

## **Supporting Information**

**MANUSCRIPT TITLE:** Size Distribution of Airborne Particle-Bound Polybrominated Diphenyl Ethers and Its Implications for Dry and Wet Deposition

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**NO. OF TABLES:** 7

**NO. OF FIGURES:** 3

**NO. OF PAGES:** 16

### **Lists of PBDEs cited in the main text**

List S1: BDE-17, 28, 47, 66, 85, 99, 100, 138, 153, 154, 183 and 209

List S2: BDE-28, 47, 66, 99, 100, 138, 153, 154, 183 and 209

List S3: BDE-7, 8, 11, 12, 13, 15, 17, 25, 28, 32, 33, 35, 47, 49, 66, 85, 99, 100, 116, 119, 138, 153, 154, 155, 166, 183, 190, 196, 197, 201, 202, 203, 206, 207, 208 and 209

**Table S1. Dry Deposition Velocities ( $V_i$ , cm s<sup>-1</sup>) and Wet Removal Efficiencies ( $\eta_i$ , %) of Size-Fractionated Particles**

$D_p^a$	20	13.4	7.5	4.2	2.4	1.3	0.75	0.42	0.24	0.13	0.075
$V_i^b$	1.82	0.56	0.19	0.080	0.051	0.053	0.071	0.10	0.17	0.28	0.49
$\eta_i^c$	100	100	95	80	75	65	35	60	65	55	50

<sup>a</sup> Particle average aerodynamic diameter. <sup>b</sup> Data from Zhang et al.<sup>1</sup> <sup>c</sup> Data from Radke et al.<sup>2</sup>

**Table S2. Median and Range of Concentrations (pg m<sup>-3</sup>) of Particle-Bound PBDEs in the Atmosphere at Different Heights above the Ground in the E-Waste Recycling Zone**

	1.5 m	5 m	20 m
BDE-28	9.7 (5.0–33)	10.7 (1.3–18.9)	9.0 (2.6–29)
BDE-47	81 (40–170)	74 (11.5–120)	78 (44–270)
BDE-66	11.3 (3.7–25)	15.3 (2.6–37)	12.6 (3.9–72)
BDE-85	12.6 (6.3–28)	21 (1.0–78)	20 (6.3–97)
BDE-99	31 (15.5–150)	57 (5.1–160)	42 (13.9–190)
BDE-100	12.2 (4.2–110)	36 (3.4–69)	10.7 (2.9–120)
BDE-153	102 (95–120)	68 (15.4–480)	100 (8.1–730)
BDE-154	55 (46–63)	101 (11.2–330)	54 (10.3–230)
BDE-181	30 (18.5–40)	54 (5.9–120)	31 (4.9–160)
BDE-183	89 (75–110)	170 (57.0–370)	96 (17.4–330)
BDE-190	27 (21–31)	42 (8.3–97)	28 (8.0–130)
BDE-196	310 (250–340)	490 (112–1200)	360 (43–900)
BDE-203	68 (54–80)	140 (16.5–300)	80 (19.5–170)
BDE-204	171 (150–230)	440 (85–970)	270 (37–400)
BDE-206	400 (250–680)	570 (190–1400)	570 (220–810)
BDE-207	600 (420–680)	980 (510–2300)	1000 (360–1400)
BDE-208	58 (48–69)	95 (32–280)	110 (23–220)
BDE-209	3600 (2500–4100)	4300 (3700–5300)	5500 (2600–7900)
$\sum \text{PBDE}^a$	5700 (4500–6400)	7700 (5200–12300)	9200 (3400–11500)

<sup>a</sup> Sum of 18 PBDE congeners.

