

## Supporting Information

2 **MANUSCRIPT TITLE:** Emissions and Occupational Exposure Risk of Halogenated  
3 Flame Retardants from Primitive Recycling of E-Waste

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## **11 NO. OF TABLES:**

12 NO. OF FIGURES: 1

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15      **Text S1. Experimental Setup and Sampling, Sample Extraction and Purification.**

16            Three plastic casings (A1–A3) and three printed circuit boards (B1–B3) were elected  
17          from 161 e-waste samples from e-waste dismantling workshops and a resource utilization  
18          company in Guangdong Province, China, for chamber experiments. These plastic casings  
19          and printed circuit boards were composition of televisions or computers, detailed product  
20          information can be obtained in our previous work.<sup>1</sup> Raw materials were cleaned and smashed  
21          into pieces (830–1700 µm) before extraction and simulation. Thermal treatment (at 300 °C)  
22          and open burning (~ 800–1350 °C) were conducted in a stainless steel chamber (500 mm  
23          length × 500 mm width × 1500 mm height). The inner walls of the apparatus were buffed to  
24          reduce adsorption, rinsed with acetone after each experiment, and these rinsing samples were  
25          collected. The air flowed through the inlet purifying unit into the chamber, and finally to a  
26          Micro-Orifice Uniform Deposit Impactor (MOUDI; MSP Corporation, Shoreview, MN),  
27          which collected particulate samples in 11 size fractions (i.e., >18, 10–18, 5.6–10, 3.2–5.6,  
28          1.8–3.2, 1.0–1.8, 0.56–1.0, 0.32–0.56, 0.18–0.32, 0.10–0.18, and 0.056–0.10 µm), followed  
29          by a polyurethane foam plug (PUF; 6.5 cm diameter × 8.0 cm; density: 0.03 g cm<sup>-3</sup>) for  
30          collection of gaseous samples at the chamber outlet, at a constant flow rate of 30 L min<sup>-1</sup>.  
31          The sampling duration were in cohesion with the progression of the experiments and  
32          continued for another 180 min after the experiments were completed. Schematic diagram  
33          showing the simulation experiment setup for measuring emissions of FRs from e-waste  
34          processing can be obtained in our previous work.<sup>1</sup>

35            Raw e-waste samples were extracted by 20 mL of toluene for 30 min with an  
36          ultrasonic extractor three times, while a mixture of *n*-hexane, dichloromethane, and acetone  
37          (2:2:1 in volume) was applied in the ultrasonic extraction of airborne particle and residual ash  
38          samples. Soxhlet extraction by the above-mentioned solvent mixture lasting for 48 h were  
39          applied for the extraction of PUF plugs. Each extract was concentrated to approximately 1  
40          mL in *n*-hexane by a pressure blowing concentrator and then purified on a silica gel column

41 (i.d.=1.0 cm) consisted of neutral silica gel (12 cm) and anhydrous sodium sulfate (1 cm)  
42 from bottom to top. The column was eluted with 45 mL of *n*-hexane, and 50 mL of  
43 dichloromethane in sequence.

44 The first fraction containing AHFRs was then concentrated to 100  $\mu$ L and purified by  
45 another glass column (i.d.=0.23 cm) packed with 5 cm sulfuric acid silica (44% sulfuric acid,  
46 w/w) and 1 cm anhydrous sodium sulfate with an eluate of 1.5 mL *n*-hexane. The second  
47 fraction containing BPhs was then concentrated to 100  $\mu$ L in hexane and spiked with internal  
48 standards (fluorene- $d_{10}$ , pyrene- $d_{10}$ ) before instrumental analysis. And then this fraction  
49 containing TBBPA and HBCDs was evaporated to near-dryness under a gentle N<sub>2</sub> stream and  
50 re-dissolved in 100  $\mu$ L methanol. Finally, internal standards ( $d_{10}$ -labeled TBBPA,  $d_{18}$ -labeled  
51  $\alpha$ -,  $\beta$ -, and  $\gamma$ -HBCD) were added before liquid chromatography/mass spectrometry analysis.  
52

### 53 **Text S2. Instrumental Analysis**

54 Concentrations of AHFRs were determined with a Shimadzu Model 2010 Plus gas  
55 chromatograph coupled with a Model QP2010 Ultra mass spectrometer in the negative  
56 chemical ionization mode. A DB-5MS capillary column (30 m  $\times$  0.25 mm i.d. with 0.25  $\mu$ m  
57 film thickness) was used for chromatographic separation for all but DBDPE which was  
58 separated on a DB-5HT capillary column (15 m  $\times$  0.25 mm i.d. with 0.1  $\mu$ m film thickness).  
59 Concentrations of BPhs were quantified with an Agilent 7890B gas chromatograph coupled  
60 to a 5977 mass spectrometer in the electron impact ionization mode, coupled with a HP-5MS  
61 capillary column (30 m  $\times$  0.25 mm i.d. with 0.25  $\mu$ m film thickness).

62 The analysis of TBBPA and HBCDs were conducted by a Shimadzu LC-30AD liquid  
63 chromatography (LC) system coupled to an AB SCIEX Triple QUAD 5500 electrospray  
64 triple quadrupole mass spectrometer. An Agilent Eclipse Plus C18 column (100 mm  $\times$  4.6  
65 mm i.d., 3.5  $\mu$ m) was applied to separate TBBPA and the HBCD diastereoisomers. The flow  
66 rate was set at 0.3 mL min<sup>-1</sup> with an injection volume of 2  $\mu$ L. The mass spectrometer was

67 operated in the electrospray ionization (ESI) negative ion mode. The ions were selected at  
68 the ([M-H] $^-$ ) transition of m/z 640.6/79, 652.6/79, and 657.6/79 for HBCD, 13C12-labeled  
69 HBCD, and d18-labeled HBCD isomers, respectively. The ions m/z 542.8/79 for TBBPA,  
70 m/z 554.8/79 for 13C12-labeled TBBPA, and m/z 551.8/79 for d10-labeled TBBPA were  
71 monitored.

72 A DB-5MS capillary column (30 m  $\times$  0.25 mm i.d. with 0.25  $\mu\text{m}$  film thickness) was  
73 used for chromatographic separation for TBECH, HCDBCO, HBB, TBB, TDBPP, BTBPE,  
74 TBPH, Dec-602, Dec-603, Dec-604, *syn*-DP, and *anti*-DP, with temperature program  
75 increasing from 110 °C (held for 1 min) to 200 °C (held for 1 min) at 12 °C min $^{-1}$ , then  
76 increasing at 5 °C min $^{-1}$  to 240 °C (held for 5 min), increasing at 2 °C min $^{-1}$  to 280 °C (held  
77 for 5 min), and finally increasing at 30 °C min $^{-1}$  to 310 °C (held for 10 min).

