

## Brominated flame retardants in black plastic kitchen utensils:

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1           **Brominated flame retardants in black plastic kitchen**  
2           **utensils: Concentrations and human exposure implications**

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7

## 8 **Abstract**

9 Concerns exist that restricted brominated flame retardants (BFRs) present in waste  
10 polymers may have, as a result of recycling, inadvertently contaminated items not  
11 required to meet flame retardancy regulations (e.g. plastic kitchen utensils). To  
12 investigate the extent to which kitchen utensils are contaminated with BFRs and the  
13 potential for resultant human exposure, we collected 96 plastic kitchen utensils and  
14 screened for Br content using a hand-held X-ray fluorescence (XRF) spectrometer.  
15 Only 3 out of 27 utensils purchased after 2011 contained detectable concentrations of  
16 Br ( $\geq 3 \mu\text{g/g}$ ). In contrast, Br was detected in 31 out of the 69 utensils purchased before  
17 2011. Eighteen utensils with Br content higher than  $100 \mu\text{g/g}$ , and 12 new utensils were  
18 selected for GC-MS analysis of BFRs. BFRs targeted were polybrominated diphenyl  
19 ethers (PBDEs) BDE-28, 47, 99, 100, 153, 154, 183 and 209, and novel BFRs (NBFRs)  
20 pentabromoethylbenzene (PBEB), 2-ethylhexyl-2,3,4,5-tetrabromobenzoate (EH-  
21 TBB), 1,2-bis(2,4,6-tribromophenoxy)ethane (BTBPE), bis(2-ethylhexyl)-3,4,5,6-  
22 tetrabromo-phthalate (BEH-TEBP) and decabromodiphenyl ethane (DBDPE). The  
23 ability of XRF to act as a surrogate metric of BFR concentration was indicated by a  
24 significant (Spearman coefficient = 0.493;  $p=0.006$ ) positive relationship between Br  
25 and  $\Sigma\text{BFR}$  concentration. Measurements of  $\Sigma\text{BFRs}$  were always exceeded by those of  
26 Br. This may be due partly to the presence of BFRs not targeted in our study and also  
27 to reduced extraction efficiency of BFRs from utensils. Of our target BFRs, BDE-209

28 was the most abundant one in most samples, but an extremely high concentration (1,000  
29  $\mu\text{g/g}$ ) of BTBPE was found in one utensil. Simulated cooking experiments were  
30 conducted to investigate BFR transfer from selected utensils (n=10) to hot cooking oil,  
31 with considerable transfer (20 % on average) observed. Estimated median exposure via  
32 cooking with BFR contaminated utensils was 60 ng/day for total BFRs. In contrast,  
33 estimated exposure via dermal contact with BFR-containing kitchen utensils was  
34 minimal.

35

36 **Keywords**

37 BFR, kitchen utensil, recycled plastic, human exposure, UK

38

## 39 **1. Introduction**

40 Brominated flame retardants (BFRs) are a group of organic compounds added widely  
41 to consumer goods such as electronic devices, textiles, and upholstery etc. to meet flame  
42 retardancy regulations. Over the life cycle of such items, BFRs may undergo emission  
43 to the environment and as a consequence are ubiquitous in the environment, including  
44 air (Abdallah et al., 2008; Sun et al., 2016), dust (Cristale et al., 2016; Harrad et al.,  
45 2008; Zhu et al., 2017), soil (Leung et al., 2007; Zhu et al., 2017), sediment (Barón et  
46 al., 2014; Guerra et al., 2010), as well as biota (including humans) exposed to such  
47 media (Carignan et al., 2013; Drage et al., 2017; Shi et al., 2016; Tao et al., 2017; Zhu  
48 et al., 2017). Such environmental contamination, coupled with evidence of their toxicity,  
49 means that BFRs are of great concern. As a consequence, BFRs like polybrominated  
50 diphenyl ethers (PBDEs) have been listed as persistent organic pollutants (POPs) under  
51 the Stockholm Convention and subject to bans and restrictions on their manufacture  
52 and new use in a number of jurisdictions. While to date, the majority of attention has  
53 focused on BFR exposure as a result of emissions from in-use materials, there is  
54 growing realization that the presence of BFRs in waste items also constitutes a potential  
55 problem.

56

57 Waste electrical and electronic equipment (WEEE) may be dismantled to recover  
58 precious metals and plastics, with the plastics recovered being recycled. However, use

59 of recycled plastics containing BFRs in new materials has led to concerns that restricted  
60 BFRs may be present in newly manufactured goods, including those which are not  
61 subject to flame retardancy regulations such as plastic food contact utensils and toys.  
62 To minimise contamination of newly manufactured goods that are not subject to flame  
63 retardancy regulations (e.g. food contact articles and children's toys) with BFRs via use  
64 of BFR-containing recycled polymers, the European Commission has under its  
65 Restriction of Hazardous Substances (RoHS) and WEEE directives, set Low POP  
66 Concentration Limits (LPCLs) for some BFRs to ensure waste plastics exceeding such  
67 limits are not recycled. These values are currently 1,000 ppm for PBDEs (not including  
68 BDE-209) and hexabromocyclododecane (HBCDD). However, reports exist that  
69 plastic goods exceeding LPCLs may still be purchased in the EU. Guzzonato et al.  
70 (2017) investigated 26 samples of toys and food-contact articles purchased from the  
71 European market, finding that ~1/3 of food-contact articles were bromine positive and  
72 around half of the toys examined exceeded LPCLs. Samsonik and Puype (2013)  
73 investigated the Br and BFR content of 30 black plastic kitchen utensils purchased from  
74 the European market, and reported a 30 % detection rate for Br. BDE-209 was the major  
75 BFR found in Br positive samples, with tetrabromobisphenol-A (TBBP-A) and  
76 decabromodiphenyl ethane (DBDPE) detected in some samples as well. Elsewhere,  
77 Chen et al. (2009) found PBDEs, DBDPE, 1,2-bis(2,4,6-tribromophenoxy)ethane  
78 (BTBPE) and polybrominated biphenyls (PBBs) in plastic toys purchased from Chinese  
79 market, while Ionas et al. (2014) found PBDEs and phosphate flame retardants (PFRs)

80 in toys from the European market. The Br concentrations measured by Samsonik and  
81 Puype (2013) ranged from not detected to 2,000 µg/g, while BFR concentrations  
82 measured by Chen et al. (2009) and Ionas et al. (2014) ranged from not detected to  
83 5,000 µg/g, all of which were insufficient to impart flame retardancy, indicating these  
84 BFRs were not intentionally added into kitchen utensils or toys, and highly possibly  
85 came from recycled plastics. Considering the background above, this study seeks to  
86 augment significantly the database on the presence of BFRs in consumer goods by  
87 measuring Br (using a hand-held X-ray fluorescence (XRF) spectrometer) and a range  
88 of BFRs including PBDEs in both used and new plastic kitchen utensils from the UK.  
89 Concentrations of PBDEs and other BFRs in these utensils are compared with LPCL  
90 values, and for the first time, the potential for human exposure arising from consumer  
91 use of such utensils is assessed. This is assessed via examining BFR transfer from  
92 selected utensils to culinary oil during simulated cooking experiments and via  
93 modelling dermal uptake from handling utensils.

94

95 Given the above, the objectives of this study are to: 1) investigate the extent to which  
96 kitchen utensils from the UK market are contaminated by Br and BFRs; 2) evaluate the  
97 extent to which the XRF measurements of Br provide an accurate metric of BFR  
98 concentrations; and; 3) evaluate the potential for human exposure to BFRs as a result  
99 of using plastic kitchen utensils containing BFRs.