**Table S3. *t*-test Results (values < 0.05 are highlighted) of Particle-**

**Bound PBDEs in the Atmosphere at Different Heights above the Ground in the E-Waste Recycling Zone**

	1.5 m vs 5 m	1.5 m vs 20 m	5 m vs 20 m
BDE-28	0.596	0.786	0.793
BDE-47	0.298	0.791	0.299
BDE-66	0.249	0.298	0.750
BDE-85	0.165	0.256	0.849
BDE-99	0.583	0.958	0.559
BDE-100	0.238	0.879	0.806
BDE-153	0.484	0.425	0.789
BDE-154	0.127	0.604	0.235
BDE-181	0.100	0.343	0.572
BDE-183	0.064	0.368	0.244
BDE-190	0.115	0.271	0.739
BDE-196	0.075	0.227	0.275
BDE-203	0.070	0.321	0.146
BDE-204	<b>0.047</b>	0.122	0.115
BDE-206	0.191	0.310	0.461
BDE-207	<b>0.032</b>	<b>0.007</b>	0.581
BDE-208	0.104	0.068	0.899
BDE-209	<b>0.017</b>	<b>0.008</b>	0.066
$\sum$ PBDE <sup>a</sup>	<b>0.024</b>	<b>0.010</b>	0.698

<sup>a</sup> Sum of 18 PBDE congeners.

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**Table S4. Geometric Mean Diameter (GMD,  $\mu\text{m}$ )<sup>a</sup> of PBDEs Calculated for the Coarse ( $> 1.8 \mu\text{m}$ ), Fine ( $< 1.8 \mu\text{m}$ ) and Total Particles at Different Heights above the Ground in the E-Waste Recycling Zone**

	1.5 m			5 m			20 m		
	Coarse	Fine	Total	Coarse	Fine	Total	Coarse	Fine	Total
BDE-28	8.04±1.88	0.34±0.07	1.93±0.85	6.94±1.05	0.36±0.05	1.34±0.38	7.23±2.68	0.33±0.09	1.52±1.51
BDE-47	7.04±1.01	0.33±0.08	1.31±0.55	7.19±0.99	0.33±0.04	1.39±0.44	6.84±1.19	0.34±0.08	1.11±0.24
BDE-66	8.28±1.14	0.35±0.07	2.47±1.98	7.21±1.43	0.33±0.05	1.48±0.59	6.86±2.46	0.35±0.11	1.46±1.36
BDE-85	8.88±1.54	0.35±0.06	2.18±0.97	7.58±2.32	0.33±0.06	1.55±0.77	7.00±2.36	0.39±0.11	1.66±1.34
BDE-99	8.31±0.68	0.35±0.03	1.94±0.27	7.30±1.54	0.38±0.05	1.58±0.64	6.75±1.70	0.39±0.17	1.27±0.43
BDE-100	7.78±1.10	0.39±0.20	2.18±1.85	6.78±2.13	0.38±0.13	1.72±1.48	6.73±3.02	0.40±0.12	1.98±1.78
BDE-153	8.23±0.62	0.37±0.03	2.40±0.42	7.12±1.95	0.32±0.05	1.67±0.94	7.53±2.50	0.37±0.10	1.62±1.39
BDE-154	7.73±0.55	0.36±0.04	2.32±0.28	7.04±2.05	0.35±0.08	1.57±0.73	6.28±1.71	0.41±0.09	1.10±0.43
BDE-181	8.68±0.94	0.39±0.03	2.39±0.27	6.86±1.54	0.38±0.07	1.50±0.63	5.93±1.01	0.38±0.08	0.98±0.31
BDE-183	8.43±0.59	0.40±0.03	2.16±0.26	7.54±2.22	0.36±0.06	1.87±1.00	6.12±1.29	0.38±0.09	1.03±0.37
BDE-190	8.40±0.48	0.38±0.04	2.35±0.19	7.04±1.39	0.39±0.08	1.56±0.56	5.96±0.71	0.39±0.07	1.05±0.29
BDE-196	8.21±1.08	0.30±0.02	2.18±0.40	6.98±1.64	0.36±0.09	1.67±0.76	6.11±1.47	0.46±0.08	1.16±0.25
BDE-203	8.13±0.65	0.33±0.01	2.12±0.18	7.14±1.49	0.38±0.08	1.80±0.61	5.52±0.97	0.47±0.07	1.24±0.38
BDE-204	8.68±1.36	0.29±0.01	2.30±0.48	7.16±1.47	0.36±0.07	1.86±0.67	5.70±1.34	0.49±0.10	1.26±0.32
BDE-206	8.71±1.38	0.29±0.05	2.07±0.77	7.52±1.06	0.35±0.04	1.80±0.56	5.87±0.99	0.46±0.06	1.19±0.27
BDE-207	9.31±0.62	0.33±0.05	1.99±0.64	7.36±1.57	0.33±0.08	1.35±0.48	5.82±1.09	0.46±0.09	1.19±0.32
BDE-208	9.40±0.97	0.30±0.01	1.78±0.29	7.44±1.56	0.36±0.07	1.75±0.54	5.83±1.06	0.48±0.08	1.23±0.28
BDE-209	8.76±0.63	0.26±0.03	1.61±0.15	7.01±0.75	0.32±0.03	1.65±0.17	5.88±0.93	0.46±0.08	1.48±0.39