78 For DBDPE, a DB-5HT capillary column (15 m  $\times$  0.25 mm i.d. with 0.1  $\mu\text{m}$  film  
79 thickness) was used, with temperature program increasing from 100 °C (held for 2 min) to  
80 255 °C at 25 °C min $^{-1}$ , then increasing at 1 °C min $^{-1}$  to 265 °C, and finally increasing at 25  
81 °C min $^{-1}$  to 325 °C (held for 6 min). Carrier gas was ultrahigh purity helium at a flow rate of  
82 1.5 mL min $^{-1}$ . The ion source and quadrupole temperatures were set at 200 and 150 °C,  
83 respectively.

84 The concentrations of 4-BPh, 2,4-DBPh, 2,6-DBPh, and 2,4,6-TBPh were quantified  
85 with an Agilent 7890B gas chromatograph coupled to a 5977 mass spectrometer in the  
86 electron impact ionization mode. A HP-5MS capillary column (30 m  $\times$  0.25 mm i.d. with  
87 0.25  $\mu\text{m}$  film thickness) was applied for their separation with temperature program increasing  
88 from 60 °C (held for 1 min) at 30 °C min $^{-1}$  to 200 °C, then increasing at 15 °C min $^{-1}$  to 278  
89 °C, and finally increasing at 25 °C min $^{-1}$  to 300 (held for 14 min).

90 An Agilent Eclipse Plus C18 column (100 mm  $\times$  4.6 mm i.d., 3.5  $\mu\text{m}$ , Agilent, CA)  
91 was used to separate the TBBPA and HBCD diastereoisomers. The mobile phases A (30:70  
92 methanol/water) and B (30:70 methanol/acetonitrile) were used, starting at 20:80 A/B (held

93 for 2 min) and increasing to 0:100 A/B over 4 min (held for 2 min) followed by a return to  
94 20:80 A/B over 2 min (held for 10 min).

95

96 **Text S3. The Estimation of the Concentration of Particulate and Gaseous FRs in**  
97 **Workshop Air based on the EFs**

98 The estimation was based on information of informal e-waste dismantling workshops  
99 in Guiyu. According to the data, there are more than 5,500 household workshops in Guiyu  
100 Town, which are engaged in the dismantling and processing of e-waste, with annual  
101 dismantling e-waste amount reached 1.55 million tons.<sup>2,3</sup> Thus the recycling amount for a  
102 workshop per day ( $W_j$ , g d<sup>-1</sup>) is estimated by:

103 
$$W_j = W_{\text{annual}} \times f_j \div 5500 \div 365 \quad (\text{S1})$$

104 where  $f_j$  is the fraction of plastics or printed circuit boards to the total dismantling  
105 amount of e-waste in Guiyu, assumed to be consistent with those in formal recycling  
106 company (29.55% and 4.73%, respectively).<sup>4</sup>

107 As estimated for the dismantling procedure of waste printed circuit boards in facilities  
108 by Guo et al.,<sup>5</sup> after 20–30 s of heating time, the solder on the waste printed circuit boards  
109 melted, and the worker started to manually remove the electrical components, such as knock  
110 and pull lasting about 20 s. The thermal process was assumed to similar to this process in  
111 informal workshops and concentration of FRs in the workshop was assumed to be uniform  
112 and steady during this cycle. Thus the concentration of FRs accumulated in air after one-day  
113 dismantling was estimated by:

114 
$$C_{j,i} = EF_{j,I} \times t \times W_j \div V \quad (\text{S2})$$

115 where  $EF_{j,I}$  is the emission factors (ng g<sup>-1</sup> s<sup>-1</sup>) for size-fractioned particulate or  
116 gaseous FRs in the thermal treatment of plastic casings or printed circuit boards;  $t$  is the  
117 heating lasting time (30 s);  $V$  is the volume of a workshop which was normally around 100

118  $\text{m}^2$  with an average height of 3.5 m, thus with a volume at 350  $\text{m}^3$ . The FR concentrations in  
119 air were supposed to accumulate in a liner increase, thus the average value per day was  
120 applied in the health assessment.

121

122 **Text S4. Exposure and Health Assessment of Flame retardants**

123 To estimate the deposition efficiency and flux of inhaled HFRs in the human  
124 respiratory tract, we adopted the simplified equations based on the International Commission  
125 on Radiological Protection model (ICRP).<sup>6,7</sup> The model calculates the deposition fractions of  
126 inhaled particles in three main regions of the human respiratory tract, i.e., head airways (HA,  
127 including nose, mouse, pharynx, and larynx), tracheobronchial region (TB), and alveolar  
128 region (AR).

129 The particle deposition efficiency in the head airway ( $\text{DF}_{\text{HA}}$ ) was estimated by:

130 
$$\text{DF}_{\text{HA},i} = \text{IF}_i \times \left( \frac{1}{1 + \exp(6.84 + 1.183 \ln D_{p,i})} + \frac{1}{1 + \exp(0.924 - 1.885 \ln D_{p,i})} \right) \quad (\text{S3})$$

131 where  $D_{p,i}$  ( $\mu\text{m}$ ) is geometric mean diameter of the particle at stage i, and this value for the  
132 first stage ( $D_p > 18 \mu\text{m}$ ) was assumed to be 20  $\mu\text{m}$ ; and IF was inhalable fraction of all  
133 particles, estimated by:

134 
$$\text{IF} = 1 - 0.5 \left( 1 - \frac{1}{1 + 0.00076 D_{p,i}^{2.8}} \right) \quad (\text{S4})$$

135 The particle deposition efficiency in the tracheobronchial region ( $\text{DF}_{\text{TB}}$ ) was  
136 calculated by:

137 
$$\text{DF}_{\text{TB},i} = \frac{0.00352}{D_{p,i}} \times (\exp(-0.234(\ln D_{p,i} + 3.40)^2) + 63.9 \exp(-0.819(\ln D_{p,i} - 1.61)^2)) \quad (\text{S5})$$

138 The particle deposition efficiency in the alveolar region ( $\text{DF}_{\text{AR}}$ ) was estimated by:

139 
$$DF_{AR,i} = \frac{0.0155}{D_{p,i}} \times (\exp(-0.416(\ln D_{p,i} + 2.84)^2) + 19.11\exp(-0.482(\ln D_{p,i} - 1.362)^2))$$

140 (S6)

141 The total deposition efficiency ( $DF_{Total}$ ) of particle in the human respiratory tract was  
142 the sum of the three region deposition efficiencies.

