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## Selected Organohalogenated Flame Retardants in Egyptian Indoor and Outdoor Environments: Levels, Sources and Implications for Human Exposure

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1 **Selected Organohalogenated Flame Retardants in Egyptian Indoor and Outdoor**  
2 **Environments: Levels, Sources and Implications for Human Exposure**

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31 **Highlights**

- 32 • Indoor levels of flame retardants were significantly higher than outdoors.
- 33 • PBDEs>NFRs and BDE-47, 99, HBB, BTBPE and DDC-CO were the most abundant
- 34 analytes.
- 35 • Significant log-linear  $K_{\text{dust-air}}-K_{\text{OA}}$  relationships indicated an equilibrium state.
- 36 • Low adverse health effects from inhalation/ingestion/dermal exposure pathways.

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## 60 Abstract

61 There is scant information on the presence of the polybrominated diphenyl ethers (PBDEs) and  
62 other alternative flame-retardants (NFRs) in Africa. Hence, to investigate their levels, sources,  
63 and human exposure scenarios, elevated fine dust (EFD) samples from apartments (n = 12),  
64 working places (n = 9) and cars (n = 12), floor dust (FD) samples (n = 5) and outdoor dust  
65 samples (n = 21) were collected from Alexandria, Egypt during 2014. Gaseous concentrations  
66 were estimated using low density polyethylene sheets (n = 33 and 21 for indoor and outdoor sites  
67 respectively). Indoor gaseous and dust PBDE (7.0 – 300 pg/m<sup>3</sup>; 4.0 – 770 ng/g), and NFR (0.40 –  
68 48 pg/m<sup>3</sup>; 0.50 – 8.5 ng/g) concentrations were significantly higher (p = 0.004 – 0.02) than  
69 outdoor concentrations (PBDEs: 3.0 – 41 pg/m<sup>3</sup>, 1.5 – 195 ng/g; NFRs: 0.20 – 13 pg/m<sup>3</sup>, 0.50 –  
70 4.0 ng/g). Median PBDE concentration in cars (210 ng/g) was higher in apartments and working  
71 places (129 and 131 ng/g respectively). PBDE concentrations in FD were 7.0 – 14-folds lower  
72 than EFD concentrations. Outdoor PBDE concentrations were significantly higher (p < 0.01) at  
73 residential – industrial places with older buildings. All samples were dominated by BDE-47 and  
74 99. HBB, BTBPE and DDC-CO were the most abundant NFRs in EFD samples. Profiles of  
75 PBDE and NFR in FD closely matched those of outdoor dust, indicating a possible carryover  
76 from the outdoor environment. Although factors such as number of electronics, construction year  
77 and floor type significantly correlated with the majority of PBDE congeners and some NFRs in  
78 apartments and working places, sources were not clearly identified for NFRs. Significant log-  
79 linear relationships were obtained between theoretical and calculated dust-air partitioning  
80 coefficients for all samples indicating an equilibrium state between dust and vapor. Low  
81 possibility of occurrence of adverse health effects was concluded, with the inhalation pathway  
82 (for adults) and dust ingestion (for children) acting as the most important exposure routes.

83 **Keywords:** PBDEs, NFRs, LDPE, Indoor, Outdoor, dust, health effects

## 84 **1. Introduction**

85 Polybrominated diphenyl ethers (PBDEs) have been widely used worldwide since the 1970s (de  
86 Wit, 2002) to reduce fire risks. Although their production has phased out, they still exist in  
87 products and thus become released into the environment (Abbasi et al., 2015). Due to the phase  
88 out of PBDEs (Abbasi et al., 2015), **alternative** halogenated (NFRs) and organophosphate flame  
89 retardants (OPFRs) are now widely used in consumer products as substitutes (McDonough et al.,  
90 2016). PBDEs and NFRs can accumulate in humans (Leonetti et al., 2016; Liu et al., 2015) and  
91 thus numerous adverse health effects could occur (Costa and Giordano, 2007; Nakari and  
92 Huhtala, 2010).

93  
94 **PBDEs and NFRs** have been detected in the indoor air (Al-Omran and Harrad, 2016; Cao et al.,  
95 2014; Fromme et al., 2014; Hassan and Shoeib, 2015; Kademoglou et al., 2017; Shoeib et al.,  
96 2012) and outdoor air and dust **worldwide** (Kurt-Karakus et al., 2017; Mahmood et al., 2015;  
97 McDonough et al., 2016; Salamova et al., 2014). The indoor environment with expected higher  
98 concentrations and longer exposure periods received special attention (Wilford et al., 2004).

99 The majority of the indoor studies concentrated on the floor dust (FD) (Ali et al., 2013; Cequier  
100 et al., 2014; Dodson et al., 2012; Fromme et al., 2014; Hassan and Shoeib, 2015; Kurt-Karakus  
101 et al., 2017; Shoeib et al., 2012). Few studies investigated (elevated) fine dust collected ~ 1  
102 meter above the ground (Al-Omran and Harrad, 2016; Newton et al., 2015; Thuresson et al.,  
103 2012). In countries with high ambient particulate levels and dust content like Egypt, we  
104 hypothesized that floor dust will not properly represent the indoor environment due to the  
105 possible carryover of dust and dirt by the shoes from outdoors. However, although dust collected

106 from elevated surfaces would be a better representative of exposure for adults, children are more  
107 exposed to the floor dust.

