BFR contamination in UK kitchen utensils: Implications for human exposure via transfer to cooking oil and direct dermal contact

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Introduction

Brominated flame retardants (BFRs) are widely used in textiles, electronic devices and upholstery to protect people from fire. However, this wide application has led to ubiquitous contamination of both the biotic and abiotic environment. This combined with concerns about their toxicity, persistence and capacity for bioaccumulation has led to restrictions and bans on the use BFRs like polybrominated diphenyl ethers (PBDEs). While intentional use of brominated flame retardants (BFRs) in food contact materials is proscribed, many black plastic kitchen utensils like spoons, spatulas and ladles, may contain BFRs as unintentional trace contaminants (UTCs) stemming from incorporation of plastics recycled from other applications where BFRs were employed. Considering the frequent contact of kitchen utensils with food and human skin, there is thus a risk of BFR exposure via consumption of food contaminated during cooking and via dermal contact when handling such utensils. To explore the extent to which UK kitchen utensils are contaminated by BFRs and the associated human exposure risk, we measured the bromine content of 91 kitchen utensils used in Birmingham UK using a hand-held X-ray fluorescence (XRF) spectrometer. Further, 30 utensils with high Br content were selected to measure concentrations of specific BFRs, including 8 polybrominated diphenyl ethers (PBDEs - BDE-28, 47, 99, 100, 153, 154, 183 and 209) and 5 novel BFRs (NBFRs – i.e. pentabromoethylbenzene (PBEB), 2-ethylhexyl-2,3,4,5-tetrabromobenzoate (EH-TBB), 1,2-bis(2,4,6-tribromophenoxy)ethane (BTBPE), bis(2-ethylhexyl)-3,4,5,6-tetrabromo-phthalate (BEH-TEBP) and decabromodiphenyl ethane (DPDPE)). Simulated cooking experiments were conducted on 10 of the 30 samples analysed for BFRs to investigate potential exposure via cooking.

Materials and Methods

Sampling and screening

New utensils were purchased from retail outlets in Birmingham, UK, while used utensils >5 years old were donated by University of Birmingham staff. All utensils were first screened for their bromine content using a hand-held X-ray fluorescence (XRF) spectrometer (Thermo Scientific Niton Analyzer). All utensils (n=18) found to have a Br content > 100 µg/g, together with a random selection of other utensils (n=12) were retained for BFR analysis.

Simulated cooking experiments

10 kitchen utensils shown to contain elevated concentrations of BFRs were subjected to experiments designed to mimic the process of cooking in oil. A small part of kitchen utensil

weighing ~0.5 g, ~ 5 mm × 5 mm × 2 mm was immersed in 0.5 mL olive oil contained in a test tube. The test tube was maintained at 160 ˚C 15 min to simulate the cooking process. This experiment was conducted in triplicate for each utensil tested.

BFR Analysis

Plastic utensils were first cut into small pieces and ground to a powder using a Fritsch Pulverisette 0 cryo-vibratory micro mill (Idar-Oberstein, Germany). Powdered samples were extracted via 15 mins sonication with three successive 10 mL aliquots of hexane. The combined extracts were washed with 98% sulfuric acid, concentrated to incipient dryness and re-dissolved in 200 µL iso-octane prior to analysis of PBDEs and NBFRs by GC-MS. Oil samples generated in the simulated cooking experiments were first dissolved in 3~4 mL hexane and then partitioned against 98% sulfuric acid before concentration ready for GC-MS analysis. Details of the instrumental methods used can be found elsewhere (Kuang et al., 2016).

Results and discussion

Data from Br screening revealed higher Br content in old utensils (≥5 years old) than in new ones. Among the 27 new utensils examined, only one showed Br content > 100 µg/g (Table 1), 2 showed 5µg/g and all the rest not detected (<1µg/g). In contrast, 17 of the 67 old utensils contained Br >100 µg/g, 14 ranged between 10-100µg/g and 36 not detected (<1µg/g).

Table 1: Type, age and Br content of Kitchen Utensils containing >100 µg/g Br

Utensil type	Age (years)	Br content (µg/g)
Thermos cup lid	New	180±5
Ladle	8	350±9
Slotted spatula	8	300±10
Slotted spoon	7	100±5
Solid spoon	7	600±14
Solid spoon	10	6000±69
Solid spoon	5	200±6
Ladle	15	120±7
Slotted spatula	15	400±10
Solid spoon	10	150±8
Masher	7	90±11
Solid spoon	10	170±35
Slotted spoon	10	150±16
Ladle	10	140±14
Slotted spoon	7	100±5
Slotted spoon	14	170±6
Scissors	14	132±7
Scissors	14	4000±43

ΣBFR concentrations in the 18 utensils containing >100 µg/g Br, ranged from 0.1 µg/g to >1000

µg/g (>0.1 %), with a median value of 3.6 µg/g. Interestingly, there was considerable variation between samples with respect to the BFR pattern and no common distribution pattern could be found. BDE-209 was the most frequently detected BFR and usually predominated, but extremely high concentrations of other BFRs like BDE-153 (100 µg/g) and BTBPE (1000 µg/g) were also found in some samples. Table 2 shows a more detailed concentration range for each BFR.

Table 2: Maximum, minimum, and median concentrations (µg/g) of selected BFRs in Kitchen Utensils containing >100 µg/g Br (n=18)

BFR	Maximum	Minimum	Median
BDE-28	0.1	N.D.[a]	N.D.
BDE-47	0.3	N.D.	0.03
BDE-99	0.9	N.D.	0.1
BDE-100	0.1	N.D.	0.02
BDE-153	120	N.D.	0.03
BDE-154	0.2	N.D.	0.01
BDE-183	10	N.D.	0.04
BDE-209	130	0.1	2
PBEB	0.03	N.D.	N.D.
EH-TBB	2	N.D.	N.D.
BTBPE	1100	N.D.	0.08
BEH-TEBP	20	N.D.	0.006
DBDPE	4	N.D.	0.1

[a] not detected

Simulated cooking experiments showed considerable BFR transfer from utensil to oil (up to 50 % of the BFR mass present in the utensil portion tested), suggesting exposure to BFRs during cooking using such utensils is plausible.

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References

Kuang, J., Ma, Y., Harrad, S., 2016. Concentrations of "legacy" and novel brominated flame retardants in matched samples of UK kitchen and living room/bedroom dust. Chemosphere 149, 224-230.