143 The deposition flux ( $D$ , pg h<sup>-1</sup>) of particle-bound HFRs in the respiratory tract was  
144 estimated by:

145 
$$D_j = \sum (DF_{j,i} \times C_i) \times V \quad (S7)$$

146 where  $DF_{j,i}$  is the particle deposition efficiency in each region for  $D_{p,i}$ ;  $C_i$  is FR  
147 concentration in particle  $D_{p,i}$  (pg m<sup>-3</sup>); and  $V$  is the breathing rate (0.54 m<sup>3</sup> h<sup>-1</sup> at the sitting  
148 level of exercise).

149 Inhalation intake flux of gaseous ( $D_g$ ) was estimated by the following:

150 
$$D_g = \sum C_i \times V \quad (S8)$$

151 Dermal intake of particulate and gaseous FRs was assessed using the formula  
152 proposed by Weschler and Nazaroff as followed:<sup>8</sup>

153 
$$D_{erm,p} = \sum C_i \times k_{p,d} \times BSA \quad (S9)$$

154 
$$D_{erm,g} = C_g \times k_{p,g} \times BSA \quad (S10)$$

155 where BSA is the body surface area, and 3300 cm<sup>2</sup> for adults<sup>9</sup> was adopt in this study;  $k_{p,d}$   
156 and  $k_{p,g}$  are the transdermal permeability coefficients (m h<sup>-1</sup>) of target FRs in the particulate  
157 and gaseous phases, and 0.13 and 0.34 m h<sup>-1</sup> were adopt from literature,<sup>10</sup> respectively.

158 The total exposure (EI, ng kg<sup>-1</sup> day<sup>-1</sup>) of FRs for the worker was estimated by:

159 
$$EI = \frac{\sum D_i \times T}{BW} \quad (S11)$$

160 where T is the daily exposure time (h d<sup>-1</sup>) which was assumed to be 8 h; BW is the body  
161 weight of an adult worker assumed to be 60 kg at an average level.

162 Monte Carlo simulation was used to evaluate the uncertainty and variability of intake  
163 amounts of FRs via dermal and inhalation pathways. The concentration of individual FR was  
164 assumed to be lognormal distributed with average value and standard deviation obtained in  
165 our studies.

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167

168 **Table S1. The Concentrations of Halogenated Flame Retardants in Plastic Casings (A1–A3) and Printed Circuit Boards (B1–B3).**

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	A1		A2		A3		B1		B2		B3	
	Mean	SD										
$\alpha$ -HBCD	$1.1 \times 10^4$	680	$6.2 \times 10^3$	280	$3.4 \times 10^3$	310	120	13	170	9.6	430	52
$\beta$ -HBCD	$3.4 \times 10^3$	160	$1.8 \times 10^3$	99	240	47	<RL <sup>a</sup>	<RL	<RL	<RL	88	10
$\gamma$ -HBCD	$1.3 \times 10^3$	89	660	85	560	34	14	2.0	21	4.2	280	38
TBBPA	$4.2 \times 10^5$	$1.3 \times 10^4$	$2.0 \times 10^5$	$9.6 \times 10^3$	$1.3 \times 10^5$	$1.2 \times 10^4$	$2.7 \times 10^4$	$4.5 \times 10^3$	$1.7 \times 10^5$	$4.4 \times 10^4$	$1.0 \times 10^5$	$4.7 \times 10^3$
TBECH	850	38	320	35	<RL							
HBB	350	23	190	36	150	31	33	24	<RL	<RL	30	9.2
DBDPE	$2.8 \times 10^4$	$4.7 \times 10^3$	$5.6 \times 10^4$	$5.7 \times 10^4$	$2.0 \times 10^4$	$3.8 \times 10^3$	<RL	<RL	<RL	<RL	$7.2 \times 10^3$	510
Dec-602	200	7.0	40	6.8	36	5.1	<RL	<RL	<RL	<RL	<RL	<RL
Dec-604	280	90	93	24	130	20	<RL	<RL	<RL	<RL	<RL	<RL
<i>syn</i> -DP	$1.1 \times 10^4$	920	$3.8 \times 10^3$	660	320	89	<RL	<RL	<RL	<RL	<RL	<RL
<i>anti</i> -DP	$6.3 \times 10^4$	$4.5 \times 10^3$	$2.3 \times 10^4$	$4.6 \times 10^3$	$1.6 \times 10^3$	450	<RL	<RL	<RL	<RL	<RL	<RL

183	4-BPh	650	42	<RL	<RL	<RL	<RL	180	55	140	35	850	100
184	2,4-DBPh	640	83	220	42	240	47	51	10	140	71	10	15
185	2,6-DBPh	$1.1 \times 10^3$	15	600	110	550	84	110	92	850	190	210	100
186	2,4,6-TBPh	$2.4 \times 10^4$	$5.0 \times 10^3$	$1.2 \times 10^4$	160	$1.6 \times 10^4$	$1.8 \times 10^3$	$9.8 \times 10^3$	$1.8 \times 10^3$	$7.8 \times 10^3$	$1.5 \times 10^3$	$4.0 \times 10^3$	960
187	$\Sigma_{15}$ HFR	$5.7E \times 10^5$	$1.3 \times 10^4$	$3.0 \times 10^5$	$4.6 \times 10^4$	$1.3 \times 10^5$	$5.2 \times 10^4$	$7.1 \times 10^4$	$5.0 \times 10^4$	$1.8 \times 10^5$	$4.4 \times 10^4$	$1.2 \times 10^5$	$4.2 \times 10^3$

188 <sup>a</sup> The concentration is below the reporting limits.

