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Fate of Polybrominated Diphenyl Ethers during Cooking of Fish in a New Model Cooking Apparatus and a Household Microwave

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Supporting Information

ABSTRACT: Fish is a major source of human exposure to polybrominated diphenyl ethers (PBDEs). Because fish is mainly consumed after cooking, this measure may alter the pattern and amounts of PBDEs that are finally consumed. To investigate this issue, we developed a model cooking apparatus consisting of a small glass bowl and a beaker glass with an exhaust fitted with a polyurethane foam filter connected to a water jet pump. In this model cooking apparatus, fish (1 g) and/or sunflower oil (0.2/0.4 g) spiked with three PBDE congeners was cooked for 30 min. Small amounts of the semi-volatile PBDEs were evaporated from the fish (BDE-47 < BDE-15), while the non-volatile BDE-209 was partly transformed. Additional experiments in a household microwave provided similar results, except that no transformation was observed for BDE-209. The model cooking apparatus proved to be well-suited to study the fate of polyhalogenated compounds in fish during cooking.

KEYWORDS: *cooking, polybrominated diphenyl ethers, hydrodebromination, volatilization*

INTRODUCTION

Polybrominated diphenyl ethers (PBDEs) are persistent, bioaccumulative, and toxic chemicals, which have been partly classified as persistent organic pollutants (POPs) under the Stockholm Convention on POPs.[1] The physicochemical properties of PBDEs have led to their detection all around the world. One main source for the human exposure with PBDEs is uptake via food,[2,3] with fish/seafood being identified as one primary source of exposure.[4−6] Typically, human dietary exposure to POPs, such as PBDEs, is estimated from their content in raw food. Fish, however, is mainly consumed after cooking, and this process may alter both the amounts and composition of PBDEs that is actually consumed.[7−9] However, the few studies focusing on the fate of PBDEs during cooking provided non-uniform and partly contradictory results.[7−12] Alternatively, the behavior of PBDEs during cooking was suggested to be similar to that of polychlorinated biphenyls (PCBs),[6] which is mainly governed by a reduction of the concentrations because of their partial leaching out with fat or direct volatilization,[13] i.e., similar to the processes observed during deodorization of edible oils.[14] However, in another study, it was shown that only PCBs but not the heavier polybrominated compounds were removed during deodorization of dietary fish oil.[15]

The goal of the present study was to study the fate of PBDEs during cooking in fish by means of a miniaturized model cooking apparatus. A similar approach has been used by Dong et al. during the cooking of polychlorinated compounds but at a larger scale.[16−18] The miniaturized model cooking apparatus enabled us to analyze the whole sample. Accordingly, we could obtain a quantitative mass balance of the PBDEs, because no sample aliquotation was necessary.[8] With this design, we wished to determine what portions of two semi-volatile (BDE-15 and BDE-47) PBDE congeners and the non-volatile BDE-209 were transformed or evaporated from the fish sample. Complementary experiments were performed in a household microwave.

MATERIALS AND METHODS

Samples, Standards, and Chemicals. *Chemicals.* Details of chemicals, standards, and salmon (*Salmo salar*) fillet samples (farmed in Chile; tested free of PBDEs[8,19]) used were reported by Bendig et al.[8] Additional polybrominated dibenzofuran (PBDF) standards were from Cambridge Isotope Laboratories, Andover, MA [1,2,3,4,6,7,8-heptabromodibenzofuran (HpBDF) and 1,2,3,4,7,8-hexabromodibenzofuran (HxBDF)] or from Wellington Laboratories, Guelph, Ontario, Canada [1,2,3,4,6,7,8,9-octabromodibenzofuran (OcBDF)]. Polyurethane foam (PUF), certified "flame retardant free", was from Tisch Environmental (Cleves, OH). The PUF was pre-extracted by open-vessel microwave-assisted extraction (MAE) using the method described by Bendig et al.[8]