$\Sigma$ PBDE <sup>b</sup>	8.71±0.49	0.28±0.02	1.75±0.20	7.02±0.79	0.33±0.04	1.59±0.35	5.95±0.95	0.45±0.07	1.36±0.32
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<sup>a</sup> Calculated as  $\log \text{GMD} = \frac{\sum (C_i \times \log D_{pi})}{\sum C_i}$ ; where  $C_i$  is the concentration of target compound in size class i and  $D_{pi}$  is the geometric mean particle diameter collected on stage i. <sup>b</sup> Sum of 18 PBDE congeners.

**Table S5. Geometric Standard Deviations (GSD,  $\mu\text{m}$ )<sup>a</sup> of Particle-Bound PBDEs Calculated for the Coarse ( $> 1.8 \mu\text{m}$ ) and Fine ( $< 1.8 \mu\text{m}$ ) Particles at Different Heights above the Ground in the E-Waste Recycling Zone**

	1.5 m			5 m			20 m		
	Coarse	Fine	Total	Coarse	Fine	Total	Coarse	Fine	Total
BDE-28	2.09±0.11	3.68±0.62	5.62±0.68	2.10±0.13	4.12±0.54	5.20±0.63	2.00±0.23	4.03±0.66	4.72±0.90
BDE-47	2.10±0.11	4.12±0.35	5.51±0.80	2.13±0.07	4.21±0.75	5.57±0.35	2.13±0.11	4.06±0.42	5.31±0.73
BDE-66	2.14±0.08	3.52±0.92	5.42±1.07	2.04±0.13	4.00±0.68	5.34±0.70	2.00±0.24	3.98±0.65	4.67±0.84
BDE-100	2.16±0.09	3.26±0.97	5.21±1.27	2.08±0.24	3.27±0.74	4.69±1.10	1.84±0.45	3.84±0.97	4.44±1.11
BDE-99	2.17±0.05	4.06±0.48	5.64±0.29	2.09±0.10	3.94±0.78	5.19±0.39	2.05±0.14	3.93±0.75	4.84±1.31
BDE-85	2.08±0.22	3.61±0.50	5.69±0.95	2.04±0.20	4.03±0.62	5.40±0.86	2.03±0.28	3.99±0.47	4.62±0.64
BDE-154	2.16±0.06	3.69±0.47	5.22±0.32	2.05±0.17	3.85±0.84	5.21±0.88	2.04±0.15	3.71±0.65	4.33±0.51
BDE-153	2.11±0.08	3.48±0.30	5.20±0.27	1.99±0.27	4.76±0.74	4.87±1.06	1.96±0.16	3.95±0.83	4.81±0.97
BDE-183	2.11±0.07	3.70±0.18	5.24±0.26	2.08±0.13	3.96±0.78	5.15±0.46	2.07±0.07	3.94±0.44	4.63±0.52
BDE-181	2.10±0.06	3.73±0.12	5.22±0.26	2.10±0.17	3.89±0.76	5.01±0.68	2.11±0.08	4.00±0.41	4.62±0.60
BDE-190	2.11±0.06	3.65±0.16	5.28±0.10	2.13±0.14	3.84±0.69	5.04±0.52	2.12±0.07	3.97±0.48	4.61±0.53
BDE-203	2.17±0.06	4.38±0.54	5.54±0.16	2.11±0.16	3.99±0.77	5.09±0.46	2.04±0.11	4.05±0.44	4.19±0.50
BDE-204	2.14±0.12	5.12±0.33	5.49±0.38	2.07±0.15	4.14±1.02	5.09±0.38	2.00±0.11	3.91±0.59	4.19±0.62