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190 **Table S2. Emission Factors (EFs) of 15 Kinds of Halogenated Flame Retardants (ng g<sup>-1</sup>) from the Thermal Treatment (T) and Open  
191 Burning (B) of Plastic Casings (A1–A3) and Printed Circuit Boards (B1–B3).**

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	<u>A1</u>		<u>A2</u>		<u>A3</u>		<u>B1</u>		<u>B2</u>		<u>B3</u>	
	T	B	T	B	T	B	T	B	T	B	T	B
α-HBCD	$3.6 \times 10^3$	$3.7 \times 10^3$	155	85	$5.0 \times 10^3$	198	240	100	210	200	270	240
β-HBCD	280	310	100	150	540	6.3	93	45	190	100	170	67
γ-HBCD	$3.2 \times 10^3$	$1.7 \times 10^3$	25	11	$5.0 \times 10^3$	250	580	27	190	55	220	77
TBBPA	$1.3 \times 10^5$	$6.1 \times 10^4$	$1.7 \times 10^4$	446	$3.6 \times 10^4$	140	$5.4 \times 10^4$	$1.1 \times 10^4$	$3.8 \times 10^5$	$2.8 \times 10^4$	$3.2 \times 10^5$	$1.6 \times 10^4$
TBECH	21	<RL <sup>a</sup>	3.9	<RL	10	<RL	2.4	<RL	0.69	<RL	1.5	<RL
HBB	230	120	92	3.0	8.6	27	6.5	16	13	15	1.2	290
DBDPE	$4.4 \times 10^3$	$1.2 \times 10^4$	$2.7 \times 10^3$	$1.1 \times 10^3$	$9.7 \times 10^3$	780	<RL	<RL	<RL	<RL	$6.0 \times 10^3$	$1.7 \times 10^3$
Dec-602	90	65	6.6	<RL	2.9	<RL	<RL	<RL	<RL	<RL	<RL	<RL
Dec-604	99	<RL	2.0	<RL	50	<RL	<RL	<RL	<RL	<RL	<RL	<RL
syn-DP	$2.8 \times 10^3$	$1.2 \times 10^3$	470	<RL	280	<RL	<RL	<RL	<RL	<RL	<RL	<RL

205	<i>anti</i> -DP	$9.6 \times 10^3$	$7.5 \times 10^3$	$2.1 \times 10^3$	4.2	97	<RL						
206	4-BPh	990	$1.6 \times 10^3$	480	930	390	$1.1 \times 10^3$	500	$1.4 \times 10^3$	940	$2.8 \times 10^3$	$1.3 \times 10^3$	$3.1 \times 10^3$
207	2,4-DBPh	$1.1 \times 10^3$	$1.9 \times 10^3$	240	$1.6 \times 10^3$	$1.0 \times 10^3$	$1.6 \times 10^3$	$1.9 \times 10^3$	$3.3 \times 10^3$	$1.4 \times 10^3$	$2.2 \times 10^3$	$1.8 \times 10^3$	$3.2 \times 10^3$
208	2,6-DBPh	$1.7 \times 10^3$	$3.1 \times 10^3$	470	$2.4 \times 10^3$	990	$2.9 \times 10^3$	$2.7 \times 10^3$	$4.2 \times 10^3$	$3.2 \times 10^3$	$4.5 \times 10^3$	$4.0 \times 10^3$	$5.8 \times 10^3$
209	2,4,6-TBPh	$6.5 \times 10^3$	$6.5 \times 10^3$	$2.0 \times 10^3$	$2.1 \times 10^3$	$8.2 \times 10^3$	$3.1 \times 10^3$	$9.2 \times 10^3$	$4.7 \times 10^3$	$4.7 \times 10^3$	$1.9 \times 10^3$	$4.6 \times 10^3$	$2.5 \times 10^3$
210	$\sum_{15}$ HFR	$1.6 \times 10^5$	$1.0 \times 10^5$	$2.6 \times 10^4$	$8.8 \times 10^3$	$6.7 \times 10^4$	$1.0 \times 10^4$	$7.0 \times 10^4$	$2.5 \times 10^4$	$3.9 \times 10^5$	$4.0 \times 10^4$	$3.4 \times 10^5$	$3.3 \times 10^4$

211 <sup>a</sup> The concentration of HFRs in after-treatment products were below reporting limits.

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214 **Table S3. Output over Input Mass Ratios by Thermal Process (T) and Open Burning (B) of Plastic Casings (A1–A3) and Printed  
215 Circuit Boards (B1–B3).**

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	<u>A1</u>		<u>A2</u>		<u>A3</u>		<u>B1</u>		<u>B2</u>		<u>B3</u>	
	T	B	T	B	T	B	T	B	T	B	T	B
$\alpha$ -HBCD	0.33	0.34	0.03	0.01	1.32	1.09	2.06	0.89	1.22	1.15	0.64	0.57
$\beta$ -HBCD	0.08	0.09	0.06	0.08	1.78	0.26	— <sup>a</sup>	—	—	—	1.92	0.77
$\gamma$ -HBCD	2.52	1.32	0.04	0.02	8.15	6.76	42.0	1.92	9.01	2.68	0.78	0.27
TBBPA	0.30	0.15	0.09	$2.3 \times 10^{-3}$	0.25	0.01	2.04	0.42	2.28	0.17	3.10	0.16
TBECH	0.02	< RL <sup>b</sup>	0.01	< RL	—	—	—	—	—	—	—	—
HBB	0.65	0.34	0.47	0.02	0.05	0.18	0.19	0.49	—	—	0.04	9.75
DBDPE	0.16	0.43	0.05	0.02	0.48	0.04	—	—	—	—	0.84	0.24
Dec-602	0.44	0.32	0.17	< RL	0.08	< RL	—	—	—	—	—	—
Dec-604	0.35	< RL	0.02	< RL	0.37	< RL	—	—	—	—	—	—
syn-DP	0.25	0.11	0.12	< RL	0.87	< RL	—	—	—	—	—	—

229	<i>anti</i> -DP	0.15	0.12	0.09	$1.9 \times 10^{-4}$	0.06	< RL	–	–	–	–	–	–
230	4-BPh	1.52	2.50	–	–	–	–	2.76	7.78	6.93	20.9	1.56	3.71
231	2,4-DBPh	1.71	2.94	1.12	7.48	4.33	6.96	37	65	9.79	15.5	174	312
232	2,6-DBPh	1.53	2.71	0.79	3.96	1.80	5.32	24	38	3.74	5.30	19	27
233	2,4,6-TBPh	0.28	0.27	0.16	0.17	0.51	0.19	0.94	0.48	0.59	0.24	1.15	0.63
234	$\sum_{15}$ HFR	0.29	0.18	0.09	0.03	0.36	0.21	1.89	0.69	2.22	0.23	2.91	0.29

235 <sup>a</sup> Invalid. The concentrations are below the reporting limits in raw materials and the output over input mass ratio therefore could not be  
 236 calculated;

237 <sup>b</sup> The concentration of HFRs in after-treatment products were below reporting limits.