Model Cooking Apparatus. The model cooking apparatus consisted of a glass bowl (40 mm outer diameter and 15 mm high) equipped with a central cavity (33 mm internal diameter and 10 mm depth), which was placed on a heating plate (MR Hei-Standard, Heidolph Instruments, Schwabach, Germany) (Figure 1). The sample (oil and fish; see next section) was placed into the cavity. The glass bowl was fitted with a 500 mL glass beaker (70 mm diameter and 130 mm height) placed upside down with a distance of ∼5−10 mm above the heating plate (Figure 1). The beaker was equipped with an exhaust (3.5 mm internal diameter) at the bottom (Figure 1), which was connected to a water jet pump by means of a plastic tube (600 mm length × 5 mm internal diameter). This device was used to simulate a kitchen hood. The exhaust was filled with a ∼1000 mm^3 piece of PUF (see previous section), which served as a trap for volatilized PBDEs. The top of the beaker was covered with a plastic bag filled with crushed ice, to promote cold condensation of lipid fumes and PBDEs.

Cooking Experiments and Sample Preparation. The cavity of the glass bowl was first furnished with 200 mg of sunflower oil. Then, a

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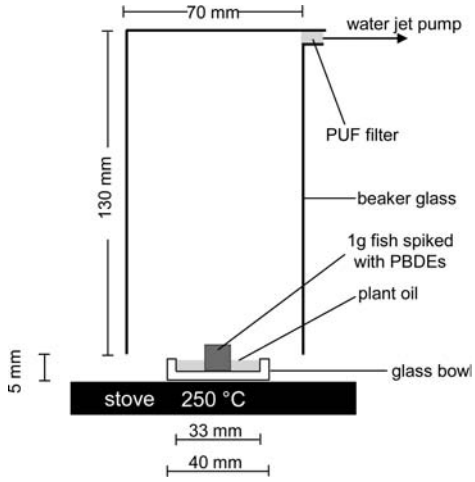

Figure 1. Schematic diagram of the model cooking apparatus.

cubic piece of 1 g of salmon fillet spiked with 200 ng of BDE-15, BDE-47, and BDE-209 was added to the cavity of the glass bowl (Figure 1). Alternatively, 400 mg of sunflower oil (no fish) was spiked with 200 ng of the three PBDE congeners (in 10 μL of toluene for BDE-209 and 10 μL of methanol for BDE-15 and BDE-47). The glass bowl with the sample was set on the preheated heating plate (250 °C) and immediately covered with the glass beaker, which had the water jet pump already switched on. The cooking experiment in the apparatus lasted for 30 min. Temperatures in the fish were measured with an 830 T1 infrared thermometer (Testo, Lenzkirch, Germany) suited for a range from −30 to +400 °C (precision of ±1.5 °C) or with a simple mercury-in-glass thermometer.

After the cooking experiment, (i) the PUF in the exhaust, (ii) the glass beaker, and (iii) the glass bowl containing the sample (salmon fillet together with the remaining lipids) were separated and spiked with 200 ng of the internal standard 2,3′,4,4′-tetrabromo-6′-methoxy diphenyl ether (6′-MeO-BDE-66).[20] Then, the (i) PUF was transferred with tweezers into a 250 mL microwave vessel. The (iii) sample in the glass bowl was washed with 25 mL of cyclohexane/ethyl acetate (azeotropic mixture, 46:54, w/w) into a 250 mL beaker and then rinsed with 5× 2 mL of cyclohexane/ethyl acetate. The two subsamples were microwave-assisted extracted according to Bendig et al.[8] After extraction, the solvent of the extracts was exchanged to isooctane. The (ii) glass beaker was rinsed with 5× 2 mL of cyclohexane/ethyl acetate (46:54, w/w). The rinsing solvents were combined in a pear-shaped flask, and the solvent was exchanged to isooctane as well. These three sub-samples were individually treated with 5 mL of sulfuric acid followed by a silica gel cleanup as described by Bendig et al.[8]

For additional experiments in a household microwave, 1 g of fish was placed in a glass bowl (see above) and heated with a household microwave (HF 26540, type SS-652T, Siemens, Munich, Germany) for varied times (2, 5, and 30 min) with a power of 600 W. The sample preparation was as shown above.