BDE-196	2.18±0.09	5.09±0.31	5.37±0.33	2.12±0.18	4.03±0.85	5.12±0.39	2.04±0.13	3.84±0.55	4.40±0.54
BDE-208	2.10±0.04	4.92±0.19	6.12±0.35	2.03±0.20	4.30±1.31	5.15±0.47	2.06±0.09	3.83±0.61	4.22±0.65
BDE-207	2.09±0.09	4.48±0.60	5.96±0.55	2.09±0.19	4.64±1.57	5.39±0.49	2.11±0.10	3.94±0.49	4.37±0.69
BDE-206	2.07±0.28	4.73±0.56	5.75±0.64	2.02±0.15	4.20±1.04	5.19±0.38	2.11±0.07	3.93±0.43	4.40±0.60
BDE-209	2.18±0.08	4.91±0.13	6.50±0.45	2.06±0.04	4.75±0.44	5.53±0.52	2.08±0.11	3.99±0.30	4.41±0.65
$\sum$ PBDE <sup>b</sup>	2.16±0.07	4.80±0.16	6.22±0.38	2.08±0.07	4.49±0.68	5.44±0.44	2.09±0.08	3.98±0.34	4.48±0.60

<sup>a</sup> calculated as  $(\log \text{GSD})^2 = \frac{\sum [C_i \times (\log D_{pi} - \log \text{GMD})^2]}{\sum C_i}$ ; where  $C_i$  is the concentration of compound in size class i and  $D_{pi}$  is the geometric mean particle diameter collected on stage I, GMD is geometric mean diameter, calculated as  $\log \text{GMD} = \frac{\sum (C_i \times \log D_{pi})}{\sum C_i}$ ; where  $C_i$  is the

mean particle diameter collected on stage I, GMD is geometric mean diameter, calculated as  $\log \text{GMD} = \frac{\sum (C_i \times \log D_{pi})}{\sum C_i}$ ; where  $C_i$  is the concentration of compound in size class i and  $D_{pi}$  is the geometric mean particle diameter collected on stage i. <sup>b</sup> Sum of 18 PBDE congeners.

**Table S6. Calculated dry ( $F_{dry}$ ) and Wet ( $F_{wet}$ ) Deposition Fluxes (ng m<sup>-2</sup> d<sup>-1</sup>) of PBDEs in Size-Fractionated Particles at 20 m Height above the Ground in the E-Waste Recycling Zone**