100



101 To achieve these objectives, we examined 96 kitchen utensils from the UK. As a first  
102 step, these were all screened for their Br content using hand-held XRF. Thirty of these  
103 utensils were then analysed for their concentrations of BFRs, including 8  
104 polybrominated diphenyl ethers (PBDEs) (BDE-28, 47, 99, 100, 153, 154, 183 and 209),  
105 pentabromoethylbenzene (PBEB), 2-ethylhexyl-2,3,4,5-tetrabromobenzoate (EH-  
106 TBB), BTBPE, bis(2-ethylhexyl)-3,4,5,6-tetrabromo-phthalate (BEH-TEBP) and  
107 DBDPE. Ten representative utensils were then subjected to controlled experiments to  
108 study the transfer of BFRs from kitchen utensils to hot culinary oil.

## 109 **2. Method and materials**

### 110 **2.1 Sampling**

111 New utensils were purchased from retail outlets in Birmingham, UK between Dec 2015  
112 and Jun 2016, while used utensils  $\geq 5$  years old were donated by University of  
113 Birmingham staff. All utensils were first screened for their Br content using a hand-  
114 held XRF spectrometer (Niton<sup>TM</sup> XL3t GOLDD+ XRF Analyzer, Thermo Fisher  
115 Scientific). The platform on which utensils were placed for measurement was pre-  
116 cleaned with ultra-pure water and ethanol, and measured using XRF to ensure no  
117 background interference existed. Measurements of Br were taken at 3-5 randomly  
118 selected points on each utensil to minimize the impact of heterogeneity and the highest  
119 result was recorded. Utensils displaying a Br content  $> 100 \mu\text{g/g}$  (n=18), along with a  
120 further 12 utensils containing  $< 100 \mu\text{g/g}$  Br were selected for measurement of their

121 BFR content.

## 122 **2.2 Chemicals**

123 Native BDE-77 was used as the internal standard (IS) to quantify BDE-28, 47, 99, 100,  
124 as well as PBEB and EH-TBB; BDE-128 as internal standard for BDE-153, 154 and  
125 183; <sup>13</sup>C-BTBPE for BTBPE; <sup>13</sup>C-BEH-TEBP for BEH-TEBP; and <sup>13</sup>C-BDE-209 for  
126 BDE-209 and DBDPE. A mixed IS solution of all the above mentioned internal  
127 standards (500 pg/μL) in iso-octane was prepared. 2,2',3,3',4,5-hexachlorobiphenyl  
128 (PCB-129) was used as a recovery determination standard (RDS) to determine the  
129 recovery of BDE-77, 128, <sup>13</sup>C-BTBPE, <sup>13</sup>C-BEH-TEBP, and <sup>13</sup>C-BDE-209. The RDS  
130 solution was prepared in iso-octane at a concentration of 250 pg/μL. All standards were  
131 purchased from Wellington Laboratories Inc. and all solvents used (acetone, hexane,  
132 iso-octane and methanol) were HPLC grade.

## 133 **2.3 Pre-treatment of plastic samples**

134 Plastic utensil samples were first cut into small pieces and then ground into a powder  
135 using a Fritsch Pulverisette 0 cryo-vibratory micro mill (Idar-Oberstein, Germany).  
136 This was achieved by adding the sample along with a 25 mm diameter stainless steel  
137 ball to the stainless steel grinding mortar (50 mL volume), cooled with liquid nitrogen.  
138 The cryogenically-cooled sample was then ground at a vibrational frequency of 30 Hz  
139 for 5 min and repeated 2-3 times. After 1 min vortexing with 10 mL hexane to achieve  
140 complete mixing, the resultant plastic powder was then extracted under 15 min

141 sonication and supernatant was then collected. The process of vortexing and  
142 ultrasonication were repeated for 2 more cycles and for the last extraction, the  
143 supernatant was left in contact with the sample overnight before collection to maximise  
144 recoveries. Combined extracts were reduced in volume to ~ 2 mL under a gentle stream  
145 of nitrogen gas, before mixing with 3-4 mL 98 % sulfuric acid. The hexane-acid mixture  
146 was then vortexed for 20 s followed by centrifugation at 2,000 g for 5 min. The  
147 supernatant was then collected. To ensure complete transfer, the residue was rinsed with  
148 hexane (2 mL) three times. The combined supernatant was then reduced to incipient  
149 dryness under a gentle stream of nitrogen gas. The final concentrate was re-dissolved  
150 in 200  $\mu$ L PCB-129 RDS solution prior to analysis of PBDEs and NBRs by GC-MS.

#### 151 **2.4 Experiments examining BFR transfer from utensil to culinary oil**

152 Ten kitchen utensils shown to contain elevated concentrations of BFRs were subjected  
153 to experiments designed to mimic the process of cooking in oil. A small portion of  
154 kitchen utensil weighing ~0.05 g, ~ 5 mm  $\times$  4 mm  $\times$  2 mm was immersed in 0.5 mL  
155 olive oil in a test tube. The test tube was maintained at 160  $^{\circ}$ C for 15 min to simulate  
156 the cooking process and oil collected for analysis. After “cooking” each utensil, the  
157 experiment was repeated twice more using the same aliquot of the utensil to investigate  
158 the impact of repeated cooking in oil on BFR transfer efficiency. The collected oil  
159 samples were first diluted in 3~4 mL hexane, before added with 5~6 mL 98 % sulfuric  
160 acid. The hexane-acid mixture then underwent the same process describe in section 2.3,  
161 before dissolution in 200  $\mu$ L PCB-129 RDS solution for analysis.

## 162 **2.5 GC-MS Protocols**

163 PBDEs and NBFRs were analysed by GC-MS in electron capture negative ionisation  
164 (ECNI) mode using the same method to our previous study (Kuang et al., 2016). For  
165 some plastic kitchen utensil samples with extremely high BDE-209 concentrations and  
166 the corresponding oil extracts, recoveries of  $^{13}\text{C}$ -BDE-209 could reach 400 % ~ 1000 %,  
167 which exceeded the normal range. The reason is that when intensity is too high, the  
168 overlap between response peaks of ions on mass spectrometer could not be neglected,  
169 especially when peaks are very close. In this case, response of  $^{13}\text{C}$ -BDE-209 (m/z 492.6,  
170 494.6) was severely interfered by the extremely high response of BDE-209 (m/z 486.6,  
171 488.6), so an exceptional high “apparent recovery” was observed. To address this issue,  
172 we re-injected affected samples in electron ionisation (EI) mode and satisfactory  
173 recoveries were obtained, as interference between the quantifying ions used for BDE-  
174 209 (m/z 799.4, 801.4) and  $^{13}\text{C}$ -BDE-209 (809.4, 811.4) was weaker given the greater  
175 difference in m/z values.

## 176 **2.6 QA/QC**

177 For measurement of Br, the XRF analyzer was calibrated regularly using manufacturer-  
178 supplied solid disk standards. And for BFR measurement, three blank oil samples were  
179 analysed along with experimental samples. Satisfactory results were obtained with  
180 recoveries of internal standards ranging from 60 %~130 % (Table S1) with all native  
181 compounds not detected, except BDE-209 (Table S2). Concentrations of BDE-209 in

182 oil samples were corrected for blank contamination by subtracting the mean value  
183 detected in blanks. Satisfactory recoveries of 70 %~130 % were obtained for both  
184 kitchen utensil plastic (Table S3) and cooking experiment oil (Table S4) samples.

185

186 In addition, to evaluate BFR losses during cooking experiments, a matrix spike  
187 experiment was conducted 5 times by spiking known amounts of all target compounds  
188 and internal standards into blank oil samples before the cooking experiment. These  
189 matrix spike samples were then analysed and recoveries of all compounds calculated  
190 (Table S5). Recoveries of all compounds showed good performance ranging from 70 %  
191 to 170 %, and recoveries of target compound showed consistent deviation with  
192 coordinating internal standard recoveries (Table S5), ensuring a precise quantification.