Gas Chromatography. Measurements were performed with a 5890 series II Plus gas chromatograph (Hewlett-Packard, Waldbronn, Germany) equipped with an electron capture detector (GC/ECD) and a 15 m × 0.25 mm × 0.25 μm BGB-1 column (BGB-Analytik, Boeckten, Switzerland) using parameters previously reported in detail.[8] Confirmatory gas chromatography with electron-capture negative-ion mass spectrometry (GC/ECNI-MS) analyses were performed with a 7890/5975C system (Agilent, Waldbronn, Germany). Analyses were run in the selected ion monitoring mode using the parameters by Bendig et al.,[8] except for the 15 m length × 0.25 mm internal diameter column coated with a 0.1 μm film of RTX-1 (Restek, Bellefonte, PA), which was used in the present study.

Quality Control. All glassware was covered with aluminum foil or amber glass to prevent photolysis of PBDEs.[21,22] Procedural blanks were tested free of PBDEs (LOD ∼ 0.1−0.5 ng/sample). Experiments with fish in the model cooking apparatus were performed in 8-fold replication. Spiked fish method blank samples were prepared in 4-fold replication and treated just as the samples (exposed to light for the same time and place as the samples) but not heated. Additionally, unheated, spiked sample blanks (n = 4) were performed to rule out PBDE transformation that was not related to the cooking process. Furthermore, method blank samples were prepared in the same way in duplicate but kept under light exclusion all of the time, to control the impact of light on the hydrodebromination of BDE-209. Recovery rates of the internal standard 6′-MeO-BDE-66 were 91 ± 11% (ranging from 70 to 108%). The purity of the BDE-209 was ∼97%, and the remaining share came from Br_9DEs. A further portion of BDE-209 was transformed into Br_9DEs during the processing of the samples. Possible hydrodebromination in fish during cooking versus the occurrence of the transformation products in sample blanks was evaluated by means of one-way analysis of variation (ANOVA) as implemented in Excel. Resulting p values of $p < 0.05$ were considered significant. Also, duplicate method blank samples of neat sunflower oil spiked with PBDEs were prepared (treated just as the samples but not heated). Experiments with fish in the household microwave were performed in triplicate (2 and 5 min) and quadruplicate (30 min) replications.

RESULTS

Specifications of the Model Cooking Apparatus. The model cooking apparatus (Figure 1) was designed to provide a miniature simulation of the real kitchen scenario. In household kitchens, the volume between the stove and kitchen hood (i.e., the cooking area) is about 1 m^3. Producers of kitchen hoods report capacities of 300−1000 m^3/h air flow through kitchen hoods. Hence, the ratio of the volume of air passed through the kitchen hood to the volume of the cooking area is ∼300−1000 per hour. In our model cooking apparatus, the cooking area (defined by the volume of the glass beaker) was 0.5 dm^3 and the throughput of the water jet pump through the exhaust was determined to be ∼360 dm^3/h in our laboratory. In our model apparatus, ∼720 times the volume of the cooking area was transported hourly over the cooking food. This setup was in good agreement with the real scenario in private households. This model apparatus was used to study the distribution of PBDE congeners (BDE-15, BDE-47, and BDE-209) in the cooking bowl with sample, the glass beaker, and the PUF in the exhaust.

