasuga, T., Senthilkumar, K., Nakamura, N., & Nakanishi, J. (2005). Risk assessment of short-chain chlorinated paraffins in Japan based on the first market basket study and species sensitivity distributions. *Environmental Science & Technology, 39*(3), 859–866.

Krätschmer, K., Cojocariu, C., Schächtele, A., Malisch, R., & Vetter, W. (2018). Chlorinated paraffin analysis by gas chromatography Orbitrap high-resolution mass spectrometry: Method performance, investigation of possible interferences and analysis of fish samples. *Journal of Chromatography A, 1539*, 53–61.

Krätschmer, K., Schächtele, A., Malisch, R., & Vetter, W. (2019). Chlorinated paraffins (CPs) in salmon sold in southern Germany: Concentrations, homologue patterns and relation to other persistent organic pollutants. *Chemosphere, 227*, 630–637.

Mandalakis, M., Atsarou, V., & Stephanou, E. G. (2008). Airborne PBDEs in specialized occupational settings, houses and outdoor urban areas in Greece. *Environmental Pollution, 155*(2), 375–382.

Oehme, M., Vetter, W., & Schlabach, M. (1994). Quantitative Analysis of Persistent Organochlorines in Samples From Polar Regions a Challenge For The Analytical

Chemist. *International Journal of Environmental Analytical Chemistry, 55*(1-4), 261–266.

Perkons, I., Pasecnaja, E., & Zacs, D. (2019). The impact of baking on chlorinated paraffins: Characterization of C10–C17 chlorinated paraffins in oven-baked pastry products and unprocessed pastry dough by HPLC–ESI–Q–TOF–MS. *Food Chemistry, 298*, 125100. https://doi.org/10.1016/j.foodchem.2019.125100.

Reth, M., Zencak, Z., & Oehme, M. (2005). New quantification procedure for the analysis of chlorinated paraffins using electron capture negative ionization mass spectrometry. *Journal of Chromatography A, 1081*(2), 225–231.

Rudan, M. A., & Barbano, D. M. (1998). A model of Mozzarella cheese melting and browning during pizza baking. *Journal of Dairy Science, 81*(8), 2312–2319.

Schinkel, L., Lehner, S., Heeb, N. V., Lienemann, P., McNeill, K., & Bogdal, C. (2017). Deconvolution of mass spectral interferences of chlorinated alkanes and their thermal degradation products: Chlorinated alkenes. *Analytical Chemistry, 89*(11), 5923–5931.

Schinkel, L., Lehner, S., Knobloch, M., Lienemann, P., Bogdal, C., McNeill, K., & Heeb, N. V. (2018). Transformation of chlorinated paraffins to olefins during metal work and thermal exposure - Deconvolution of mass spectra and kinetics. *Chemosphere, 194*, 803–811.

Sprengel, J., & Vetter, W. (2019). Synthesis and characterization of eight single chain length chlorinated paraffin standards and their use for quantification. *Rapid Communications in Mass Spectrometry, 33*(1), 49–56.

Sprengel, J., Wiedmaier-Czerny, N., & Vetter, W. (2019). Characterization of single chain length chlorinated paraffin mixtures with nuclear magnetic resonance spectroscopy (NMR). *Chemosphere, 228*, 762–768.

Statistische Ämter des Bundes und der Länder (2018). Gebiet und Bevölkerung nach Ländern – Haushalte. http://www.statistikportal.de/de/bevoelkerung/haushalte. Accessed 07.04.2020.

Tomy, G. T. (2010). Analysis of chlorinated paraffins in environmental matrices: The ultimate challenge for the analytical chemist. In J. de Boer, & T.-S. Ali (Eds.), *The Handbook of Environmental Chemistry: vol. 10. Chlorinated Paraffins* (pp. 83–106). Berlin: Springer.

van Mourik, L. M., Gaus, C., Leonards, P. E. G., & de Boer, J. (2016). Chlorinated paraffins in the environment: a review on their production, fate, levels and trends between 2010 and 2015. *Chemosphere, 155*, 415–428.

Verbraucherzentrale Rheinland-Pfalz (2014). Energieverbrauch von Backofen und Herd. https://www.verbraucherzentrale-rlp.de/sites/default/files/migration_files/media232292A.pdf. Accessed 28.07.2020.

Vetter, W., & Luckas, B. (1995). Synthesis, isolation, and chromatography of perdeuterated α-1,2,3,4,5,6-hexachlorocyclohexane. *Journal of High Resolution Chromatography, 18*(10), 643–646.

Vetter, W., & Luckas, B. (1998). On the significance of [M−HCl]⁻ fragment ions in gas chromatography/electron capture negative ionization mass spectra of octachloro compounds of technical toxaphene. *Rapid Communications in Mass Spectrometry, 12*(6), 312–316.

Weichbrodt, M., Vetter, W., & Luckas, B. (2000). Microwave-assisted extraction and accelerated solvent extraction with ethyl acetate-cyclohexane before determination of organochlorines in fish tissue by gas chromatography with electron-capture detection. Journal of AOAC International, 83, 1334–1343.

Wilson, W?D, MacKinnon, I?M, & Jarvis, M?C (2002). Transfer of heat and moisture during oven baking of potatoes. *Journal of the Science of Food and Agriculture, 82*(9), 1074–1079.

Xue, J., Lefort, G., & Walker, C. E. (2004). Effects of oven humidity on foods baked in gas convection ovens. *Journal of Food Processing and Preservation, 28*(3), 179–200.

Xue, J., & Walker, C. E. (2003). Humidity change and its effects on baking in an electrically heated air jet impingement oven. *Food Research International, 36*(6), 561–569.

Yuan, B.o., Strid, A., Darnerud, P. O., de Wit, C. A., Nyström, J., & Bergman, Å. (2017). Chlorinated paraffins leaking from hand blenders can lead to significant human exposures. *Environment International, 109*, 73–80.

Zellmer, S., Heiserich, L., Kappenstein, O., Merkel, S., Schulte, A., & Luch, A. (2020). MCCP: Are medium-chain chlorinated paraffins of concern for humans? *Archives of Toxicology, 94*(3), 955–957.