### 193 **3. Results and discussion**

#### 194 **3.1 Bromine content of kitchen utensils**

195 Table 1 reports Br concentrations in the utensil samples analysed using hand-held XRF.  
196 Of the 96 samples analysed, 69 were reported by the donors to be 5 years or older, 6  
197 were aged 2 years, while 21 were purchased for this study between December 2015 and  
198 July 2016. It should be noted that “age” in this study refers only to the donor-reported  
199 date of purchase to the nearest year. It is important to note not only the uncertainty  
200 associated with such self-reported data, but that the date of purchase does not equate to  
201 the date of manufacture but to the date of availability on the market. Notwithstanding

202 this, for convenience, we use “age” as an abbreviation of “date of availability on the  
 203 market” from herein. Table 1 also lists the utensil type, with the main categories being:  
 204 spoons (n=33), spatulas (n=18) and ladles (n=12). Of the 27 utensils aged < 5 years,  
 205 only 1 (3.7 %) contained >100 µg Br/g, 2 (7.4 %) contained ~ 5 µg Br/g, with the  
 206 remaining 24 (88.9 %) containing <3 µg Br/g. In contrast, for utensils aged ≥5 years,  
 207 17 (24.6 %) contained > 100 µg Br/g, 13 (18.9 %) contained between 5 and 100 µg Br/g,  
 208 and 34 (49.3 %) containing <3 µg Br/g. Given this apparent dichotomy between “older”  
 209 and “newer” utensils, we evaluated the significance of this using non-parametric  
 210 statistical tests as our data did not display a normal distribution. We first conducted a  
 211 Mann-Whitney rank test to compare Br concentrations between the two age groups.  
 212 This revealed Br concentrations to be significantly greater in utensils ≥5 years old  
 213 (p=0.016). This was consistent with a Spearman correlation analysis which showed  
 214 utensil age and Br content to be significantly and positively correlated (r=0.237,  
 215 p=0.020).

216

217 **Table 1 Bromine Concentrations (µg/g) in Kitchen Utensils**

Sample # <sup>a</sup>	Utensil type	Br content, µg/g	Date of purchase <sup>b</sup>	Age, years
P1	Solid spoon	<3	2015	New
P2	Thermos cup lid	<3	2015	New
P3	Thermos cup lid	180	2015	New
P4	Thermos cup lid	<3	2015	New
P5	Thermos cup lid	<3	2015	New
P6	Food package	<3	2015	New
P7	Food package	<3	2015	New
P8	Food package	<3	2015	New

P9	Food package	<3	2015	New
P10	Ladle	350	2008	8
P11	slotted spatula	300	2008	8
P12	spaghetti server	<3	2013	2
P13	Solid spatula	<3	2013	2
P14	solid spatula	<3	2013	2
P15	Food clip	<3	2013	2
P16	slotted spoon	100	2009	7
P17	Solid spoon	600	2009	7
P18	Solid Spoon	6,000	2006	10
P19	solid spoon (grip)	200	Before 2011	>5
P20	Ladle	120	2001	15
P21	slotted spatula	400	2001	15
P22	solid spoon (grip)	150	2006	10
P23	Masher	90	2009	7
P24	solid spoon (grip)	170	2006	10
P25	slotted spoon (grip)	150	2006	10
P26	Ladle (grip)	140	2006	10
P27	slotted spoon	100	2009	7
P28	slotted spoon (grip)	170	2002	14
P29	Scissors	130	2002	14
P30	Scissors	4,000	2002	14
	slotted spatula	<3	2009	7
	Solid spatula	<3	2009	7
	Ladle	<3	2009	7
	slotted spatula	<3	2009	7
	slotted spoon	40	Before 2011	>5
	Solid Spoon	<3	2007	9
	slotted spoon	<3	2001	15
	Solid Spoon	<3	2016	New
	Ladle	<3	2016	New
	slotted spatula	<3	2016	New
	Solid spoon	30	2009	7
	Solid spoon	<3	2009	7
	Masher	<3	2008	8
	slotted spatula	50	2008	8
	slotted spatula	<3	2008	8
	spaghetti server	<3	2008	8
	Solid spoon	<3	2008	8
	Ladle	<3	2008	8
	slotted spoon	85	2006	10
	skimming spoon	<3	2006	10

---

Masher	<3	2006	10
Not recorded	<3	2006	10
Not recorded	<3	2006	10
Not recorded	<3	2006	10
Not recorded	<3	2006	10
Not recorded	<3	2006	10
Cut board	10	2009	7
Spatula	20	2009	7
Ladle	<3	2009	7
Solid spoon	<3	2006	New
slotted spatula	<3	1996	20
Solid spoon	<3	1996	20
Ladle	<3	1996	20
slotted spoon	20	1996	20
Masher	<3	1996	20
Spatula	<3	1998	18
dotted spoon	<3	1998	18
Masher	<3	1998	18
Spatula	10	2002	14
Masher	<3	2002	14
Scissors	60	2002	14
Whisk	<3	2014	2
Masher	<3	2014	2
spaghetti server	10	2001	15
slotted spatula	<3	2001	15
Ladle	<3	2001	15
slotted spoon	<3	2001	15
Masher	30	2001	15
solid spoon	<3	2016	New
slotted spatula	<3	2016	New
Masher	<3	2016	New
Ladle	5	2016	New
slotted spoon	<3	2016	New
slotted spoon	<3	2016	New
slotted spoon	7	2016	New
Scissors	<3	2016	New
solid spoon	<3	2011	5
slotted spatula	<3	2011	5
Ladle	<3	2011	5
Ladle	8	2011	5
Fork	<3	2011	5
Spatula	<3	Before 2011	>5

---



Solid spoon	50	Before 2011	>5
Solid spoon	<3	Before 2011	>5
Slotted spoon	60	Before 2011	>5
Skimming spoon	<3	Before 2011	>5

218 <sup>a</sup>Sample # refers to sample analysed for BFR content – see Table 2. Samples not  
 219 assigned a number were not analysed for their BFR content

220 <sup>b</sup>Owner’s estimate of purchase date

### 221 **3.2 BFR concentrations in kitchen utensils**

222 Based on the Br concentration data, those utensils containing >100 µg Br/g (n=18) were  
 223 subjected to GC-MS determination of their BFR content, together with 12 utensils  
 224 containing <100 µg Br/g to provide context. These 30 samples are numbered 1~30 in  
 225 Table 1.

226

227 Table 2 shows that utensils with high Br content (>100 µg/g) display a higher BFR  
 228 concentration than those indicated by XRF to contain <100 µg/g Br. We tested the  
 229 statistical significance of this relationship using non-parametric tests as our data did not  
 230 display a normal distribution. Specifically, a Mann-Whitney rank test showed the  
 231 difference to be statistically significant (p=0.007), with the positive relationship  
 232 between Br and BFR concentrations confirmed by Spearman correlation analysis  
 233 (r=0.493, p=0.006). However, more detailed inspection of Table 2 reveals there is  
 234 substantial discrepancy between our BFR and Br data for the same samples. To be  
 235 explicit, our ΣBFR measurements are always lower than the corresponding Br  
 236 measurements – and in some cases substantially so, for example, sample 18 contained  
 237 6,000 µg Br/g, but displayed a ΣBFR concentration of 0.6 µg/g. This is most likely due

238 to some compounds not included in our list of target BFRs for example TBBP-A, and/or  
239 low extraction efficiency for BFRs using our method.