Heating of Sunflower Oil Spiked with BDE-209, BDE-47, and BDE-15 in the Model Cooking Apparatus. After 30 min of heating sunflower oil spiked with the PBDEs (no fish), 95% of BDE-209 was recovered from the cooking bowl. The remaining share of BDE-209 was detected in the beaker glass (3%) and in the PUF in the exhaust (2%). In comparison to this non-volatile congener, only 32% of the initial amount of the semi-volatile BDE-47 was detected in the sample, while $^2/_3$ was found outside the sample, i.e., in the glass beaker (42%) and the PUF in the exhaust (27%) (Figure 2). The more volatile BDE-15 was almost completely removed from the sample. The largest fraction of BDE-15 (∼55%) was detected in the PUF in the exhaust, and ∼43% was identified in the glass beaker (Figure 2). All three PBDE congeners provided a quantitative mass balance, and accordingly, metabolites were not detected at all. This chemical integrity of PBDE congeners during heating in sunflower oil was in agreement with a previous study in a (long and narrow) test tube.[8] However, only with the new and more realistic design, the partial volatilization of PBDEs from the sample could be observed. After 30 min of cooking, the remaining share of the PBDEs in the fish sample was linearly linked with the molecular weight

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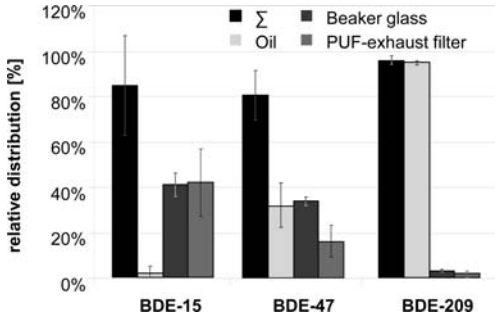

Figure 2. Relative distribution (%) of BDE-15, BDE-47, and BDE-209 in the model cooking apparatus after 30 min of heating in sunflower oil.

(Figure 3). However, removal of even traces of BDE-209 from sunflower oil into the gas phase was completely unexpected because of the non-volatile nature of this high-molecular-weight compound (959 g/mol). Most likely, this effect was due to adsorption to vapors of hot sunflower oil released from the glass bowl during cooking. In agreement with that, the (initially colorless) PUF in the exhaust turned yellow during heating (see Figure S1 of the Supporting Information). This attested that oil was evaporated from the glass bowl. Apparently, the oil could not be effectively condensed on the cooled beaker glass. Likewise, the more volatile BDE-15 was condensed to a lesser degree on the cooled beaker glass than the heavier tetrabrominated BDE-47 (Figure 2).

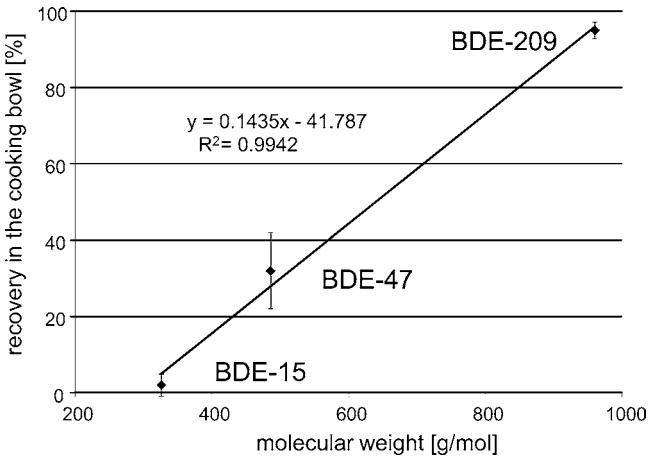

Figure 3. Correlation of the recovery of BDE-15, BDE-47, and BDE-209 in the cooking glass bowl with the molecular weight after 30 min of heating in sunflower oil in the model cooking apparatus.