108 Low density polyethylene passive samplers (LDPE) have been widely used for monitoring  
109 hydrophobic organic compounds (HOCs) in the atmosphere (Bartkow et al., 2004; Kennedy et  
110 al., 2007; Khairy and Lohmann, 2014, 2013; McDonough et al., 2016). However, they have been  
111 rarely used to monitor indoor air pollution. HOCs accumulate in passive air samplers via  
112 diffusion and absorption into the sampler matrix, with a high enrichment in LDPE (Sacks and  
113 Lohmann, 2011) thus offering lower detection limits compared to conventional samplers.  
114 Accordingly, LPDE can be used as a very useful tool in screening level risk assessment studies.

115  
116 Although **PBDEs and NFRs** were extensively investigated in the indoor dust and the ambient air  
117 worldwide, only few studies investigated their concentrations in the indoor dust (Abdallah and  
118 Covaci, 2014; Hassan and Shoeib, 2015) and ambient air (Arinaitwe et al., 2014; Pozo et al.,  
119 2009) collected from Africa. Additionally, none of the studies investigated **their** levels in the  
120 elevated fine dust (EFD), outdoor dust and/or their gaseous levels in the indoor and outdoor  
121 environments.

122 In the current study, we studied the levels of PBDEs and **NFRs** in the indoor and outdoor  
123 environments of Alexandria, Egypt. Vapor phase (using LDPE), in- and outdoor dust were  
124 collected to: i) determine concentrations, profiles and sources of **PBDEs and NFRs** in the indoor  
125 and outdoor air (gas phase + dust), ii) compare **their** concentrations among **the** different indoor  
126 microenvironments, and between floor dust and elevated fine dust, iii) assess the link between  
127 outdoor dust and indoor contamination, and iv) estimate the human intakes via inhalation and  
128 dust ingestion for adults and toddlers. To our knowledge, this is the first comprehensive study to

129 measure **PBDEs and NFRs** in the indoor and outdoor environments, and to utilize LDPE as an  
130 indoor passive sampler.

131  
132 **2. Materials and methods**

133 Detailed descriptions of the study area, LDPE passive sampling, extraction, cleanup,  
134 instrumental analysis, and estimation of the vapor phase concentrations from LDPE are provided  
135 in Appendix A, and are briefly summarized below.

136  
137 **2.1. Sampling**

138 Indoor and outdoor samples were collected from Alexandria, Egypt (Figure A1) during the late  
139 autumn – winter (minimum ventilation effect) of 2014 (November – December). EFD was  
140 collected 1 m above the floor from homes (n = 12) and working places (n = 9) using an  
141 Electrolux JetMaxx Bag Canister Vacuum cleaner (Model EL4042A). **Dust was collected in**  
142 **cellulose thimbles fitted between the crevice tool and the vacuum tube. The thimble was secured**  
143 **with a stainless-steel ring. At each sampled homes, dust was collected from the living area**  
144 **including the living room, dining room and the bedrooms, where people spend most of their**  
145 **indoor times (greatest exposure likelihood), and many products exist that may be sources of the**  
146 **target flame retardants. Prior to sampling, participants were asked not to vacuum or ventilate**  
147 **their places for a week. The crevice tool was passed on all the available surfaces in the rooms.**  
148 **After each of the 6 dust collection events at each location, the thimble was carefully removed,**  
149 **wrapped in a pre-combusted aluminum foil, sealed in plastic zip bag, and stored at -4 °C. After**  
150 **the final sampling event, thimbles were stored at -20 °C until analysis. Similarly, EFD from cars**  
151 **(n = 12) were obtained from all available surfaces except the floor (to prevent the effect of**  
152 **carryover from street dust). Participants were asked not to vacuum their vehicles at least two**

153 weeks before sampling and until the end of the sampling event (45 days). 15 indoor samples  
154 (other than what was analyzed in the current study) were excluded from further consideration  
155 because volunteers failed to follow our pre-sampling guidelines (see Appendix A).

156  
157 FD was collected from selected homes (n = 5) using the same vacuum cleaner equipped with  
158 dust collection bag (Schreder and La Guardia, 2014). Information about the sampled indoor  
159 microenvironments is given in Table A1.

160  
161 For outdoor samples (n = 21), settled dust on elevated smooth surfaces were collected using  
162 small plastic brushes and a small aluminum shovel from the same locations as the indoor  
163 samples, and stored in small amber glass jars at -20 °C until analysis.

164  
165 For the outdoor vapor – phase samples, LDPE sheets were pre-cleaned, spiked with performance  
166 reference compounds (PRCs) according to Booij et al. (2002) and deployed in duplicates in the  
167 atmosphere for 2 months at the same sites as the outdoor dust (n = 42) in two inverted bowls as  
168 a shelter for protection against sunlight and precipitation (see Appendix A for more details).

169  
170 Indoor LDPE sheets were deployed in duplicates for 2 months at each of the sampled rooms (n =  
171 2 – 3) of the living area starting two weeks before the dust sampling campaign. Samplers were  
172 hung in protected places away from direct exposure to the light and direct contact with  
173 participants. The total number of deployed LDPE were 46 (23 x duplicates) at apartments, 24 at  
174 the cars, and 38 at working places (total = 108 samples). For the extraction/analysis step, all the  
175 LDPE sheets deployed at the different rooms within an apartment or working place were  
176 combined and analyzed together (n = 12, 9 and 12 for apartments, working places and cars  
177 respectively x duplicates).