240

241 We first tested the hypothesis that the discrepancy between Br and  $\Sigma$ BFR was because  
242 the former was due to the presence of one or more BFRs not targeted by our GC-MS  
243 analyses. To do so, we studied sample 18 in more detail. Tentative support for this  
244 explanation is supplied by the observation of several unidentified peaks on the m/z 79  
245 and 81 traces in the GC mass chromatogram for sample 18. Hence, following solvent  
246 exchange from iso-octane to methanol we re-analysed this sample on a LC-high  
247 resolution MS system (UPLC-Orbitrap-MS, Thermo Fisher Scientific, Bremen,  
248 Germany) in an attempt to identify BFRs not quantified via our GC-MS method such  
249 as TBBP-A or HBCDD. However, this did not provide an obvious explanation for the  
250 discrepancy, and thus incomplete extraction efficiency can not be ruled out as a cause  
251 in this instance at least. To avoid dissolving the plastic during BFR extraction and thus  
252 expedite more rapid analysis, a low polarity aliphatic solvent (hexane) was chosen for  
253 extraction. We note that other studies have used different solvents (Allen et al. (2008),  
254 Aldrian et al. (2015) used toluene, and Gallen et al. (2014) used dichloromethane), and  
255 thus our BFR measurements may be underestimates of the true value. Also, as TBBP-  
256 A is a reactive BFR which binds more firmly with polymers than additive BFRs like  
257 PBDEs, hexane may be less effective at extracting it from polymers, leading it to be not  
258 detected even in our LC-high resolution MS screening.



Table 2 BFR concentrations in kitchen utensils, ng/g

Sample #	BDE-28	PBEB	BDE-47	BDE-100	BDE-99	EH-TBB	BDE-154	BDE-153	BDE-183	BTBPE	BEH-TEBP	BDE-209	DBDPE	ΣBFRs, μg/g	Br, μg/g
P1	<0.2	0.2	6.3	7.0	42	<0.2	7.8	16	36	530	<0.2	1,100	72	1.8	<3
P2	<0.2	<0.2	37.4	6.9	26	<0.2	1.3	2.7	14	78	<0.2	620	16	0.8	<3
P3	<0.2	<0.2	110	36	150	<0.2	12	22	100	1,200	<0.2	2,500	23	4.1	180
P4	<0.2	<0.2	0.5	<0.2	1.4	<0.2	0.4	1.1	16	3.8	27	260	<9.2	0.3	<3
P5	<0.2	<0.2	1.2	0.3	2.3	<0.2	<0.4	0.5	3.9	5.4	<0.2	37	<9.2	0.1	<3
P6	<0.2	0.2	<0.2	0.5	4.6	0.5	<0.4	0.7	<1.0	<1.0	150	14	12	0.2	<3
P7	<0.2	<0.2	<0.2	<0.2	1.3	<0.2	<0.4	<0.4	<1.0	<1.0	<0.2	<2.6	<9.2	<0.01	<3
P8	<0.2	<0.2	<0.2	<0.2	1.1	<0.2	<0.4	<0.4	<1.0	<1.0	<0.2	<2.6	<9.2	<0.01	<3
P9	<0.2	<0.2	<0.2	<0.2	0.7	<0.2	<0.4	1.1	4.4	8.4	<0.2	340	290	0.6	<3
P10	130	<0.2	360	68	330	<0.2	48	90	330	1,400	<0.2	17,000	<9.2	20	350
P11	100	<0.2	210	82	93	<0.2	4.6	21	36	60	<0.2	2,200	<9.2	2.8	300
P12	<0.2	<0.2	7.4	1.3	7.7	<0.2	0.9	1.8	14	<1.0	<0.2	1300	<9.2	1.4	<3
P13	0.6	<0.2	25	4.8	30	<0.2	2.9	6.2	34	1.1	<0.2	2,500	<9.2	2.6	<3
P14	<0.2	<0.2	11	4.1	21	<0.2	3.6	5.6	24	<1.0	<0.2	1,200	<9.2	1.3	<3
P15	<0.2	<0.2	38	9.9	49	<0.2	5.4	9.1	46	<1.0	<0.2	2,100	<9.2	2.3	<3
P16	<0.2	<0.2	9.5	<0.2	10	<0.2	8.9	36	27	<1.0	6.8	660	58	0.8	100
P17	<0.2	<0.2	36	34	180	<0.2	1,000	1,800	1,600	<1.0	<0.2	1,000	340	6.0	600
P18	<0.2	1.1	15	82	100	<0.2	21	14	23	210	<0.2	140	<9.2	0.6	6,000
P19	<0.2	<0.2	8.8	1.8	10	<0.2	1.3	2.3	8.8	<1.0	350	260	110	0.8	200
P20	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.4	<0.4	<1.0	<1.0	<0.2	81	<9.2	0.1	120
P21	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.4	<0.4	<1.0	<1.0	17	220	<9.2	0.2	400

P22	<0.2	4.0	57	30	240	<0.2	15	25	130	<1.0	46	110,000	5,500	120	150
P23	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.4	<0.4	<1.0	<1.0	<0.2	78	<9.2	0.1	90
P24	120	11	1,000	110	530	900	40	170	139	280	30,000	8,100	5,200	47	170
P25	79	14	1,000	110	370	950	23	110	66	180	25,000	1,900	3,700	34	150
P26	15	8.4	970	43	130	830	5.2	29	49	200	22,000	2,700	7,200	34	140
P27	<0.2	<0.2	<0.2	<0.2	9.0	<0.2	3.7	14	45	35	10	2,500	280	2.9	100
P28	64	8.3	82	30	260	<0.2	30	560	1,100	1,500	140	81,000	5,700	90	170
P29	<0.2	<0.2	10	0.2	12	<0.2	7.6	1,600	180	18,000	5.7	3,200	420	23	130
P30	<0.2	33	<0.2	<0.2	12	<0.2	210	120,000	13,000	1,100,000	<0.2	140,000	1,900	1,400	4,000

262 Given our observation that Br concentrations were significantly higher in samples  $\geq 5$   
263 years old, than in younger utensils, we examined our data for similar age-related  
264 differences in  $\Sigma$ BFR concentrations, again using non-parametric tests in accordance  
265 with the distribution of our data. A Mann-Whitney rank test found significantly  
266 ( $p=0.014$ ) higher  $\Sigma$ BFR concentrations in utensils  $\geq 5$  years old than in those  $< 5$  years  
267 in age. This was consistent with Spearman correlation analysis ( $r=0.501$ ,  $p=0.005$ ) that  
268 showed a positive relationship between BFR concentration and utensil age. These  
269 findings are likely attributable to two main factors: (1) the introduction in restrictions  
270 in use of PBDEs in the mid-2000s onwards, and (2) the more recent introduction of  
271 restrictions on the recycling of BFR-treated plastics.

272

273 In terms of the BFR distribution pattern, BDE-209 was the most abundant BFR detected  
274 and in 17 out of 30 samples (56.7%), BDE-209 accounted for more than 70 % of  $\Sigma$ BFR.  
275 This is consistent with the fact that BDE-209 is mainly used in hard plastics like  
276 polyamide (Arias, 2001 cited by Alaei et al., 2003) which is used widely in kitchen  
277 utensils. Aside of this general predominance of BDE-209 however, the BFR pattern  
278 varied widely between individual utensils. For example, while P22, P23 and P24, which  
279 came from the same donor and were purchased at the same time, all contained a high  
280 percentage of BEH-TEBP (65 % ~ 75 %  $\Sigma$ BFR); P10 and P11 (donated by the same  
281 individual and purchased at the same time) contained substantial contributions of less  
282 brominated PBDEs like BDE-47 and -99; while P29 and P30 (which were the two  
283 handles of the same pair of scissors) were dominated (~80 %  $\Sigma$ BFR) by BTBPE. These

284 3 examples indicate that as well as age, production batch may be an important additional  
285 factor influencing the Br and BFR concentration and pattern.