Cooking of Fish in the Model Cooking Apparatus: Temperatures and Conditions of the Fish. Within 1−2 min, the temperature at the bottom of the fish reached 70 °C and the fish started cooking. Between 4 and 5 min of heating, water condensed inside the beaker glass. After ∼10 min, the lower part of the fish that was dipped into the sunflower oil turned brown, while the top still looked raw or only slightly cooked. At this stage, the temperature on the bottom and the top of the fish was 160 and 110 °C, respectively (Figure 4). When 15 min had passed, the bottom of the fish had reached the maximum temperature of 170 °C. At this point, no further water condensed on the glass. The brown color of the fish intensified with the heating time (pictures are shown in Figure S2 of the Supporting Information). The temperature on the top

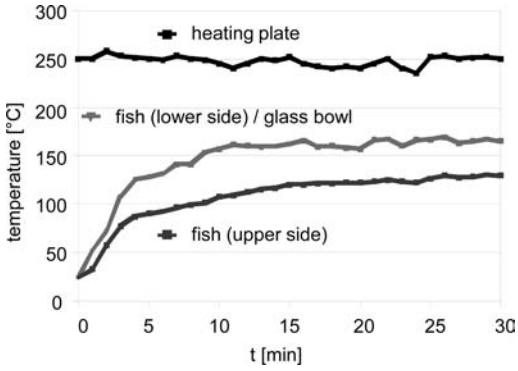

Figure 4. Temperature profile of the heating plate, with the upper and lower sides of the fish during the cooking experiment.

of the fish (125 °C) still lagged behind its lower part. At the same time, the PUF in the exhaust turned yellow (similar to the effect observed with sunflower oil; see Figure S1 of the Supporting Information) and spots of lipids became visible inside the glass beaker.

Cooking of PBDE-Spiked Fish in the Model Cooking Apparatus. When heated in fish for 30 min, between 22 and 79% (50 ± 19%) of BDE-15 was recovered in the fish, which was on average ∼25 times more than in neat sunflower oil. In comparison to the experiment with sunflower oil, heating of fish was more difficult to reproduce because, in some replicate samples, the oily matrix was splashed from the sample. As a consequence, the variations between replicate samples were higher than during heating of BDE-15 in sunflower oil (Figures 2 and 5). Two samples that showed the most excessive

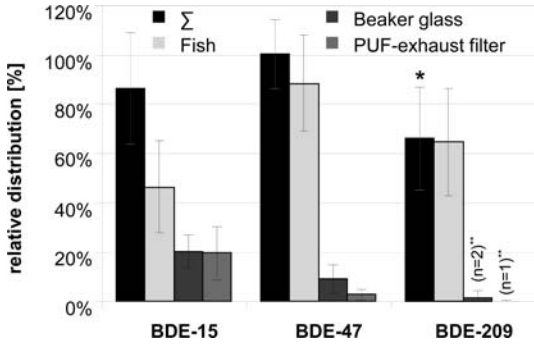

Figure 5. Relative distribution of BDE-15, BDE-47, and BDE-209 in the model cooking apparatus when heated in sunflower oil and fish. (∗) Recovery of BDE-209 was reduced because of its transformation, and the missing ∼35% was detected in the form of Br_8DEs and Br_9DEs. (∗∗) *n* refers to the number of samples in which BDE-209 was actually detected (because of splashing of the sample).

splashing of the oily matrix from the sample were those with the lowest overall recovery of BDE-15 from all compartments (see Table S1 of the Supporting Information). The released share of BDE-15 was almost equally distributed between the PUF in the exhaust (24%) and the beaker glass (20%), i.e., in a similar ratio as observed during the cooking of the PBDEs spiked in sunflower oil alone (compare Figures 2 and 5). The lower rate of BDE-15 released from the fish compared to sunflower oil was possibly due to the water content of the fish tissue, which is accompanied with a lower temperature in the fish. In addition, the temperature gradient observed within the

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fish (Figure 4) could be a further explanation for the observed variations between the experiment with fish and oil.

A much higher amount of BDE-47 (89 ± 19% of the spike) was retained in the fish compared to pure sunflower oil (Figures 2 and 5). Similar to the experiment with oil, a higher share of BDE-47 released from the fish was found in the beaker glass and much less in the PUF in the exhaust (Figure 5). Altogether, the mass balance indicated no loss/transformation of BDE-47 from the sample. Higher but still low amounts of BDE-47 were only eliminated when high amounts of the oily matrix were splashing from the fish.