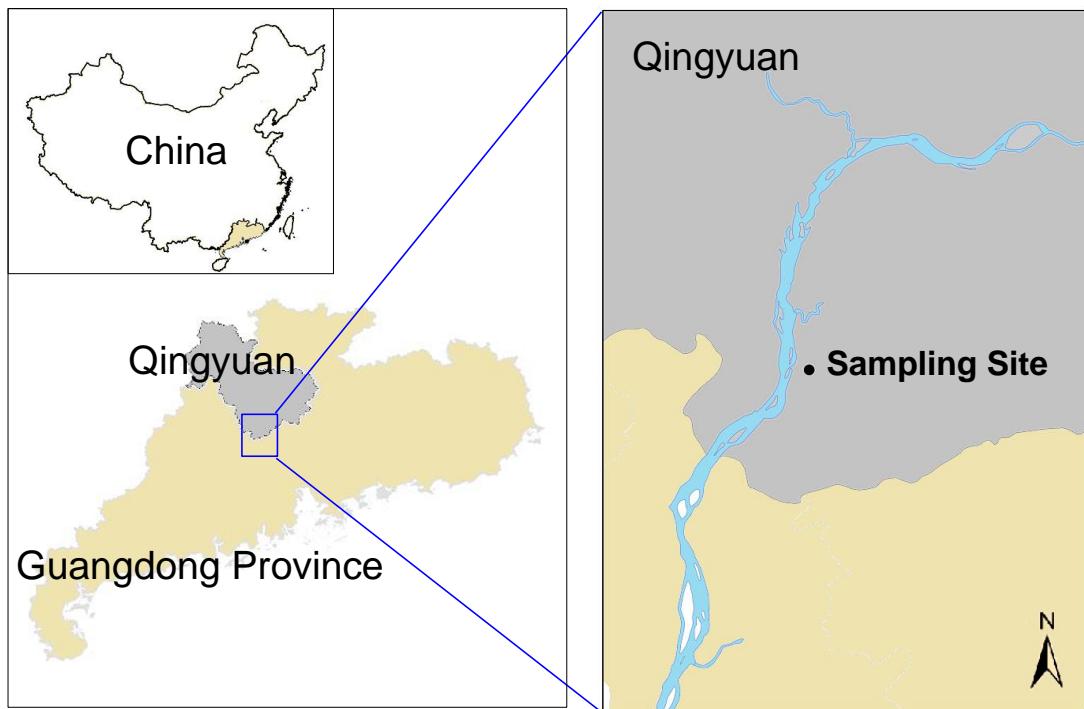
	Coarse Particle <sup>a</sup>			Fine Particle <sup>b</sup>			Total <sup>c</sup>		
	$F_{dry}$	$F_{wet}$	$F_{total}^d$	$F_{dry}$	$F_{wet}$	$F_{total}$	$F_{dry}$	$F_{wet}$	$F_{total}$
BDE-28	2.2±3.4	0.87±1.26	3.1±4.7	0.97±0.64	0.63±0.46	1.60±1.09	3.2±3.5	1.51±1.33	4.7±4.9
BDE-47	16.7±13.8	6.4±4.9	23±19	10.5±8.5	6.2±4.8	16.7±13.3	27±22	12.6±9.6	40±32
BDE-66	3.3±4.2	1.31±1.55	4.6±5.8	2.2±3.1	1.49±2.13	3.7±5.2	5.5±5.8	2.8±2.8	8.3±8.5
BDE-85	4.4±4.8	1.77±1.73	6.2±6.5	2.8±4.2	1.85±2.75	4.6±7.0	7.2±7.4	3.6±3.6	10.8±10.9
BDE-99	6.3±4.1	2.65±1.41	8.9±5.4	5.7±8.0	4.1±5.5	9.8±13.5	12.0±11.5	6.7±6.6	18.7±18.0
BDE-100	2.9±4.9	1.18±1.57	4.1±6.4	1.9±2.0	1.30±1.51	3.2±3.4	4.8±5.6	2.5±2.5	7.3±7.9