### 286 **3.3 BFR transfer from utensil to oil in simulated cooking process**

287 Table 3, as well as Figures 1 and 2 show the transfer of individual BFRs and  $\Sigma$ BFR  
288 from the aliquots of utensils subjected to the simulated cooking experiments. The  
289 percentage transfer in Figure 1 and 2 was calculated as  
290  $r = m_{BFR-oil} / (c_{BFR-plastic} \times m_{plastic}) \times 100\%$ , where  $m_{BFR-oil}$  is the mass of BFR extracted  
291 by oil, measured by GC-MS,  $c_{BFR-plastic}$  is BFR concentration in plastic utensils and  
292  $m_{plastic}$  is mass of plastic used in cooking experiment. Transfer was substantial for all  
293 compounds, especially during the 1<sup>st</sup> cooking exposure (batch 1), ranging from 20 % to  
294 100 %. The extent of transfer decreased in the order batch 1 > batch 2 > batch 3 and with  
295 increasing degree of bromination for PBDEs. In particular, while BDE-209 was  
296 abundant in most utensils, its transfer to oil was negligible in 6 of 10 cases. However,  
297 for samples P22, P24, P28 and P30 that contained BDE-209 concentrations in the range  
298 10~100  $\mu\text{g/g}$ , more substantial transfer was observed. The generally lower transfer  
299 efficiency of BDE-209 in our experiments is likely due to a combination of lower  
300 solubility in oil of BDE-209 compared to other BFRs, alongside greater binding of  
301 BDE-209 to plastic.

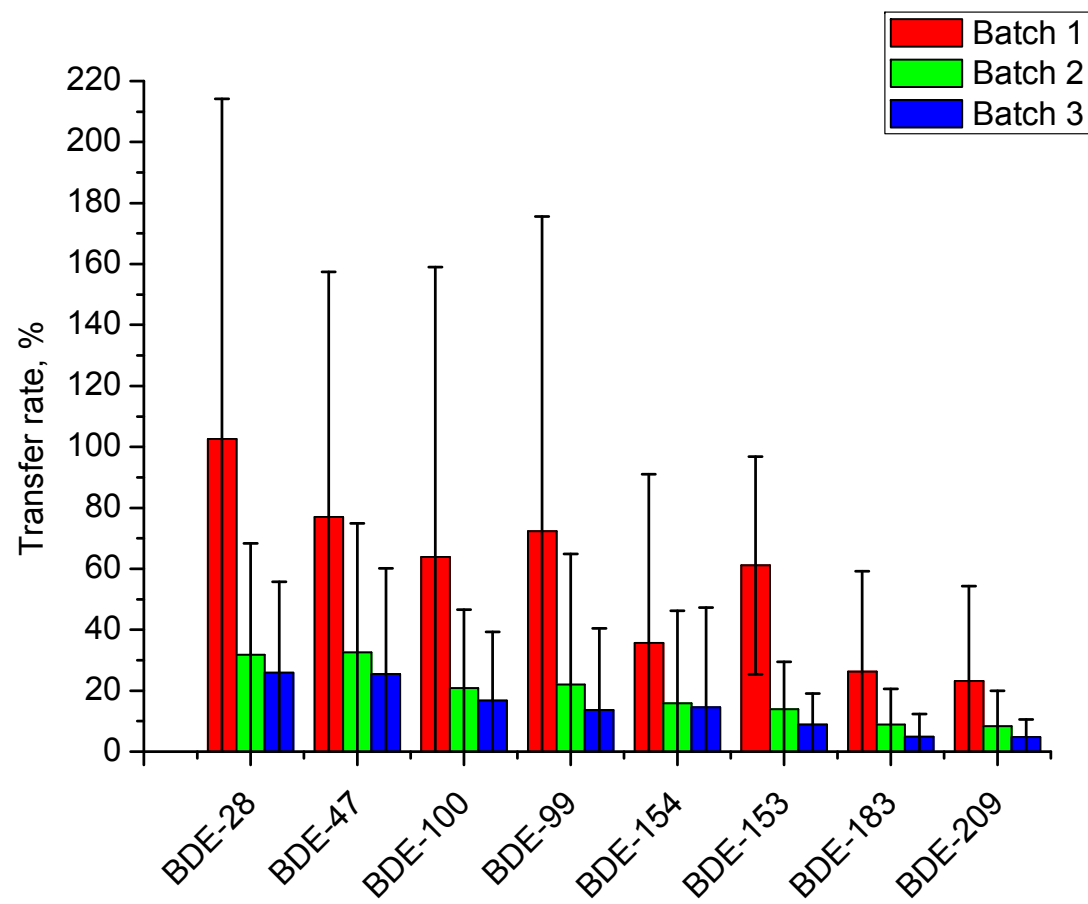
302

303 In some cases, the transfer exceeded 100 %. This may be attributable to a number of  
304 factors, namely: (a) inhomogeneous distribution of BFRs in the kitchen utensils which

305 could result in the BFR content of the aliquot of the utensil subjected to cooking  
306 differing from that in the aliquot used to determine BFR concentration; (b) that hot oil  
307 may be a more effective solvent for extracting BFRs from kitchen utensils than hexane;  
308 and (c) where transfers >100 % are observed for lower PBDEs, this may indicate some  
309 degree of thermal debromination of higher homologues such as BDE-209.



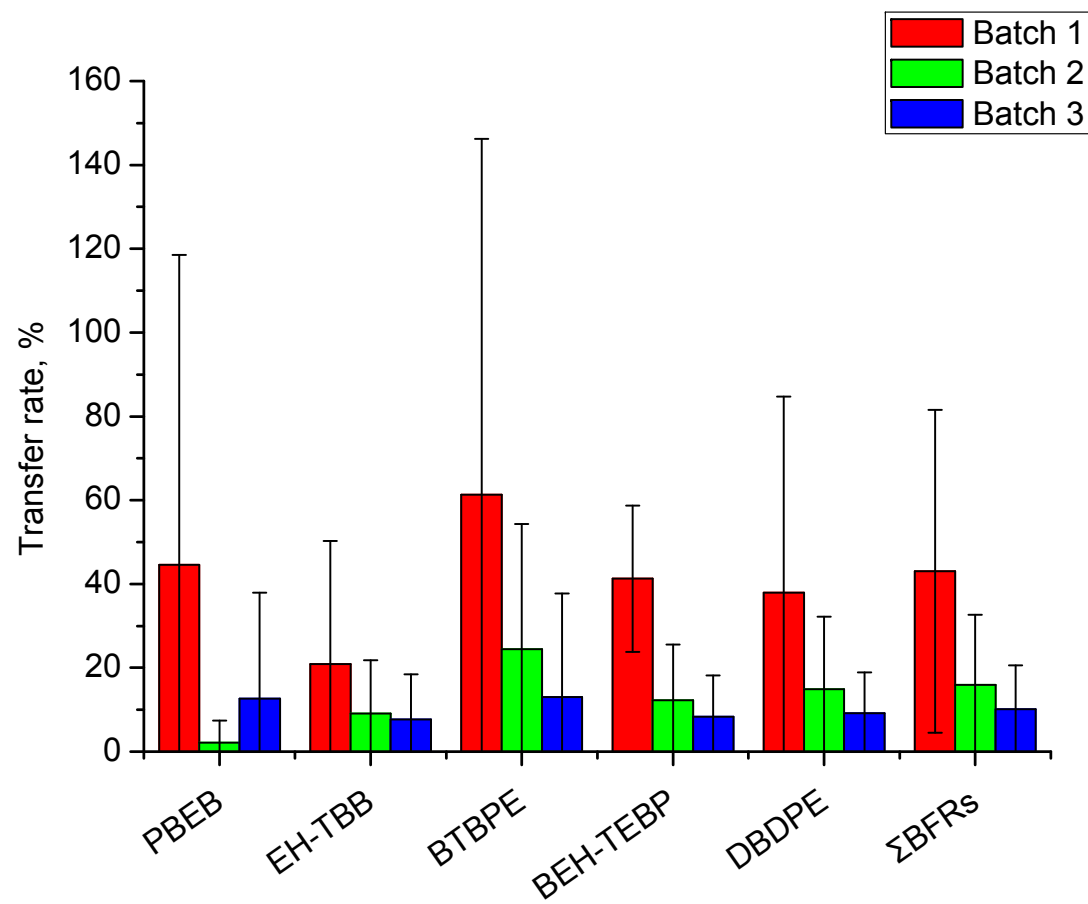
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311