## 178 2.2. Extraction, cleanup and instrumental analysis

179 Dust samples (indoor and outdoor) were sieved through a 3-inch, 100 mesh (150  $\mu\text{m}$ ) stainless  
180 steel sieve (Dodson et al., 2012), and  $\sim 0.5$  g dry weight were **sonicated** with n-hexane/acetone  
181 (1:1, v:v) after spiking with surrogate standards (**10  $\mu\text{L}$  of a 2.0 ng/ $\mu\text{L}$  solution of  $^{13}\text{C}_{12}$  BDE-28,**  
182 **47, 99, 153 and 183 in nonane**), purified and fractionated (fraction1 containing target analytes  
183 and fraction 2 containing organophosphate flame retardants, which is not discussed in the current  
184 manuscript) over silica gel and concentrated to a final volume of  $\sim 25$   $\mu\text{L}$  after the addition of the  
185 injection standards. LDPE samplers were spiked with surrogate standards and cold extracted  
186 twice **with methylene chloride and n-hexane** for 24 h each with no further cleanup.

187  
188 Extracts were analyzed for 12 PBDE congeners (BDE-2, 8, 15, 28, 30, 47, 49, 99, 100, 153, **154**  
189 **and 183**) and 9 **NFRs** [tetrabromo-*p*-xylene (TBX), pentabromobenzene (PBB),  
190 pentabromotoluene (PBT), pentabromoethylbenzene (PBEB), hexabromobenzene (HBB),  
191 hexachlorocyclo-pentadienyl-dibromocyclooctane (DBHCTD), 1,2-bis (2,4,6-tribromophenoxy)  
192 ethane (BTBPE), and anti- and syn-isomers of Dechlorane Plus (a-DDC-CO and s-DDC-CO)] on  
193 an Agilent 6890N gas chromatograph coupled to a Waters Quattro Micro mass spectrometer  
194 (GCMS/MS) in electron ionization mode (EI, 70 eV) using multiple reaction monitoring (MRM).

## 195 196 2.3. Quality assurance

197  
198 **Field blanks (Table A2), matrix spikes, and duplicate samples were included with each sample**  
199 **batch. Field blanks were composed of anhydrous sodium sulphate (3 g) contained in either a**  
200 **thimble, dust bag or a glass jar (based on the type of the sample). To prevent cross contamination**  
201 **between the sampling locations, cleaning of the vacuum cleaner was performed before and after**  
202 **each sampling event (at each location) by using a detergent, warm water, methanol and air-drying.**

203 Matrix spikes were prepared by spiking ground sea sand (1 g) contained in a thimble, dust bag or  
204 glass jar with 15 µL of a solution composed of all the target analytes at a concentration of 2.0  
205 ng/µL in nonane, and performing the same analytical steps as the samples. All the LDPE were  
206 deployed and analyzed in duplicates. For the dust samples, 30 % of the total number of samples  
207 were analyzed in duplicates. Instrumental calibrations were checked by injection of the  
208 continuing calibration solution. The GC/MSMS calibration was verified before, during, and after  
209 each analytical sequence. Three calibration standards were continuously injected every 10 dust  
210 samples and 15 LDPE samples and the calibration check was maintained within ±15% for all  
211 analytes of interest. When an analyte was not detected, the noise was quantitated for the limit of  
212 detection (LOD) calculations. LODs were determined in the different matrices as the upper limit  
213 of the 95% confidence interval for the field blanks (Table A3). We used ½ the LOD for  
214 concentrations that were below the detection limit. Recoveries of the surrogate standards  
215 generally ranged from 74 - 92% and 71 – 96 % for the outdoor and indoor LDPE respectively, 63  
216 – 86 % for the outdoor dust, 64 – 91 % for the indoor fine dust and 67 – 96 % for the floor dust.  
217 Matrix spikes recoveries (n = 5 for LDPEs and 6 for dust) ranged from 82 % (BDE 2) to 102 %  
218 (BDE 154) for the LDPE and from 76 % (BDE-2) to 103 % (BDE-100) for the dust (Table A4)  
219 with a relative standard deviation < 20%. Results of the replicate analysis of LDPE and the dust  
220 samples indicated that the reproducibility of the analysis ranged from 17.6 % – 25.3 %.

221

#### 222 **2.4. Human health risk assessment**

223 To calculate the daily exposure doses from inhalation, ingestion and dermal exposure pathways,  
224 the following models were used according to the USPEA risk assessment guidelines (Means,  
225 1989):

226 
$$DD_{inh} = \frac{C_g \cdot ET \cdot InhR}{BW} \longrightarrow 1$$

227 
$$DD_{ing} = \frac{C_{EFD} \cdot ET \cdot IngR}{BW} \longrightarrow 2$$

228 
$$DD_{der} = \frac{C_{EFD} \cdot ET \cdot SA \cdot AF_d \cdot ABS}{BW} \longrightarrow 3$$