BDE-153	21±27	8.6±10.7	30±37	15±24	10.8±18.5	26±43	36±38	19.4±21.8	55±59
BDE-154	6.0±4.3	2.8±1.4	8.8±5.4	7.5±11.0	5.3±7.3	12.8±18.3	13.5±13.1	8.1±8.1	22±21
BDE-181	4.7±4.4	2.1±1.6	6.8±5.9	4.8±5.6	3.4±4.1	8.2±9.6	9.5±9.9	5.5±5.6	15.0±15.4
BDE-183	12.9±8.6	5.9±3.3	19±12	13.2±12.7	8.9±8.6	22±21	26±21	14.9±11.3	41±32
BDE-190	4.6±3.8	2.1±1.5	6.7±5.3	4.2±4.2	3.0±3.2	7.2±7.5	8.7±8.0	5.1±4.7	13.8±12.7
BDE-196	44±27	23±14	68±40	35±22	27±16	62±38	80±45	50±30	130±74
BDE-203	9.1±5.7	5.1±3.9	14.2±9.3	6.3±3.2	4.9±2.5	11.2±5.6	15.4±7.5	10.0±5.8	25±13
BDE-204	28±17	15.0±6.9	43±24	21±11	16.3±8.5	37±19	49±24	31±14	80±36
BDE-206	66±28	30±10	96±37	46±21	35±16	81±36	112±45	65±24	177±66