312

**Figure 1** Average percentage transfer of PBDEs from kitchen utensils in simulated cooking experiments (y-error bar represents  $\sigma_{n-1}$ )



**Figure 2** Average percentage transfer of NBRs and  $\Sigma$ BRs from kitchen utensils in simulated cooking experiments (y-error bar

**Table 3 BFR transfer from kitchen utensils (ng BFR/g plastic<sup>a</sup>) in simulated cooking experiments**

Sample	BDE-28	PBEB	BDE-47	BDE-100	BDE-99	EH-TBB	BDE-154	BDE-153	BDE-183	BTBPE	BEH-TEBP	BDE-209	DBDPE	$\Sigma$ BFR <sub>s</sub>
P1	<0.2	0.2	6.3	7.0	42	<0.2	7.8	16	36	530	<0.2	1,100	72	1,800
Batch1	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.4	14	<1.0	100	<0.2	62	<9.2	170
Batch2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.4	<0.4	<1.0	<1.0	<0.2	<2.6	<9.2	<16
Batch3	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.4	<0.4	<1.0	<1.0	<0.2	<2.6	<9.2	<16
P3	<0.2	<0.2	110	35.6	150	<0.2	12	22	100	1,200	<0.2	2,500	23	4,100
Batch1	<0.2	<0.2	<0.2	<0.2	13	<0.2	<0.4	7.4	<1.0	<1.0	<0.2	<2.6	<9.2	21
Batch2	<0.2	<0.2	5.8	<0.2	0.3	<0.2	<0.4	<0.4	<1.0	<1.0	<0.2	11	<9.2	17
Batch3	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.4	<0.4	<1.0	<1.0	<0.2	<2.6	<9.2	<16
P10	130	<0.2	360	68	330	<0.2	48	90	330	1,400	<0.2	17,000	<9.2	20,000
Batch1	270	2.0	410	170	170	<0.2	<0.4	21	<1.0	92	<0.2	4.8	<9.2	1,100
Batch2	86	1.2	110	41	41	<0.2	<0.4	5.2	<1.0	<1.0	<0.2	3.6	<9.2	290
Batch3	68	1.3	85	26	30	<0.2	<0.4	4.0	<1.0	20	<0.2	10	<9.2	250
P11	100	<0.2	210	81	93	<0.2	4.6	21	36	60	<0.2	2,200	<9.2	2,800
Batch1	200	2.3	320	140	160	<0.2	<0.4	21	<1.0	150	0.6	4.6	<9.2	1,000
Batch2	63	0.5	57	14	21	<0.2	<0.4	2.3	<1.0	35	<0.2	<2.6	<9.2	190
Batch3	53	0.9	33	6.1	12	<0.2	<0.4	<0.4	<1.0	<1.0	<0.2	<2.6	<9.2	100
P17	<0.2	<0.2	36	34	180	<0.2	1,000	1,800	1,600	<1.0	<0.2	990	250	6,000
Batch1	<0.2	<0.2	12	<0.2	59	<0.2	210	560	1,300	<1.0	<0.2	41	<9.2	2,200
Batch2	<0.2	<0.2	4.9	14	16	<0.2	54	140	310	I <sup>b</sup>	<0.2	5.0	<9.2	740

Batch3	<0.2	<0.2	3.5	21	19	<0.2	86	180	330	I <sup>b</sup>	<0.2	<2.6	<9.2	920
P18	<0.2	1.1	15	82	100	<0.2	21	14	23	210	<0.2	140	<9.2	600
Batch1	<0.2	2.3	36	140	340	<0.2	38	13	<1.0	100	<0.2	8.4	<9.2	670
Batch2	<0.2	<0.2	19	52	140	<0.2	20	4.8	<1.0	52	<0.2	<2.6	<9.2	290
Batch3	<0.2	<0.2	12	33	91	<0.2	22	4.3	<1.0	30	<0.2	2.7	<9.2	200
P22	<0.2	4.0	57	30	249	<0.2	15	25	130	<1.0	46	110,000	5,500	120,000
Batch1	<0.2	<0.2	11	<0.2	130	<0.2	4.1	610	59	270	17	100,000	6,400	110,000
Batch2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.4	240	21	390	<0.2	34,000	1,900	36,000
Batch3	<0.2	0.4	5.7	<0.2	<0.2	<0.2	<0.4	94	<1.0	390	<0.2	16,000	1,200	18,000
P24	120	11	1,000	110	530	900	40	170	130	280	30,000	8,100	5,200	47,000
Batch1	<0.2	4.9	990	55	230	370	15	61	43	220	7,800	3,400	3,300	17,000
Batch2	<0.2	<0.2	810	29	78	160	4.5	16	10	220	3,100	1,000	1,200	6,700
Batch3	<0.2	<0.2	920	22	53	140	2.9	7.9	5.3	200	1,800	570	820	4,600
P28	64	8.3	82	30	260	<0.2	30	560	1,100	1,500	140	81,000	5,700	90,000
Batch1	7.7	3.3	34	<0.2	77	4.5	16	620	870	1,100	82	48,000	4,300	55,000
Batch2	<0.2	<0.2	9.8	<0.2	53	<0.2	9.4	260	380	430	36	21,000	2,200	25,000
Batch3	<0.2	5.7	<0.2	<0.2	14	<0.2	2.7	100	150	170	26	10,000	1,100	12,000
P30	<0.2	33	<0.2	<0.2	12	<0.2	210	120,000	13,000	1,100,000	<0.2	140,000	1,900	1,400,000
Batch1	<0.2	7.2	<0.2	<0.2	0.2	<0.2	66	39,000	2,900	100,000	<0.2	32,000	220	180,000
Batch2	<0.2	4.6	<0.2	<0.2	<0.2	<0.2	25	12,000	1,200	56,000	<0.2	15,000	120	85,000
Batch3	<0.2	3.0	<0.2	<0.2	<0.2	<0.2	30	13,000	1,200	51,000	<0.2	15,000	140	81,000
Batch4	<0.2	2.7	<0.2	<0.2	<0.2	<0.2	24	7,400	980	40,000	0.2	13,000	120	62,000

318 <sup>a</sup>Amount of BFRs extracted in oil (Batch 1, 2, 3) is expressed as  $m_{\text{BFR-oil}}/m_{\text{plastic}}$ , i.e. mass of BFR detected in each olive oil extract divided by the  
319 mass of plastic tested.

320 <sup>b</sup>Interference prevented quantification.

### 321 **3.4 Preliminary exposure assessment**

322 We considered two pathways via which human exposure to BFRs in kitchen utensils  
323 may occur: (a) transfer to food when cooking, and (b) transfer through dermal contact.