A different situation was observed with BDE-209. In six of eight replicates, BDE-209 was exclusively detected in the fish. In the remaining two replicates, up to 8% were also detected in the glass beaker and, on one occasion, in the PUF in the exhaust (Figure 5). These two replicates were those characterized by remarkable amounts of fat being splashed from the fish into the beaker glass (see Table S1 of the Supporting Information). This confirmed the observation from the experiments with sunflower oil that splashing of the oily matrix appeared to be the general prerequisite for the release of BDE-209 from the fish into the "vapor phase". Accordingly, the oily matrix released from the fish served as an adsorbent for the transport of the non-volatile BDE-209 into the "vapor phase", similar to the particle-bound atmospheric transport of BDE-209.[23,24] The splashing effect in these two replicate samples was also observed for BDE-15 and BDE-47, but it pertained to even more replicate samples (Figure 6). This feature underlines the

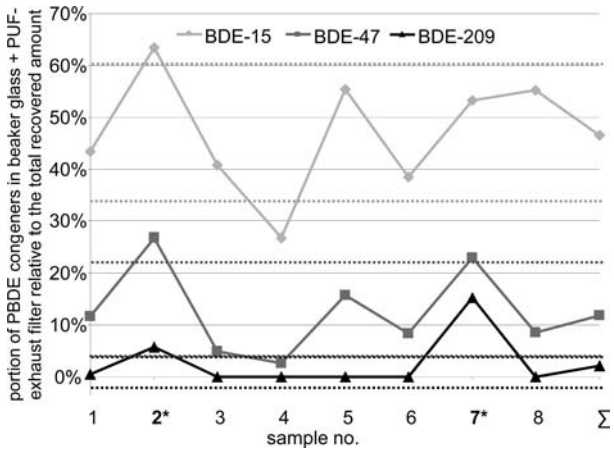

Figure 6. Recovery of BDE-15, BDE-47, and BDE-209 in the vapor phase in samples 1−8 after 30 min of heating of fish in the model cooking apparatus. Samples with significant splashing of the oily sample matrix are marked with an asterisk (∗). The dotted lines refer to the standard deviations.

particular role of adsorption to liquids for the release of BDE-209 into the vapor phase. However, the amounts of BDE-209 (66 ± 21%) detected in the fish were much lower than in sunflower oil (96 ± 1.5%). The lower recovery rate of BDE-209 in experiments with fish was due to its partial transformation.[8] The comparison between the amounts of hydrodebromination products of BDE-209 detected in the method blanks (see the Materials and Methods) and those partially formed during sample processing resulted in significant differences for BDE-206 (p value of 0.002) but not for BDE-207 and BDE-208 (p values of 0.76 and 0.21; see Figure S3 of the Supporting Information). Accordingly, the cooking-induced hydrodebromination of BDE-209 into BDE-206 was a key process in the transformation of BDE-209 in fish. Subsequent transformation into Br_8DEs showed significant differences, especially for BDE-199 and BDE-196 (p values of 0.02 and 0.006, respectively; see Figure S3 of the Supporting Information). In agreement with previous experiments in the test tube, BDE-199 and BDE-196 were found to be the key Br_8DEs of the BDE-209 hydrodebromination in fish.[8] In addition, traces of highly brominated PBDFs were detected in the samples.[8,25] Summing up the amounts of BDE-209 and the transformation products provided a quantitative mass balance (104 ± 19%). In the model cooking apparatus, the cooking of the BDE-spiked fish for 30 min was accompanied with a linear transformation rate of 1.16%/min BDE-209. This value agreed well with the transformation rate of 1.14%/min BDE-209 observed in previous experiments in test tubes,[8] although the maximum temperature was lower (i.e., 170 °C) than in the test tube (i.e., 200 °C, reached within ∼10 min[8]). The spectrum of PBDE transformation products (Br_9DEs > Br_8DEs > Br_7DEs) in the model cooking apparatus and the test tube was also comparable.