229 Where,

230  $DD_{inh}$ ,  $DD_{ing}$  and  $DD_{der}$  are the daily dose associated with the inhalation, ingestion and dermal  
231 contact (all in ng/ kg/d) from the different micrenvironments,  $C_g$  = the vapor phase concentration  
232 of each individual flame retardant ( $ng/m^3$ ),  $ET$  = the exposure time (h/d),  $InhR$  = inhalation rate  
233 ( $m^3/d$ ),  $BW$  = body weight (kg),  $C_{EFD}$  = the concentration of each flame retardant in the elevated  
234 fine dust ( $ng/g$ ),  $IngR$  = ingestion rate ( $mg/d$ ),  $ABS$  = dermal adsorption fraction  
235 (dimensionless),  $SA$  = skin surface area exposed ( $cm^2$ ),  $AF_d$  = particle-to-skin adherence factor  
236 ( $g/cm^2/event$ ).. Note that  $DD_{inh}$ ,  $DD_{ing}$  and  $DD_{der}$  are the sum of inhalation, ingestion or dermal  
237 contact of vapor phase and elevated fine dust from homes, working places (for adults only), cars  
238 and schools.

239  
240 The  $ET$  was obtained from the answers of the participants in the questionnaire. The average  
241 working hours were set as 8.0 and 6.0 hours/day for males and females respectively. For  
242 housewives, no working hours were included. For all adults and children,  $ET$  in cars was set at 2  
243 hours/day. The remainder was the time spent at home. For toddlers, all the time was spent at  
244 home.  $DW$  was 70, 15 and 12 kg for adults, children and toddlers respectively (Means, 1989).  
245 Selected inhalation rates were 13.3 and 10.9  $m^3/day$  for adults and children (toddlers)  
246 respectively (Means, 1989). In the best-case scenario, ingestion rates were set at 0.02 and 0.05  
247 d/day for adults and children, whereas they were set at 0.05, 0.1 and 0.2 g/d for adults, children

248 and toddlers respectively (Means, 1989). For the dermal exposure pathway, SA used were 8,620  
249 and 4,970 cm<sup>2</sup> for adults and children (Means, 1989), AF<sub>d</sub> (Means, 1989) was 0.000096 g/cm<sup>2</sup>  
250 and ABS was 0.03 (Cequier et al., 2014). Available reference dose values (RfD, mg/kg body  
251 weight/day) for BDE-47, 99, 153 and HBB were obtained from the Integrated Risk Assessment  
252 Information System (IRIS) of the US EPA ( <https://www.epa.gov/iris>, accessed in 10/01/2017).  
253 For BTBPE, RfD was obtained from Ali et al. (2012).

## 254 **2.5. Statistical analysis**

255 Analysis of variance (ANOVA) and the student t-test were performed with SigmaPlot 11. Before  
256 applying the statistical tests, all numbers were log transformed for normality. Bivariate  
257 correlation was performed with IBM SPSS (V 23, USA).

## 258 **3. Results and discussion**

### 259 **3.1. Indoor concentrations**

260 **3.1.1. PBDEs:**  $\Sigma$ 12 PBDE concentrations in the vapor phase ranged from 7.0 - 300 pg/m<sup>3</sup>, 36 –  
261 220 pg/m<sup>3</sup> and 13 – 303 pg/m<sup>3</sup> at apartments, working places and cars respectively (Table 1). In  
262 the EFD, concentrations ranged from 4.10 – 710, 80 – 540 and 3.0 – 770 ng/g respectively  
263 (Table 2). Concentrations of PBDEs (2.0 - 78 ng/g) in floor dust (Table A5) were lower than in  
264 EFD by a factor of 7.0 – 14 as was previously observed by Bjorklund et al. (2012) in Sweden.  
265 Working places (120 pg/m<sup>3</sup> compared to 66 – 85 pg/m<sup>3</sup> at the other microenvironments) and cars  
266 (210 ng/g compared to 129 – 131 ng/g for the other microenvironments) showed the highest  
267 median PBDE concentrations in the vapor phase and EFD samples respectively as was  
268 previously observed in the vapor phase samples of Sweden (Thuresson et al., 2012), and dust  
269 samples in Cairo, Egypt (Hassan and Shoeib, 2015), and in UK (Stuart et al., 2008). Vapor phase  
270 concentrations of BDE-2, 8 and 30 were significantly higher (One-Way Repeated Measures of  
271

272 ANOVA;  $p = 0.004 - 0.025$ ) at apartments probably due to limited ventilation at apartments  
273 compared to working places and cars.

274 Detected median vapor phase and EFD concentrations of  $\sum_{12}$ PBDE and the individual congeners  
275 were generally within the same range or lower than concentrations previously reported  
276 worldwide at working places and apartments/houses (Figure 1a,b and Table A6). In contrast,  
277 PBDE concentrations in the EFD samples of the cars were much higher than concentrations  
278 previously observed in Asia and in Cairo, Egypt (Figure 1c). PBDE concentrations in the FD  
279 samples were lower than previously detected concentrations in Europe, Asia and USA (Figure  
280 1d) but close to what was previously detected in Cairo, Egypt (Hassan and Shoeib, 2015).  
281 However, it should be pointed out that the vapor phase sampling technique used in our study  
282 (LDPE passive samplers) is different from the active or passive sampling techniques (using  
283 PUFs) used in the other studies and this should be considered when comparing the results.  
284 Additionally, differences in the sampling periods, sampled cars (manufacturer, year and  
285 mileage), and conditions of the investigated apartments/houses are all significant factors and  
286 should be considered in this comparison. Accordingly, we tried to limit our comparisons (Figure  
287 1) to studies with very close sampling periods whenever possible.