324 The following are preliminary evaluations of the likely magnitude of human exposure  
325 via such pathways.

#### 326 **3.4.1 Exposure via cooking**

327 Exposure via cooking was estimated based on the results of our simulated cooking  
328 experiments – note that as some utensils for which BFR concentrations were determined  
329 were unlikely to come into contact with hot oil during use (e.g. scissors), these utensils  
330 (P2-P5, P6-P9, plus P29 and 30) were excluded from our estimations. To estimate  
331 exposure resulting from contact between the utensil and hot oil and subsequent  
332 ingestion of the oil we made several assumptions. The first of these are that: 1) over the  
333 useful “lifetime” of every 200 mL oil (assumed 1 week) used for deep frying, the utensil  
334 is in contact with oil at 160 °C for a total period over that week of 15 min; and 2) the  
335 extent of BFR transfer is proportional to the specific surface area (i.e. surface area per  
336 unit utensil volume) of the utensil in contact with oil. We further assumed that the  
337 utensil dimensions likely to come into contact with oil during cooking are 10 cm × 8  
338 cm × 2 mm (equivalent to that of a typical spatula), yielding a specific surface area of  
339 10 cm<sup>-1</sup>. This compares quite closely with the specific surface area of 19 cm<sup>-1</sup> of the 5  
340 mm × 4 mm × 2 mm plastic cuboids used in our cooking experiments. Based on these

341 assumptions, we estimated the amount of BFR transferred from kitchen utensils to hot  
342 oil during cooking via the equation below.

$$343 \quad c_{BFR-oil} = (c_{BFR-utensil} \times m_{utensil} \times r_{real}) / V_{oil} \quad (1)$$

344 Where:

345  $c_{BFR-oil}$  is BFR concentration transferred to hot cooking oil (ng/mL);

346  $c_{BFR-utensil}$  is BFR concentration (ng/g) in kitchen utensils coming into contact with hot  
347 oil;

348  $m_{utensil}$  is mass of utensil contact with hot oil when cooking, whose size is 10 cm × 8  
349 cm × 2 mm, and for density, a value of 1.4 g/cm<sup>3</sup> was applied based on the average  
350 measured value for several utensils on this study. So  $m_{utensil} = V_{utensil} \times \rho_{utensil} = 10 \text{ cm} \times$   
351  $8 \text{ cm} \times 2 \text{ mm} \times 1.4 \text{ g/cm}^3 = 22.4 \text{ g}$ ;

352  $r_{real}$  is BFR transfer rate (unitless) in real-life scenario and is calculated based on  
353 transfer rate obtained in cooking experiment ( $r_{exp}$ ), specific surface area of utensil in  
354 experiment ( $A_{exp}$ ) and in real-life scenario ( $A_{real}$ ):

$$355 \quad r_{real} = \frac{A_{real}}{A_{exp}} \times r_{exp} = \frac{10 \text{ cm}^{-1}}{19 \text{ cm}^{-1}} \times r_{exp} = 0.53r_{exp} ;$$

356  $V_{oil}$  is volume of oil involved in cooking which is assumed to be 200 mL.

357

358 Thus,

$$359 \quad c_{BFR-oil} = \frac{c_{BFR-utensil} \times 22.4 \text{ g} \times 0.53r_{exp}}{200\text{mL}} = 0.059c_{BFR-utensil}r_{exp} \text{ ng/mL} \quad (2)$$

360

361 According to 2015-2020 dietary guidelines for Americans (U.S. DHHS and DA, 2015),  
362 the recommended daily oil intake for an adult is 27 g. We assume that deep fried oil  
363 accounts for 15 % of daily oil intake on average, and that as noted on the food  
364 information label of the oil used, the density of olive oil was 0.9 g/mL; thus the daily  
365 BFR exposure amount is:

$$\begin{aligned} E_{BFR-oil} &= 15\% \times c_{BFR-oil} \times \frac{27 \text{ g/day}}{0.9 \text{ g/mL}} \\ 366 \quad &= 15\% \times 0.059 c_{BFR-utensil} r_{exp} \frac{\text{ng}}{\text{mL}} \times \frac{27 \text{ g/day}}{0.9 \text{ g/mL}} \quad (3) \\ &= 0.27 c_{BFR-utensil} r_{exp} \text{ ng/day} \end{aligned}$$

367

368 Here we use median and maximum concentration of the 20 utensils (P1, P10~P28) as  
369 the value of  $c_{BFR-utensil}$  for median and high exposure scenario estimates, and the mean  
370 transfer rate of the 3 batches in the cooking experiments is used for the value of  $r_{exp}$ .

371 The resultant exposure estimates are shown in Table 4.

372

373

**Table 4 BFR exposure (ng/day) via cooking in median and high exposure scenarios<sup>a</sup>**

	BDE-28	PBEB	BDE-47	BDE-100	BDE-99	EH-TBB	BDE-154	BDE-153	BDE-183	BTBPE	BEH-TEBP	BDE-209	DBDPE	ΣBFRs
<i>r<sub>exp</sub></i>	53.4%	19.8%	45.0%	37.6%	40.0%	12.5%	22.3%	27.9%	13.2%	32.9%	20.6%	11.7%	20.7%	-
Median	NA <sup>b</sup>	NA <sup>b</sup>	2.4	0.8	4.4	NA <sup>b</sup>	0.3	1.1	1.3	0.1	NA <sup>b</sup>	52.2	1.7	64.2
High	18.7	0.7	125.2	10.3	51.0	31.6	58.2	135.7	55.9	130.6	1,651.4	3,545.0	393.0	6,207.3

374 <sup>a</sup>low exposure scenario was not calculated because minimum concentrations of all BFRs but BDE-209 were not detected; median and high exposure375 scenarios assume transfer from a utensil containing the median and maximum values of  $c_{BFR-utensil}$  respectively;376 <sup>b</sup>not available due to a not detected concentration.

377



378 As shown in Table 4, daily exposure to total BFRs are ~60 ng and ~6,000 ng under  
379 median and high scenarios, respectively; while those for  $\Sigma$ BDEs are ~60 ng and 4,000  
380 ng respectively. To place these exposure estimates into context, Besis and Samara (2012)  
381 reviewed daily intake of PBDEs via different exposure pathways in different countries,  
382 and found that dust ingestion could amount to up to 400 ng/day intake in the US and  
383 the UK. Intake in other countries was lower, ranging from 50 to 200 ng/day. Dietary  
384 intake, as another important exposure pathway, ranged from 50 to 75 ng/day according  
385 to Besis's review. Harrad et al. (2004) investigated concentrations of tetra-hexa BDEs  
386 in UK duplicate diet samples and estimated dietary exposure of 90 ng/day for  $\Sigma$ PBDEs  
387 (tetra-to hexa-BDEs only). D'Silva et al. (2006) investigated concentrations of 17  
388 PBDEs in typical UK diet composite samples in 2003, and the daily dietary exposure  
389 for tri- to hepta-BDEs and BDE-209 were estimated to be 80 ng/day and 270 ng/day,  
390 respectively. For NBFRs, Tao et al. (2017) detected several NBFRs including EH-TBB,  
391 BEH-TEBP, BTBPE, DBPDE and tetrabromoethylcyclohexane (DBE-DBCH) in UK  
392 food samples, estimating the average total daily dietary exposure to the sum of these  
393 NBFRs for adults was 90 ng/day. This compares with the median and high-end  
394 estimates in this study of ~2 and ~2,000 ng/day. To place our exposure estimates into  
395 context against non-dietary exposure, Harrad et al. (2008) estimated indoor dust  
396 ingestion of PBDEs, DBDPE and BTBPE, and the median exposure for UK adult was  
397 about 200 ng/day. Ni et al. (2013) estimated PBDE exposure via indoor dust ingestion  
398 in different cities of China, the median exposure for adult ranged from 20 to 100 ng/day.