Cooking Experiments in a Household Microwave. Additional cooking experiments with PBDE-spiked fish were performed in a household microwave for 2, 5, and 30 min. With a longer heating time, the rate of BDE-15 released from the fish increased (Figure 7). After 30 min, ∼30% of BDE-15 was

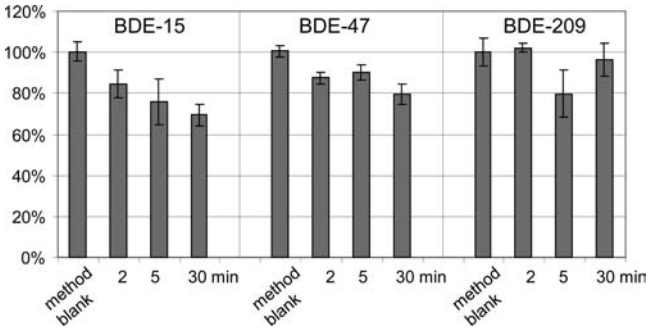

Figure 7. Relative distribution of BDE-15, BDE-47, and BDE-209 in method blanks and fish after heating in a household microwave for 2, 5, and 30 min (600 W).

removed from the fish, which was slightly less than in the cooking experiment in the model cooking apparatus (Figure 5). The evaporation rate of BDE-47 was smaller and showed alternating mean recovery rates in the fish for 2, 5, and 30 min (Figure 7). After 30 min in the microwave, about 20% of BDE-47 was released from the fish (Figure 7). For BDE-209, we noted non-characteristic reduction of the amount in the fish after 5 min, which is in contrast to the heating for 30 min in the microwave (Figure 7). However, transformation products of BDE-209 could not be detected in any heating experiment in the microwave, which is different to the model cooking apparatus. In the microwave, the temperature did not exceed 100 °C, i.e., the boiling point of water, which seemed to be the key factor for the transformation of BDE-209.

DISCUSSION

Benefits of the Small-Scale Experiments of Cooking of PBDE-Spiked Fish in the Model Cooking Apparatus and the Microwave. Switching to the small-scale design primarily helped to solve the problems associated with aliquotation of samples because the whole sample was analyzed after cooking.

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By this measure, the loss of water and/or fat from the fish did not play a role and absolute amounts determined after cooking could be compared to amounts initially spiked into the uncooked sample. This new experimental design allowed for verification of two key findings, i.e., (i) the partial evaporation of the more volatile PBDE congeners from the sample during the heating in both the model cooking apparatus and the microwave and (ii) the partial transformation of BDE-209 (but not of BDE-47 and BDE-15) during the cooking of fish, which was in contrast to its heating in oil in the model cooking apparatus and in fish in the microwave.