288  
289 BDE-47 and BDE-99 were the dominant congeners in the vapor phase, EFD and FD samples  
290 (Figure 2) comprising on average 46 - 84% of the total concentrations. In the EFD samples,  
291 BDE-99 was the dominant congener compared to BDE-47 in the vapor phase and FD samples.  
292 The ratio of BDE-47 to BDE-99 for the vapor phase (1.0 – 1.7) and FD samples (1.3 – 1.9) were  
293 slightly higher than values observed for the penta-formulations (0.6 and 1.0, La Guardia et al.,  
294 2006), whereas values in the EFD samples were consistent with the penta-formulation (0.4 –  
295 0.9), which is probably attributed to the slightly higher volatility of BDE-47 (log vapor pressure

296 = -3.61 pa) compared to BDE-99 (log vapor pressure = -4.30 pa) (Yue and Li, 2013). Our  
297 findings indicate the possible wide usage of the penta formulation in the imported consumer  
298 goods to Alexandria. Additionally, differences in the ratio values between EFD and FD samples  
299 suggest a different PBDE sources. BDE-183 (octa- formulation) showed minor contributions (<  
300 10 %) (Figure 2) indicating probably a limited usage of the octa-BDE formulation, in-line with  
301 results reported for floor dust in Cairo (Hassan and Shoeib, 2015).

302  
303 **3.1.2. NFRs:** Concentrations of NFRs ranged from 0.40 to 47 pg/m<sup>3</sup>, 0.60 – 10 ng/g and 0.50 –  
304 2.0 ng/g in the vapor phase, EFD ad floor dust samples respectively (Tables 1, 2 and A5). HBB  
305 displayed the highest concentrations (Figure 3) in the vapor phase samples, as was previously  
306 observed in Norwegian schools (Cequier et al., 2014). In all the microenvironments, PBEB, PBT  
307 and TBX were less abundant than HBB but showed significant contributions in the vapor phase  
308 (Figure 3a, c, f). BTBPE, DBHCTD, a-DDC-CO and s-DDC-CO were below LOD in all the  
309 samples probably due to their low vapor pressure (vapor pressure = -5.5 to -10.1 Pa)  
310 (McDonough et al., 2016) and accordingly, their preferential occurrence in the particulate phase  
311 as was observed in the current study (Figure 3b, d, f, g). Higher contribution of s-DDC-CO in the  
312 EFD of cars and working places was observed in the current study probably due to the usage of  
313 the technical DDC-CO as an additive in textile, automotive and in computers (Hassan and  
314 Shoeib, 2015).

315  
316 Detected concentrations of NFRs in air and dust of the microenvironments were much lower than  
317 concentrations previously recorded worldwide (Table A7). However, EFD median  
318 concentrations of HBB (0.25 ng/g), BTBPE (1.0 ng/g), s-DDC-CO (2.0 ng/g) and a-DDC-CO

319 (0.9 ng/g) in cars of Alexandria were comparable with those observed in Cairo, Egypt (0.23, 1.3,  
320 1.5 and 1.0 respectively) (Hassan and Shoeib, 2015). Additionally, median concentrations of  
321 HBB and BTBPE were very close to values reported for the FD samples of Cairo, Egypt (Hassan  
322 and Shoeib, 2015), whereas, the median concentrations of s- and a-DDC-CO in Cairo were 3.0  
323 and 7.0 fold respectively higher than our reported values. Egypt does not manufacture flame  
324 retardants and any detected compounds are due to imported goods. Nevertheless, the lower  
325 observed concentrations in the current studies could be the reason of different materials  
326 imported, different manufacturing countries, ages and types of materials and electrical equipment  
327 used in the different studies, type, age, mileage and manufacturer of the vehicles and/or less strict  
328 fire safety standards in Egypt.

329

## 330 **3.2. Outdoor concentrations**

331 **3.2.1. PBDEs:** PBDE concentrations ranged from 3.0 - 41pg/m<sup>3</sup> and 1.5 - 200 ng/g in the vapor  
332 phase and dust samples respectively (Table 3). BDE-2 was below the LOD in all the outdoor  
333 samples. Vapor phase and dust concentrations of  $\sum_{12}$ PBDE, BDE-47, 49, 99, 100, 153 and 154  
334 were significantly higher (t-test,  $p < 0.05$ ) in the residential – industrial regions. This is probably  
335 attributed to the older age of the industrial buildings, ventilation systems and/or the number of  
336 items containing PBDEs in the industrial buildings. In all samples (Figures A3, A4), BDE-47  
337 and BDE-99 dominated the profiles of vapor phase and dust samples in the outdoor environment  
338 as was previously observed for the indoor microenvironments (Figure 2). However, in the  
339 outdoor dust samples, BDE-47 showed higher concentrations than BDE-99 as was observed for  
340 the indoor floor dust samples (Figure 2g). Values of BDE-47/BDE-99 ratio (1.0 – 2.4) at all the  
341 locations were slightly higher than values of the penta-BDE formulations (La Guardia et al.,  
342 2006), EFD samples (0.4 – 0.9), but consistent with the FD samples (1.3 – 1.9) and the outdoor

343 gaseous samples (1.0 – 1.9). The ratio values observed here were probably the result of the faster  
344 photodegradation of BDE-99 compared to BDE-47 (Dickhut et al., 2012). BDE- 28, 100 and 153  
345 showed also significant contributions in the outdoor dust samples (8.0 – 15 % of the total  
346 concentrations).