BDE-207	117±48	54±19	170±62	78±27	59±20	137±45	195±65	113±35	310±90
BDE-208	10.4±3.8	5.4±2.2	16±5.8	7.8±5.0	6.6±4.4	14.4±9.3	18.3±8.4	11.9±6.4	30±14
BDE-209	840±270	400±110	1240±340	410±150	320±120	730±250	1240±330	720±210	1960±480
ΣPBDE <sup>e</sup>	1200±360	570±140	1770±450	670±230	510±180	1180±410	1870±450	1100±310	3000±680

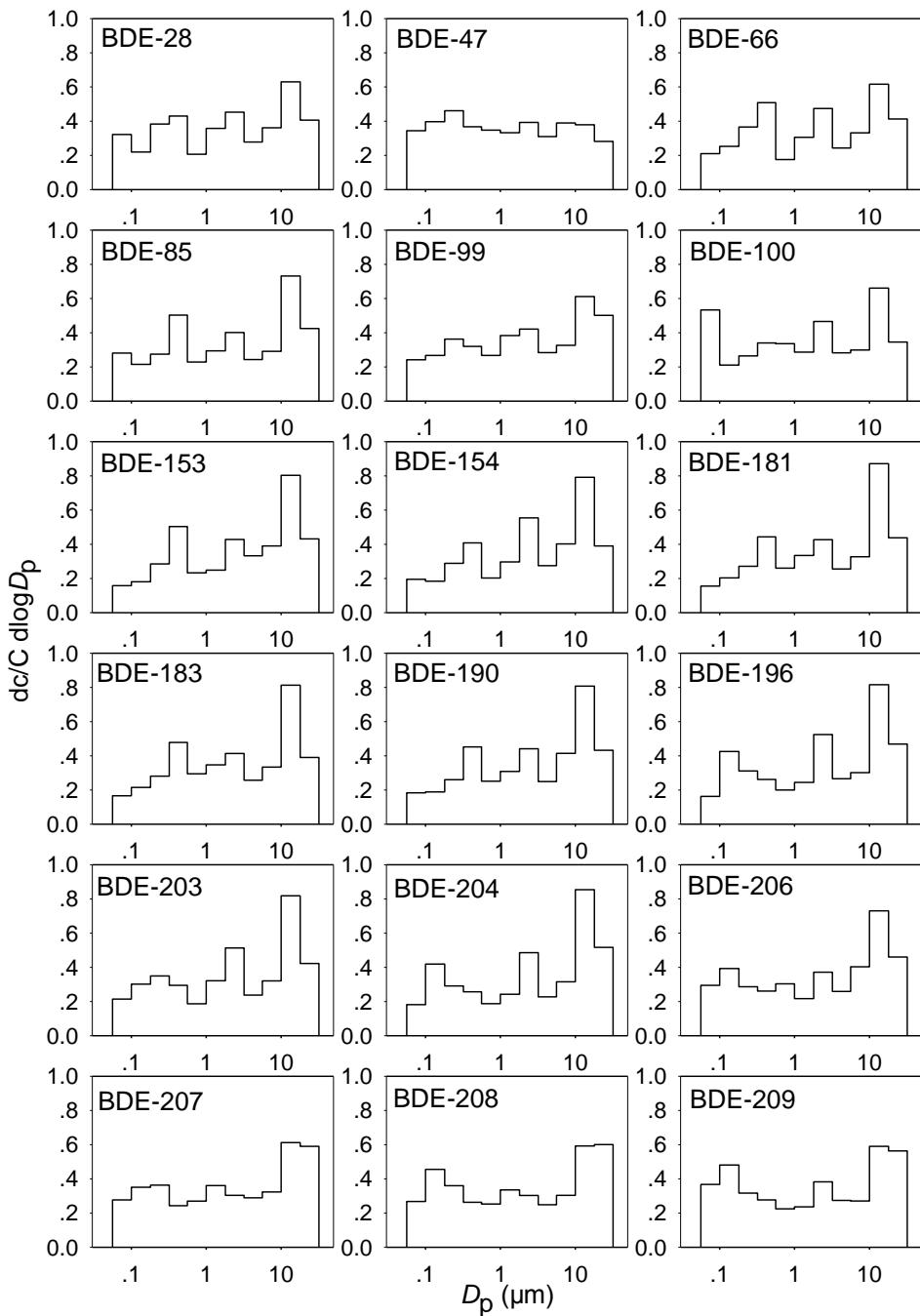
<sup>a</sup> Particle aerodynamic diameters ( $D_p$ ) > 1.8  $\mu\text{m}$ . <sup>b</sup>  $D_p$  < 1.8  $\mu\text{m}$ . <sup>c</sup> Sum of deposition fluxes of coarse and fine particles. <sup>d</sup> Sum of dry and wet deposition fluxes. <sup>e</sup> Sum of 18 PBDE congeners.