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239 **Table S4. Relative abundance of individual halogenated flame retardant derived from thermal treatment (T) and open burning (B) of**  
 240 **plastic casings (A1–A3) and printed circuit boards (B1–B3).**

241

	<u>A1</u>		<u>A2</u>		<u>A3</u>		<u>B1</u>		<u>B2</u>		<u>B3</u>		
	T	B	T	B	T	B	T	B	T	B	T	B	
243													
244	$\alpha$ -HBCD	2.2	3.7	0.59	0.96	7.55	1.9	0.34	0.41	0.05	0.50	0.08	0.73
245	$\beta$ -HBCD	0.17	0.30	0.40	1.72	0.81	0.06	0.13	0.18	0.05	0.25	0.05	0.20
246	$\gamma$ -HBCD	2.0	1.7	0.10	0.13	7.4	2.5	0.84	0.11	0.05	0.14	0.06	0.23
247	TBBPA	79	61	67	5.1	53	1.4	78	45	97	70	95	49
248	TBECH	0.01	— <sup>a</sup>	0.01	—	0.02	—	$3.5 \times 10^{-3}$	—	$1.8 \times 10^{-4}$	—	$4.3 \times 10^{-4}$	—
249	HBB	0.14	0.12	0.35	0.03	0.01	0.27	0.01	0.07	$3.3 \times 10^{-3}$	0.04	$3.6 \times 10^{-4}$	0.87
250	DBDPE	2.7	12	10	12	15	7.7	—	—	—	—	1.8	5.1
251	Dec-602	0.06	0.06	0.03	—	$4.4 \times 10^{-3}$	—	—	—	—	—	—	—
252	Dec-604	0.06	—	0.01	—	0.08	—	—	—	—	—	—	—
253	<i>syn</i> -DP	1.7	1.2	1.8	—	0.42	—	—	—	—	—	—	—

254	<i>anti</i> -DP	5.9	7.4	7.8	0.05	0.14	—	—	—	—	—	—	—
255	4-BPh	0.61	1.6	1.8	10	0.58	11	0.72	5.6	0.24	7.1	0.39	9.4
256	2,4-DBPh	0.67	1.9	0.92	18	1.5	16	2.8	13	0.36	5.6	0.52	9.6
257	2,6-DBPh	1.1	3.0	1.8	27	1.5	29	3.9	17	0.81	11	1.2	17
258	2,4-TBPh	4.0	6.4	7.5	24	12	31	13	19	1.2	4.8	1.4	7.6

259 <sup>a</sup> The concentration of HFRs in after-treatment products were below reporting limits.

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260

261 **Table S5. Geometric Mean Diameter (GMD,  $\mu\text{m}$ )<sup>a</sup> and Geometric Standard Deviations (GSD,  $\mu\text{m}$ )<sup>b</sup> of Particle-Bound HFRs Calculated**  
 262 **for Particles Released by Thermal Process of Plastic Casings (A1–A3) and Printed Circuit Boards (B1–B3).**

263

	<u>A1</u>		<u>A2</u>		<u>A3</u>		<u>B1</u>		<u>B2</u>		<u>B3</u>	
	GMD	GSD	GMD	GSD	GMD	GSD	GMD	GSD	GMD	GSD	GMD	GSD
$\alpha$ -HBCD	0.85	1.70	0.78	2.33	0.73	1.70	0.37	1.65	0.45	1.65	0.52	1.60
$\beta$ -HBCD	1.00	1.76	0.66	1.98	1.11	1.66	0.42	1.63	0.48	1.64	0.53	1.67
$\gamma$ -HBCD	0.86	1.72	0.83	2.00	0.82	1.69	0.44	1.70	0.49	1.62	0.60	1.52
TBBPA	1.12	2.02	0.75	2.07	0.78	1.63	0.48	1.96	0.48	1.65	0.56	1.64
TBECH	— <sup>c</sup>	—	1.34	1.00	—	—	—	—	—	—	—	—
HBB	0.83	2.09	0.64	1.89	—	—	—	—	0.57	1.33	—	—
DBDPE	0.88	2.95	0.67	2.48	0.73	2.45	—	—	—	—	0.48	2.14
Dec-602	0.85	1.98	0.58	2.19	1.34	1.00	—	—	—	—	—	—
Dec-604	0.75	1.74	1.34	1.00	0.75	1.00	—	—	—	—	—	—
<i>syn</i> -DP	0.82	1.87	0.65	1.82	0.76	1.59	—	—	—	—	—	—

276	<i>anti</i> -DP	0.75	1.90	0.65	1.78	1.04	1.34	—	—	—	—	—	—
277	4-BPh	1.00	2.49	0.72	3.36	—	—	—	—	—	—	—	—
278	2,4-DBPh	0.78	1.78	0.55	1.69	0.77	1.62	0.50	1.56	0.41	1.63	0.63	1.42
279	2,6-DBPh	0.78	1.42	0.72	1.59	0.73	1.41	0.46	1.89	0.47	1.81	0.56	1.69
280	2,4,6-TBPh	0.81	2.37	0.66	2.11	0.72	1.94	0.57	2.42	0.49	2.15	0.64	1.87
281	$\sum_{15}$ HFR	1.06	2.04	0.73	2.10	0.77	1.78	0.48	2.01	0.48	1.65	0.56	1.65

282 <sup>a</sup> Calculated as  $\log \text{GMD} = \frac{\sum (C_i \times \log D_{p,i})}{\sum C_i}$ ; where  $C_i$  is the concentration of target compound in size class i and  $D_{p,i}$  is the geometric mean  
283 particle diameter collected on stage i;

$$284 \quad \log \text{GSD} = \sqrt{\frac{\sum [C_i \times (\log D_{p,i} - \log \text{GMD})^2]}{\sum C_i}};$$

285 <sup>c</sup> Invalid for calculation.