347  
348 Detected vapor phase concentrations in the current study were much lower than previously  
349 detected concentrations in USA (Batterman et al., 2009), China (Hong et al., 2016) and Turkey  
350 (Kurt-Karakus et al., 2017) (Table A6) but higher than values previously observed for air around  
351 the lower Great Lakes (0.10 – 14.5 pg/m<sup>3</sup>) (McDonough et al., 2016) and Ottawa, Canada  
352 (<LOD – 4.4 pg/m<sup>3</sup>) (Wilford et al., 2004).

353  
354 **3.2.2. NFRs:** NFRs concentrations ranged from <LOD - 13 pg/m<sup>3</sup> and 0.40 – 4.0 ng/g in the  
355 outdoor vapor and dust samples respectively (Table 3), with no significant difference between  
356 the residential and residential-industrial areas as was previously observed for PBDEs.  
357 Concentrations of all the investigated NFRs were much lower than the indoor  
358 microenvironments (Table 2), and were also lower than PBDE concentrations (Table 3). Profiles  
359 of NFRs in the outdoor environment (Figures A5, A6) were similar to the profiles of NFRs in the  
360 vapor phase (dominance of HBB, PBB, PBEB and PBT) and dust samples (dominance of  
361 BTBPE, s-DDC-Co and a-DDC-CO) of the indoor microenvironments.

362  
363 Detected concentrations of PBB, HBB, PBT and PBEB in the current study were much lower  
364 than concentrations previously observed in China (Table A7) (Hong et al., 2016; Lin et al.,  
365 2013), Stockholm, Sweden (for PBT: <LOD in the current study compared to 0.17 pg/m<sup>3</sup>)  
366 (Newton et al., 2015), Istanbul, Turkey (for HBB and BTBPE; Table A7) and Great Lakes (for

367 BTBPE, DBHCTD, s-DDC-CO and a-DDC-CO) (McDonough et al., 2016). In contrast, the  
368 median concentration of HBB in the current study (0.10 pg/m<sup>3</sup>) was higher than that in Sweden  
369 (<LOD), and the range of PBB, HBB and PBEB in the current study was also higher than  
370 concentrations observed at the lower Great Lakes. Again, much lower concentrations of **NFRs**  
371 demonstrate that these compounds have not (yet) replaced PBDEs as flame retardants in  
372 Alexandria.

373  
374  
375

### 3.3. Indoor vs Outdoor Organohalogenated Flame Retardants

376 The lack of correlations between detected vapor concentrations and their subcooled vapor  
377 pressures ( $P_L$ , Pa) imply that emissions of **PBDEs and NFRs** were dominated by various source  
378 strengths, reactivity but not their volatility. All the gaseous and EFD concentrations of  
379  $\sum_{12}$ PBDEs and  $\sum$ **NFRs** in the indoor samples (excluding cars) were significantly higher (t-test,  $t$   
380 = 2.95 – 7.13,  $p < 0.006$ ) than the outdoor samples (**Figure A7**). This, in addition to the observed  
381 indoor - outdoor pattern variabilities and the between-samples variability in the indoor  
382 microenvironments suggests that detected indoor concentrations are mainly from indoor  
383 emissions with minimum contribution from the outdoor environment.

384 In the current study, several findings were observed when we compare indoor and outdoor dust.  
385 First, different patterns of PBDE congeners (**Figure A7c**) were observed in the elevated fine  
386 versus floor dust samples (dominance of BDE-47 in FD compared to BDE-99 in the EFD),  
387 whereas for the **NFRs** (**Figure A7d**) similar patterns were observed in the EFD, FD and outdoor  
388 dust samples. Second, greater similarities in concentrations and profiles (**Figure A7c, d**) of  
389 PBDEs and **NFRs** were observed between floor and outdoor dust (dominance of BDE-47,  
390 BTBPE, HBB and s-DDC-CO and similar BDE-47/99 ratios). This finding is unusual

391 considering the higher expected concentrations in the indoor samples, which is already observed  
392 for the gaseous and the elevated fine dust. Additionally, FD concentrations of HBB, BTBPE,  
393 DBHCTD and BDE-28 to 183 were more strongly correlated with the outdoor samples ( $r = 0.74$   
394  $- 0.99$ ,  $p < 0.05$ ) than with elevated fine dust samples ( $r = -0.08 - 0.90$ ), which were not  
395 significant ( $p > 0.05$ ) in most cases. The exception were a-and s-DDC-CO, which displayed  
396 stronger correlation to elevated fine dust samples ( $r = 0.80, 0.87$  compared to  $r = 0.67$  and  $0.68$   
397 for outdoor samples), although they were statistically insignificant ( $p > 0.05$ ). Third, elevated  
398 fine dust samples displayed much higher concentrations than floor and outdoor dust and slightly  
399 different profiles. Since all samples were sieved, we argue that different sources are responsible  
400 for the detected levels of PBDEs and NFRs in the EFD and FD. Floor dust is possibly  
401 contaminated by outdoor dust and dirt carried indoors by shoes, whereas this process is of  
402 minimum effect for the elevated fine dust (based on our requested pre-sampling and during-  
403 sampling ventilation conditions from volunteers).