**Table S7. Dry Deposition Velocities ( $V_d$ , cm s<sup>-1</sup>) and Wet Removal Efficiencies ( $\eta$ , %) of Size-Fractionated PBDEs at 20 m Height above the Ground in the E-Waste Recycling Zone**

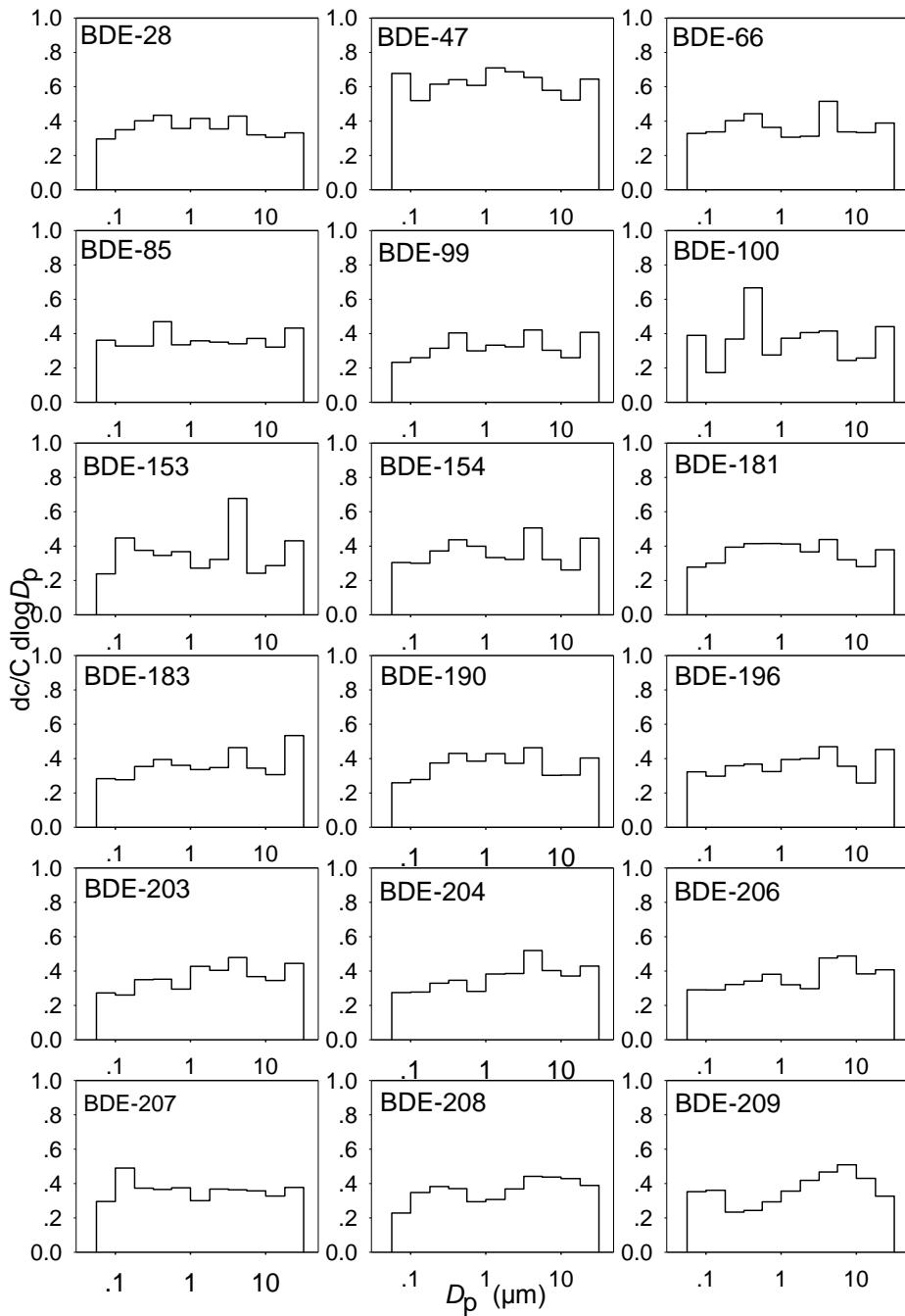
	$V_d$	$\eta$
BDE-28	0.29±0.10	69.3±8.1
BDE-47	0.29±0.05	67.1±2.8
BDE-66	0.28±0.10	68.5±8.0
BDE-85	0.29±0.09	70.2±7.0
BDE-99	0.28±0.09	69.2±4.9
BDE-100	0.27±0.10	67.6±7.4
BDE-153	0.28±0.11	67.1±10.6
BDE-154	0.24±0.07	66.7±3.5
BDE-181	0.25±0.05	65.0±3.6
BDE-183	0.25±0.04	65.6±3.5
BDE-190	0.25±0.04	65.5±3.3
BDE-196	0.24±0.06	66.0±3.0
BDE-203	0.23±0.05	66.2±4.4
BDE-204	0.23±0.06	66.9±2.9
BDE-206	0.25±0.06	66.3±2.8
BDE-207	0.25±0.07	66.1±3.0
BDE-208	0.23±0.06	66.2±3.4
BDE-209	0.26±0.07	69.3±2.8



**Figure S1.** Map showing the location of Qingyuan City in Guangdong Province of China and the sampling site within the e-waste recycling zone in Qingyuan.



**Figure S2.** Size distributions of particle-bound PBDEs in the atmosphere at 1.5 m height above the ground in the e-waste recycling zone. dc is the mass concentration on each filter, C is the sum concentration on all filters, and  $d\log D_p$  is the logarithmic size interval for each impactor stage in aerodynamic diameter ( $D_p$ ).



**Figure S3.** Size distributions of particle-bound PBDEs in the atmosphere at 5 m height above the ground in the e-waste recycling zone. dc is the mass concentration on each filter, C is the sum concentration on all filters, and  $d\log D_p$  is the logarithmic size interval for each impactor stage in aerodynamic diameter ( $D_p$ ).

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- (2) Radke, L. F.; Hobbs, P. V.; Eltgroth, M. W., Scavenging of aerosol particles by precipitation. *J. Appl. Meteorol.* **1980**, *19* (6), 715–722.