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286

287 **Table S6. Geometric Mean Diameter (GMD,  $\mu\text{m}$ )<sup>a</sup> and Geometric Standard Deviations (GSD,  $\mu\text{m}$ )<sup>b</sup> of Particle-Bound Halogenated  
288 Flame Retardants Calculated for Particles Released by Open Burning of Plastic Casings (A1–A3) and Printed Circuit Boards (B1–B3).**

289

	A1		A2		A3		B1		B2		B3	
	GMD	GSD	GMD	GSD	GMD	GSD	GMD	GSD	GMD	GSD	GMD	GSD
291 $\alpha$ -HBCD	1.02	1.95	0.49	1.78	0.46	1.71	0.53	1.76	0.69	1.92	0.63	1.83
292 $\beta$ -HBCD	— <sup>c</sup>	—	0.58	2.20	—	—	0.75	1.00	0.57	1.92	0.62	1.78
293 $\gamma$ -HBCD	0.95	2.15	0.47	1.80	0.77	1.92	0.78	1.16	0.55	1.76	0.73	1.83
294 TBBPA	1.08	1.71	0.62	2.12	0.72	2.26	0.61	1.81	0.58	2.11	0.62	1.82
295 TBECH	—	—	—	—	—	—	—	—	—	—	—	—
296 HBB	1.57	2.32	0.42	1.00	0.63	2.76	0.35	2.11	0.57	2.02	0.64	1.88
297 DBDPE	0.95	1.33	0.45	1.39	2.40	1.00	—	—	—	—	0.61	1.93
298 Dec-602	—	—	—	—	—	—	—	—	—	—	—	—
299 Dec-604	—	—	—	—	—	—	—	—	—	—	—	—
300 <i>syn</i> -DP	—	—	—	—	—	—	—	—	—	—	—	—

302	<i>anti</i> -DP	1.10	1.32	0.42	1.00	–	–	–	–	–	–	–	–
303	4-BPh	1.10	2.60	0.55	1.81	0.40	1.80	0.47	1.63	0.47	1.72	0.60	1.79
304	2,4-DBPh	1.24	2.18	–	–	0.33	1.32	0.47	1.64	0.43	1.60	0.48	1.52
305	2,6-DBPh	–	–	–	–	–	–	0.75	1.00	0.41	1.61	0.55	1.63
306	2,4,6-TBPh	0.87	3.04	0.74	3.35	0.38	2.26	0.50	2.01	0.50	1.67	0.56	1.72
307	$\sum_{15}$ HFR	0.99	2.57	0.50	1.83	0.47	2.41	0.61	1.82	0.57	2.10	0.62	1.82

308 <sup>a</sup> Calculated as  $\log \text{GMD} = \frac{\sum (C_i \times \log D_{p,i})}{\sum C_i}$ ; where  $C_i$  is the concentration of target compound in size class i and  $D_{p,i}$  is the geometric mean  
309 particle diameter collected on stage i;

$$310 \quad \log \text{GSD} = \sqrt{\frac{\sum [C_i \times (\log D_{p,i} - \log \text{GMD})^2]}{\sum C_i}};$$

311 <sup>c</sup> Invalid for calculation.

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312 **Table S7. Daily Intake (ng d<sup>-1</sup>) of Particle-bound and Gaseous Flame Retardants in the Thermal Treatment of E-waste through**  
 313 **Inhalation and Dermal Absorption, and the Estimated Daily Intake Dose (EDI, ng kg<sup>-1</sup> d<sup>-1</sup>) and Their Reference Dose (RfD, ng kg<sup>-1</sup>**  
 314 **d<sup>-1</sup>).**

315

		Inhalation			Dermal			Total	EDI	RfD
		Particle	Gas	Total	Particle	Gas	Total			
HBCDs	2.1E4±4.4E3	0±0	2.1E4±4.4E3	4.5E3±1.6E3	0±0	4.5E3±1.6E3	2.6E4±4.7E3	430±78	5000 <sup>a</sup>	
TBBPA	3.4E5±7.0E4	6.04±1.95	3.4E5±6.9E4	7.4E4±2.7E4	1.26±0.39	7.4E4±2.7E4	4.1E5±7.5E4	6.9E3±1240	10 <sup>6</sup> <sup>a</sup>	
TBECH	4.3±2.91	0±0	4.3±2.93	0.59±0.4	0±0	0.6±0.42	4.9±2.98	0.08±0.04	— <sup>b</sup>	
HBB	350±85	0±0	350±85	75±32	0±0	75±32	430±90	7.1±1.5	—	
DBDPE	1.2E4±2.7E3	0±0	1.2E4±2.7E3	2.5E3±1.1E3	0±0	2.5E3±1.1E3	1.5E4±2.9E3	240±48	330000 <sup>c</sup>	
Dec-602	130±360	0±0	130±360	25±15	0±0	25±15	150±320	2.5±5.3	—	
Dec-604	170±41	0±0	170±41	39±14	0±0	39±14	210±45	3.5±0.74	—	
DP	1.4E4±4.3E3	0±0	1.4E4±4.3E3	3.1E3±1.7E3	0±0	3.1E3±1.7E3	1.7E4±4.5E3	280±76	1.02E7 <sup>d</sup>	
4-BPh	530±143.61	4.5E3±990	5.1E3±1.0E3	120±49	950±210	1.1E3±210	6.1E3±1.2E3	10±20	—	
2,4-BPh	790±200	8.6E3±1.6E3	9.4E3±1.6E3	170±65	1.8E3±330	2.0E53±330	1.1E4±1.9E3	190±31	—	

328	2,6-DBPh	690±230	1.3E4±1.9E3	1.4E4±1.9E3	170±68	2.8E3±410	3.0E3±420	1.7E4±2.4E3	280±40	–
329	2,4,6-TBPh	1.2E4±2.2E3	1.7E4±2.8E3	2.9E4±3.6E3	2.8E3±920	3.4E3±590	6.3E3±1.1E3	3.5E4±4.2E3	590±69	–
330	ΣHFR	4.0E5±7.7E4	4.3E4±6.7E3	4.5E5±7.7E4	8.7E4±2.9E4	8.9E3±1.4E3	9.7E4±2.9E4	5.4E5±8.2E4	9.0E3±1.4E3	
331	BDE-7	230±39	11±7.3	240±41	56±18	2.27±1.54	58±18	300±44	5.0±0.73	–
332	BDE-15	164±19	79±27	240±33	40±7.1	16±5.7	56±8.9	300±38	5.0±0.64	–
333	BDE-17	57±8.5	0.26±0.18	57±8.5	14±2.9	0.05±0.03	14±3.0	71±9.0	1.2±0.14	100 <sup>e</sup>
334	BDE-28	63±12	2.8±1.9	66±12	21±6.2	0.58±0.39	21±6.3	87±14	1.4±0.22	100 <sup>e</sup>
335	BDE-47	283±68	2.8±1.9	280±66	88±39	0.57±0.39	89±38	370±77	6.3±1.3	100 <sup>e</sup>
336	BDE-49	410±74	0±0	410±73	89±24	0±0	89±23	500±77	8.3±1.3	100 <sup>e</sup>
337	BDE-66	268±87	0.53±0.36	270±86	72±25	0.11±0.07	72±25	340±90	5.7±1.5	100 <sup>e</sup>
338	BDE-71	430±85	17±1.9	450±86	110±38	3.6±0.39	110±39	560±94	9.3±1.6	100 <sup>e</sup>
339	BDE-77	380±140	0±0	380±140	87±47	0±0	87±47	460±140	7.7±2.4	100 <sup>e</sup>
340	BDE-85	100±24	0±0	100±24	24±8.8	0±0	24±8.8	130±26	2.1±0.42	100 <sup>d</sup>
341	BDE-99	740±170	0±0	740±170	170±61	0±0	170±61	910±190	15±3.1	100 <sup>e</sup>
342	BDE-100	800±260	5.3±1.3	810±270	180±94	1.1±0.26	180±92	990±280	16.6±4.7	100 <sup>e</sup>
343	BDE-119	1.6E3±490	0.69±0.47	1.6e3±490	370±170	0.14±0.09	370±170	2.0E3±500	33±8.3	100 <sup>e</sup>