#### 404 405 **3.4. Sources and relations between organohalogenated flame retardants in the samples**

##### 406 **3.4.1. Indoor**

407 To study the relations between detected flame retardants, a bivariate analysis was performed  
408 (correlations). Factors such as year of construction, number of foamed furniture, number of  
409 electronics, type of ventilation used and the number of vacuuming per week were included  
410 (Table A1). For cars, the make, year, type of seats and the ventilation system were included. All  
411 this information was obtained from the questionnaires given to the volunteers. For the indoor air  
412 (Table A8) and EFD (Table A9) samples, significant correlation between the tri- through hepta-  
413 brominated congeners (BDE- 28 to BDE- 183) was observed indicating possible common  
414 sources, the banned penta- and octa- formulations. Although a significant correlation was

415 observed between all tri- through hepta- brominated congeners, we excluded the possibility of  
416 occurrence of debromination and conversion of the higher brominated congener (BDE-183) to  
417 lower brominated ones as this finding occurred both in the indoor and outdoor environments (see  
418 below). No significant correlations were observed between PBDE congeners in the gaseous and  
419 EFD samples and the number of electronics and the number of foamed furniture. However, some  
420 within variability was observed at working places. Concentration of PBDE congeners at the car  
421 upholstery shop ( $\sum 12$  PBDE = 220  $\text{pg}/\text{m}^3$ ) was significantly higher (One-Way Repeated  
422 Measures of ANOVA;  $p < 0.001$ ) than those recorded for the four sampled school offices ( $\sum 12$   
423 PBDE = 36 - 120  $\text{pg}/\text{m}^3$ ).

424 In the elevated fine dust samples (Table A9), all the PBDE congeners were negatively correlated  
425 with the construction year ( $r = 0.43 - 0.62$ ,  $p < 0.05$ ), probably due to the increased dependence  
426 on **NFRs** and OPFRs in recent years. Nevertheless, the majority of the investigated **NFRs**  
427 showed no significant correlations with the construction year. Most of the PBDE congeners  
428 showed significant correlations with the floor type (Table A9), where concentrations increased  
429 when floors were covered with carpets, probably relating to the use of PBDEs in carpet paddings  
430 (Stapleton et al., 2008).

431  
432 For **NFRs**, only PBEB, s- and **a-DDC-CO** in the EFD samples showed significant correlations ( $r$   
433 = 0.47 - 0.68,  $p < 0.05$ ) with the number of electronics, probably due to their wide usage in  
434 electronics (Sverko et al., 2011). Although BTBPE was used as a replacement for the octa-BDE  
435 formulation by the end of 2004 (Kurt-Karakus et al., 2017), no correlation were observed with  
436 BDE-183 in the current study. Additionally, median concentrations of BTBPE in EFD of the  
437 indoor microenvironments (0.80 - 3.0  $\text{ng}/\text{g}$ ) and the outdoor dust (0.20 - 0.26  $\text{ng}/\text{g}$ ) were much  
438 lower than median concentrations of BDE-183 (2.2 - 25  $\text{ng}/\text{g}$  and 0.50 - 2.50  $\text{ng}/\text{g}$  respectively),

439 which may be related to the limited usage of the octa-PBDE formulation in the imported goods  
440 in Egypt.

441 The detected concentrations of **NFRs** in the current study are more affected by the within-site  
442 variabilities and thus sources were not clearly identified for this class of flame-retardants. A  
443 similar finding was observed in Norway (Cequier et al., 2014).

444  
445 As for cars, manufacturer was the key factor for the gaseous **organohalogenated flame retardants**  
446 (Table A14), where **vapor phase** concentrations of BDE-28 through BDE-154 in the Korean cars  
447 were significantly higher (One-Way Repeated Measures of ANOVA;  $p < 0.004$ ) than the  
448 German and Japanese cars sampled here (Table 1). Higher concentrations of the same congeners  
449 were also observed in the EFD samples collected from the Korean cars compared to the German  
450 and Japanese cars but was statistically insignificant (Table 2). Like the apartments and working  
451 places, several significant positive correlations were observed for the different **NFRs** (Tables  
452 A10, A11) **indicating** common sources. Additionally, the significant correlations between the  
453 individual penta- BDE congeners were also observed for the air and dust in cars. HBB in air and  
454 dust showed significant positive correlation with the model year ( $r = 0.677 - 0.74$ ,  $p < 0.05$ ),  
455 whereas DBHCTD and a-DDC-CO in the dust samples were significantly higher ( $r = 0.58 -$   
456  $0.85$ ,  $p < 0.05$ ) in the Korean cars.

#### 457 458 **3.4.2. Outdoor**

459 No correlations were found with temperature, wind speed or wind direction (data not shown).  
460 Generally, some significant between-analyte correlations were observed for **NFRs** and for the  
461 individual penta-BDE congeners and BDE-183. As we mentioned earlier, industrial activities  
462 were only a major source of PBDEs (significantly higher concentrations at the residential –

463 industrial areas than the residential areas) in the outdoor environment, whereas this pattern was  
464 not observed for NFRs.