Evaporation of PBDE Congeners from Both the Model Cooking Apparatus and the Microwave. Release of volatile PBDEs from the samples was observed in all experiments. In the cooking apparatus, the effect was most pronounced for BDE-15 (~45% evaporated from the fish), while only ~10% BDE-47 was released from the fish after 30 min of cooking. Slightly lower proportions were released during microwave heating, which indicated that the temperature in the fish was lower in the microwave setup and lower than during the heating in sunflower oil (as indicated by the higher evaporation rate of both PBDE congeners). Typically, cooking times for fish are much shorter (~10 min). Assuming that the evaporation rate linearly increased with time, only ~3% of BDE-47 would have been released from fish after 10 min. Accordingly, evaporation of BDE-47 during cooking will only slightly reduce the amount of BDE-47 consumed with the fish (in comparison to the initial amount). Accordingly, the human exposure of BDE-47 (and related PBDE congeners, such as BDE-99 and BDE-100) from fish can be estimated from its concentration in the fish prior cooking. In a similar way, the release of BDE-15 from fish (~45% after 30 min) would be ~15% of the initial amount of BDE-15 after 10 min. The direct link between molecular weight and evaporation rate indicated that evaporation may only play a role for compounds as light as or lighter than BDE-15 (molecular weight of 328 g/mol). This applies to polychlorinated compounds with similar volatility, such as PCBs, hexachlorocyclohexanes (HCHs), and dichlorodiphenyltrichloroethane (DDT)-related compounds.[13,26] Partial evaporation of the more volatile polyhalogenated compounds may thus slightly reduce the amounts that are actually consumed. The removal of PBDEs and other POPs from fish may be increased by the unintended splashing of the oily matrix from the sample, which is a typical side effect of cooking in homes. One point worth noting is that PBDEs could also be released from fish into cooking oil.[10] While this effect would reduce the PBDE content in fish, cooking oil is also partly adsorbed to the fish and consumed. The extent of this effect is currently difficult to assess and should stimulate further research.

Partial Transformation of BDE-209 during the Cooking of Fish in the Model System. Transformation was observed for BDE-209 in the model apparatus with fish but not with oil. Hence, the matrix fish has been verified as a key prerequisite for the transformation of BDE-209. Ingredients in fish could either be directly involved in the transformation reaction (in form of a chemical reaction) and/or may act as catalysts by lowering the activation energy of the transformation of BDE-209. In this context, Choi and Chi reported the possibility of a selective hydrodebromination of aromatic brominated compounds in the presence of a "scavenger" of bromine.[27] A suitable scavenger compound in the chemical laboratory process was sodium sulfite.[27] The presence of such a scavenger of bromine in the fish and its absence in plant oil was most likely the reason for the stability of BDE-209 during heating. The temperature seems to be a second factor in this process, because no transformation of BDE-209 was observed during heating of fish in the microwave (where this parameter was lower).

Noteworthy, no transformation was observed in the case of the low-brominated BDE-47. For radicalic substitutions and chemical substitutions, it was found that the stability of PBDEs increases with a decreasing number of bromine substituents.[28−30] For the latter reaction, Granelli et al. noted that high-brominated diphenyl ethers were more prone to this reaction because of their lower electron density at the ring carbons and their higher propensity to the hydrodebromination reaction.[29] Likewise, PBDE congeners with fully brominated rings were decidedly more stable than PBDEs without this feature.[29] For this reason, the main Br_9DE metabolite, BDE-206 (see above), was hydrodebrominated into Br_8DEs on the perbrominated ring moiety. This reaction can lead to BDE-194, BDE-196, and BDE-199, with the latter two being the most prominent Br_8DEs congeners in the fish sample.

Transfer of the Results on Other Polyhalogenated Compounds. Altogether, transformation of PBDEs during the cooking of fish required (i) temperatures above 100 °C and a high number of bromine substitutents. Because of the stronger C−Cl bonds compared to C−Br bonds, the transformation of polychlorinated aromatic compounds during cooking is less likely except for very labile compounds, like DDT (which reacts in the non-aromatic part of the molecule).[31] However, similar effects as observed for BDE-209 may exist for decabromobiphenyl (PBB 209) as well as for highly brominated novel brominated flame retardants (BFRs), such as decabromodiphenyl ethane (DBDPE). The model cooking apparatus developed in this study can be suggested for further future studies on the fate of especially these compounds but also other POPs during cooking.

■ ASSOCIATED CONTENT

Supporting Information

PUF exhaust filter before and after the cooking experiment (Figure S1), fish (*Salmo salar*) before and after the cooking experiment (Figure S2), distribution of Br_8DEs and Br_9DEs in method blanks and heated fish (Figure S3), and recoveries of PBDE congeners in the different compartments of the cooking apparatus (Table S1). This material is available free of charge via the Internet at http://pubs.acs.org.

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Notes

The authors declare no competing financial interest.

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