344	BDE-126	150±35	0±0	150±35	35±14	0±0	35±14	180±37	3.0±0.62	100	e
345	BDE-138	2.8E3±940	0±0	2.8E3±940	540±210	0±0	540±210	3.3E3±950	55±16	200	e
346	BDE-153	1.5E4±3.3E3	0±0	1.5E4±3.3E3	3.1E3±330	0±0	3.1E3±330	1.8E4±3.2E3	290±54	200	e
347	BDE-154	4.9E3±1.4E3	0±0	4.9E3±1.4E3	1.0E3±340	0±0	1.0E3±340	5.9E4±1.4E3	99±24	200	e
348	BDE-156	1.4E3±470	0±0	1.4E3±470	310±150	0±0	310±150	1.7E3±490	28±8.3	200	e
349	BDE-180	8.7E4±1.5E4	0±0	8.7E4±1.5E4	1.9E4±5.0E3	0±0	1.9E4±5.0E3	1.0E5±1.6E4	1.8E3±260	200	e
350	BDE-183	6.8E4±1.5E3	0±0	6.8E4±1.5E3	1.4E4±4.7E3	0±0	1.4E4±4.6E3	8.2E3±1.6E3	1.4E3±270	200	e
351	BDE-184	9.7E3±3.3E3	0±0	9.7E3±3.3E3	1.9E3±790	0±0	1.9E3±790	1.2E4±3.5E3	190±58	200	e
352	BDE-194	4.1E3±1.0E3	0±0	4.1E3±1.0E3	860±320	0±0	860±320	5.0E3±1.1E3	83±18	3000	e
353	BDE-195	4.7E3±1.8E3	0±0	4.7E3±1.8E3	1.1E3±550	0±0	1.1E3±550	5.8E3±1.9E3	97±32	3000	e
354	BDE-196	1.7E4±4.0E3	0±0	1.7E4±4.0E3	3.7E3±1.3E3	0±0	3.7E3±1.3E3	2.1E4±4.2E3	350±71	3000	e
355	BDE-197	1.3E4±2.9E3	0±0	1.3E4±2.9E3	2.8E3±920	0±0	2.8E3±920	1.6E4±3.0E3	270±50	3000	e
356	BDE-198	f1.2E4±4.4E3	0±0	1.2E4±4.4E3	2.7E3±1.1E3	0±0	2.7E3±1.1E3	1.4E4±4.7E3	240±78	3000	e
357	BDE-201	1.3E4±3.4E3	0±0	1.3E4±3.4E3	2.9E3±1.2E3	0±0	2.9E3±1.2E3	1.6E4±3.7E3	270±61	3000	e
358	BDE-202	5.1E3±1.4E3	0±0	5.1E3±1.4E3	1.1E3±450	0±0	1.1E3±450	6.2E3±1.5E3	100±24	3000	e
359	BDE-204	1.4E34±420	0±0	1.4E34±420	300±130	0±0	300±130	1.7E3±440	29±7.4	3000	e

360	BDE-205	$205\pm75$	$0\pm0$	$205\pm75$	$432\pm25$	$0\pm0$	$432\pm25$	$248\pm78$	$4.1\pm1.3$	3000 e
361	BDE-206	$2.2E4\pm5.3E3$	$0\pm0$	$2.2E4\pm5.3E3$	$4.7E3\pm1.7E3$	$0\pm0$	$4.7E3\pm1.7E3$	$2.7E4\pm5.6E3$	$450\pm94$	3000 e
362	BDE-207	$2.8E4\pm6.8E3$	$0\pm0$	$2.8E4\pm6.8E3$	$5.9E3\pm2.2E3$	$0\pm0$	$5.9E3\pm2.2E3$	$3.4E4\pm7.2E3$	$570\pm120$	3000 e
363	BDE-208	$1.7E4\pm4.2E3$	$0\pm0$	$1.7E4\pm4.2E3$	$3.7E3\pm1.4E3$	$0\pm0$	$3.7E3\pm1.4E3$	$2.1E4\pm4.4E3$	$350\pm73$	3000 e
364	BDE-209	$2.9E4\pm5.9E3$	$0\pm0$	$2.9E4\pm5.9E3$	$6.2E3\pm2.2E3$	$0\pm0$	$6.2E3\pm2.2E3$	$3.5E4\pm6.3E3$	$590\pm100$	7000 e
365	$\sum$ PBDE	$3.6E5\pm7.2E4$	$120\pm27$	$3.6E5\pm7.2E4$	$7.7E4\pm2.3E3$	$25\pm5.6$	$7.7E4\pm2.3E3$	$4.4E5\pm7.6E4$	$7.3E3\pm1.3E3$	-
366	TIBP	$120\pm29$	$0\pm0$	$120\pm29$	$28\pm19$	$0\pm0$	$28\pm20$	$150\pm35$	$2.5\pm0.58$	-
367	TNBP	$370\pm45$	$0\pm0$	$370\pm45$	$71\pm19$	$0\pm0$	$71\pm19$	$440\pm49$	$7.4\pm0.81$	24000 c
368	TCEP	$1.1E5\pm1.6E4$	$0\pm0$	$1.1E5\pm1.6E4$	$2.6E4\pm4.8E3$	$0\pm0$	$2.6E4\pm4.8E3$	$1.4E5\pm1.6E4$	$2.3E3\pm270$	22000 c
369	TCIPP	$8.4E4\pm1.3E4$	$3.0E3\pm720$	$8.7E4\pm1.3E4$	$1.6E4\pm4.9E3$	$610\pm150$	$1.7E4\pm4.8E3$	$1.E5\pm1.4E4$	$1.7E3\pm230$	80000 c
370	TDCIPP	$4.2E4\pm1.5E4$	$0\pm0$	$4.2E4\pm1.5E4$	$8.7E3\pm5.8E3$	$0\pm0$	$8.7E3\pm5.8E3$	$5.1E4\pm1.6E4$	$850\pm270$	15000 c
371	TPHP	$1.0E5\pm2.6E4$	$330\pm230$	$1.0E5\pm2.6E4$	$2.1E4\pm9.6E3$	$69\pm46$	$2.1E4\pm9.6E3$	$1.2E5\pm2.7E4$	$2.1E3\pm460$	70000 c
372	EHDPP	$3.5E3\pm490$	$0\pm0$	$3.5E3\pm490$	$760\pm150$	$0\pm0$	$760\pm150$	$4.3E3\pm510$	$71\pm8.6$	-
373	ToCP	$1.3E3\pm360$	$0\pm0$	$1.3E3\pm360$	$270\pm120$	$0\pm0$	$270\pm120$	$1.6E3\pm370$	$27\pm6.3$	13000 c
374	$\sum$ OPFR	$3.5E5\pm3.7E4$	$3.5E3\pm660$	$3.5E5\pm3.7E4$	$7.3E4\pm9.4E3$	$730\pm130$	$7.4E4\pm9.4E3$	$4.2E5\pm3.8E4$	$7.0E3\pm630$	
375	<sup>a</sup> Data from Wang et al.; <sup>11</sup>									