399 Compared with estimates of exposure via other pathways from by previous studies,  
400 exposure via cooking using BFR-containing utensils is not negligible. Moreover,  
401 although the transfer rate of BDE-209 during cooking is not high, it still accounts for  
402 the largest proportion (80 %) of exposure via cooking due to its high concentration in  
403 utensils.

404

405 It is important to emphasise the preliminary nature of our assessment of dietary  
406 exposure arising from using BFR-containing utensils. Our simulated cooking  
407 experiments involved deep frying, which is likely a worst-case scenario with respect to  
408 BFR extraction. Moreover, our estimate of oil-utensil contact occurring for 15 minutes  
409 over 1 week is subject to considerable uncertainty and will vary considerably between  
410 households, along with the frequency with which individuals will consume deep-fried  
411 food. Finally, we focused only on those utensils displaying elevated BFR concentrations,  
412 with our high-end exposure estimates based on the most contaminated utensil; thus our  
413 high-end estimates are likely a worst-case scenario, with our median estimates more  
414 representative of exposure at the population level. Balanced against this, it is not  
415 unreasonable to assume that utensils will have contained higher BFR concentrations  
416 when new and thus greater BFR transfer will have occurred earlier in the life of some  
417 of the older utensils studied here. On the whole therefore, we consider our estimates a  
418 reasonable first-level evaluation, and that they provide evidence to suggest that further  
419 investigation of the potential for human exposure arising from use of such utensils is

420 warranted.

### 421 **3.4.2 Dermal exposure**

422 Considering the high BFR concentration not only in the main body but also in the grip  
423 of kitchen utensils, exposure via dermal contact is of concern. Dermal uptake is a  
424 complex process involving two major steps. First, the transfer of BFRs from the plastic  
425 polymer to the skin surface film liquid (i.e. becomes bioaccessible). Second, the  
426 penetration of the skin barrier to reach the blood circulation (i.e. becomes bioavailable)  
427 (Abdallah et al., 2015). With the exception of HBCDDs (Pawar et al., 2017), an  
428 extensive survey of the literature revealed no available data on the dermal  
429 bioaccessibility of BFRs. For the second process, Abdallah et al. (2015) reported on the  
430 dermal uptake rates of mono to deca BDEs over a 24 h exposure period. Therefore, our  
431 exposure model adopts a conservative approach with the assumption of 100 %  
432 bioaccessibility of PBDEs (*in the absence of relevant data*), and data from Abdallah et  
433 al. (2015) were applied for estimation of bioavailability. Daily exposure (ng/day) via  
434 dermal contact was calculated by the equation below.

$$435 \quad E = C \times SA \times F \times EF \quad (4)$$

436 where  $E$  is daily dermal exposure (ng/day),  $C$  is the concentration of BFRs in the  
437 utensil (ng/cm<sup>2</sup>),  $SA$  is the skin surface area exposed (cm<sup>2</sup>),  $F$  is the fraction absorbed  
438 by the skin (unitless),  $EF$  is the fraction of time in contact with the item (day<sup>-1</sup>).

439

440 To transfer BFR concentration in ng/g to concentration per surface area, a 0.5 mm

441 depth ( $h$ ) plastic from the surface of the utensil was assumed. For utensil density

442 ( $\rho_{utensil}$ ) a value of 1.4 g/cm<sup>3</sup> was applied as indicated in section 3.4.1. So

443  $C (area) = h \times \rho_{utensil} \times C (mass) = 0.05 \text{ cm} \times 1.4 \text{ g/cm}^3 \times C (mass) = 0.07 C (mass)$ .

444

445 For the exposure area, we used data from the US EPA exposure factors handbook (U.S.

446 EPA, 2011) stating the average surface area of an adult hand is 1070 cm<sup>2</sup> for male and

447 890 cm<sup>2</sup> for a female. The average area of a single palm was estimated as  $1/2 \times 1/2 \times$

448  $(1070+890)/2 \text{ cm}^2 = 245 \text{ cm}^2$ . Considering that not the whole palm will contact with

449 kitchen utensils upon handling, a 75 % coefficient was assumed resulting in an exposed

450 skin area ( $SA$ ) of 184 cm<sup>2</sup>. Finally, parameters  $F$  and  $EF$  were obtained from Abdallah

451 et al. (2015), who measured various absorbed fraction of PBDEs at different exposure

452 times from 15 min to 24 h.

453

454 Over a daily contact time of 15 min, no dermal uptake was observed for any PBDEs

455 which is consistent with the “lag time” reported by Abdallah et al. (2015) for the studied

456 compounds. Lag time is defined as the time required by a specific chemical from its

457 initial contact with the skin surface to reach the systemic circulation. Low dermal

458 uptake was observed when the contact time was prolonged to 0.5 h and 1 h, except for

459 higher brominated BDEs (Table 5).

460

461

462 Table 5 PBDE exposure (ng/day) via dermal contact in median and high scenarios<sup>a</sup>

	BDE-28	BDE-47	BDE-100	BDE-99	BDE-154	BDE-153	BDE-183	BDE-209	ΣPBDEs
F (0.5 h) <sup>b</sup>	0.07%	0.04%	- <sup>c</sup>	-	-	-	-	-	
Median	NA <sup>d</sup>	0.05	-	-	-	-	-	-	0.05
High	1.19	5.41	-	-	-	-	-	-	6.60
F (1 h)	0.20%	0.13%	0.08%	0.08%	0.03%	0.03%	-	-	
Median	NA	0.17	0.03	0.17	0.02	0.04	-	-	0.43
High	3.40	17.58	1.18	5.51	3.85	456.43	-	-	487.95

463 <sup>a</sup>exposure in low scenario was not calculated because minimum concentrations of all  
 464 BFRs but BDE-209 were not detected; median and high exposure scenarios were  
 465 calculated based on median and maximum BFR concentration of P1~P30;

466 <sup>b</sup>data obtained from Abdallah et al. (2015);

467 <sup>c</sup>no transfer observed;

468 <sup>d</sup>not available due to a not detected concentration.

469

470 Our results indicate that human uptake of PBDEs via dermal contact with cooking  
 471 utensils is much lower than our intake estimates based on cooking and other pathways  
 472 (section 3.4.1). The exception to this is for BDE-153 in the 1 h contact high-end  
 473 scenario, due to the extremely high BDE-153 concentration in scissor sample P30. This  
 474 could be attributed to the limited daily contact time with utensils, and low penetration  
 475 efficiency into skin, especially for BDE-209 whose concentration was the highest.  
 476 Therefore, our findings suggest when using BFR-contaminated kitchen utensils,  
 477 exposure is dominated by utensil-oil transfer, rather than utensil-skin transfer.

#### 478 4. Conclusions

- 479 ● 34 % of plastic kitchen utensils analysed in this study contained measurable  
 480 concentrations of Br.

- 481 ● Under our extraction procedure, BDE-209 was predominant among our target  
482 BFRs in most utensils, but the pattern of other BFRs varied substantially between  
483 utensils. Elevated concentrations of BTBPE and BDE-153 were found in some  
484 utensils.
- 485 ● BFR transfer from utensils into hot oil during simulated cooking experiments was  
486 considerable, and differed between BFRs and utensils. Transfer efficiency  
487 decreased with increasing Br substitution of PBDEs.
- 488 ● Using BFR containing utensils for frying may lead to considerable dietary exposure,  
489 whilst exposure via dermal contact is negligible due to limited contact time and  
490 barrier effect of skin.

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