376 <sup>b</sup> Reference dose is unavailable;

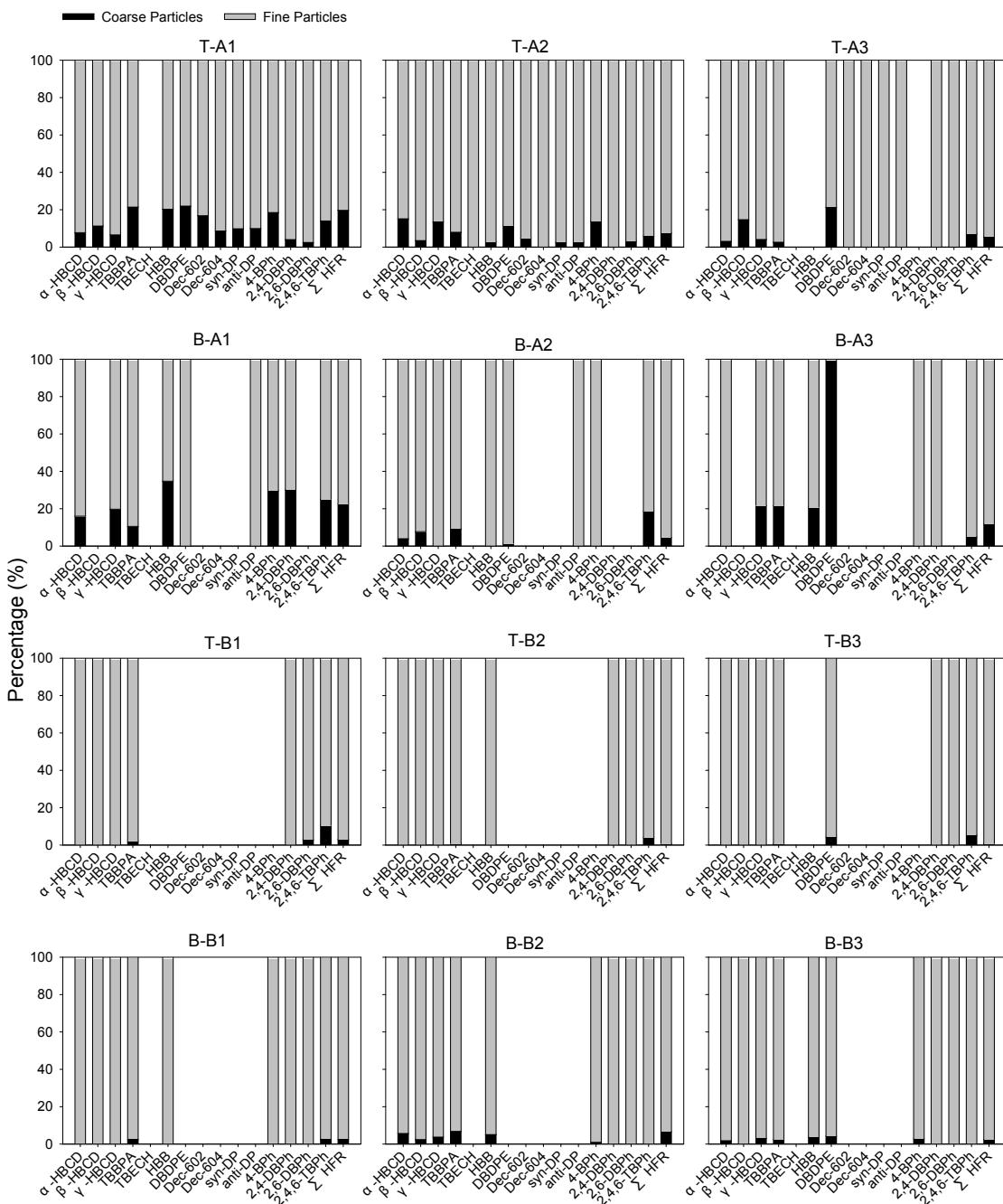
377 <sup>c</sup> Data from Ali et al.;<sup>12</sup>

378 <sup>d</sup> Data from Wang et al.;<sup>13</sup>

379 <sup>e</sup> Reference dose for some BDEs were suggested by the United States Environmental Protection Agency Integrated Risk Information System  
380 ([https://cfpub.epa.gov/ncea/iris\\_drafts/AtoZ.cfm](https://cfpub.epa.gov/ncea/iris_drafts/AtoZ.cfm)) and other BDE congeners were assumed to be equivalent reference dose of BDE congeners with the  
381 same number of bromine atoms;

382 <sup>f</sup> The sum of BDE-198, -203, -199, and -200.

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383

384 **Figure S1. The relative abundances of particle-bound halogenated flame retardants on**  
 385 **coarse ( $D_p > 1.8 \mu\text{m}$ ) and fine particles ( $D_p < 1.8 \mu\text{m}$ ).**

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