#### 465 466 **3.4. Partitioning of the Organohalogenated Flame Retardants in Air and Dust**

467 To investigate the relationship between the detected concentrations in the dust and air samples,  
468 dust-air partitioning coefficients ( $K_{\text{dust-air}}$ ;  $\text{m}^3/\text{g}$ ) were calculated [concentrations in the dust  
469 ( $\text{ng}/\text{g}$ )/air concentrations ( $\text{ng}/\text{m}^3$ )]. For equilibrium conditions,  $K_{\text{dust-air}}$  is expected to be directly  
470 proportional to the octanol-air partition coefficient ( $K_{\text{OA}}$ ) (Cequier et al., 2014; Liagkouridis et  
471 al., 2017; Weschler and Nazaroff, 2010). In contrast, if the gaseous fraction was freshly released,  
472 or if the flame retardants in the air and dust were emitted from different sources, this relationship  
473 would be insignificant. Calculated  $\log K_{\text{dust-air}}$  values were compared with a theoretical  $K_{\text{dust-air}}$   
474 calculated based on  $K_{\text{OA}}$ , fraction of organic carbon ( $f_{\text{OC}}$ ) and density of dust ( $\rho_{\text{dust}}$ ,  $\text{g}/\text{m}^3$ ) as  
475 follows:

$$476 \quad K_{\text{dust-air}} = (f_{\text{OC}} \cdot K_{\text{OA}}) / \rho_{\text{dust}} \quad (4).$$

477  $f_{\text{OC}}$  and  $\rho_{\text{dust}}$  were selected as 0.2 and  $17 \times 10^5 \text{ g}/\text{m}^3$  (Liagkouridis et al., 2017). Thus, the less  
478 volatile flame retardants (higher  $K_{\text{OA}}$ ) will partition preferentially into the dust, thus resulting in  
479 higher  $K_{\text{dust-air}}$ .

480 In the current study, significant log-linear relationships between the calculated and theoretical  
481  $K_{\text{dust-air}}$  ( $R^2 = 0.71 - 0.82$  for EFD and FD, and  $0.73 - 0.81$  for the outdoor samples,  $p < 0.001$ )  
482 were observed for elevated fine and floor dust (Figure 4a) and the outdoor samples (Figure 4b).  
483 Additionally, slopes of the regression relationships ( $b = 0.53 - 0.65$ ) were significantly different  
484 from zero ( $p < 0.001$ ) indicating equilibrium distribution of the flame retardants between dust  
485 and vapor. Surprisingly, this relationship explained 75 % of the total variability for the elevated  
486 fine and floor dust samples, and the slopes were identical ( $b = 0.60$ ) and insignificantly different

487 from one ( $p < 0.001$ ). Assuming that floor dust was contaminated by outdoor dust, a stronger  
488 relationship for EFD samples is expected. However, this was not observed probably due to the  
489 similarity between the profiles of PBDEs and NFRs in the vapor phase samples of the indoor and  
490 outdoor environments (Figure 2a, b), the similarities between profiles of NFRs in the EFD and  
491 FD samples and/or the possible ongoing emission of flame retardants in the indoor  
492 microenvironment, which affected the strength of the relationship.

493

### 494 **3.5. Human Exposure Assessment**

495 Daily intake of the flame retardants from dust ingestion (based on EFD samples), air inhalation  
496 (vapor phase concentrations) and dermal absorption was estimated in the current study for adult  
497 males and females (working and housewives), children (age more than 6 years) and toddlers.  
498 Calculated daily intakes are given in Table A12. Values generally ranged from  $6.8 \times 10^{-6} - 0.17$ ,  
499  $2.0 \times 10^{-5} - 0.60$ ,  $7.0 \times 10^{-8} - 0.03$  and  $1.0 \times 10^{-5} - 0.04$  ng/kg body weight/day for the ingestion  
500 pathway (best case and worst-case scenarios respectively), inhalation and dermal exposure  
501 pathways. Calculated daily dose for the three exposure routes were generally higher for PBDEs  
502 than NFRs (Figure 5). Toddlers showed the highest ingestion ( $2.9 \times 10^{-3} - 1.0 \times 10^{-2}$ ), inhalation  
503 ( $1.4 \times 10^{-3} - 5.5 \times 10^{-3}$ ) and dermal ( $2.1 \times 10^{-4} - 1.3 \times 10^{-3}$ ) daily dose values compared to the  
504 other groups. Ingestion of NFRs was the most important exposure route (higher calculated daily  
505 dose values; Table A12) for toddlers, children, adult males and housewives (Figure 5a, 6), and  
506 for all the groups with respect to PBDEs. In contrast, inhalation (especially of HBB and TBX)  
507 was more important for adult working females. For most of the NFRs, comparable daily dose  
508 values (from all exposure routes) were observed for adult males and working females, which  
509 were higher than values calculated for housewives (Figure 5a, c, e). This observed pattern is  
510 probably attributed to the higher observed concentrations of most of the NFRs at the working

511 places. In contrast, the pattern was not observed for PBDEs (Figure 5b, d, f). Generally,  
512 inhalation pathway was more important for the more volatile flame retardants (TBX, PBB, HBB,  
513 PBT, PBEB, BDE-2, 8 and 15), whereas the ingestion pathway was the most important exposure  
514 route for BTBPE, DBHCTD, s-DDC-CO, BDE-28, 47, 99, 100 and 153 (Figure 5). Nevertheless,  
515 calculated values for BDE-47, 99, 153 and HBB were at least three orders of magnitude lower  
516 than their corresponding oral reference doses (RfD) (Table A12).

517

## 518 **Conclusions**

519 To our knowledge, this is the first study reporting levels of PBDEs and NFRs in the air and dust  
520 samples from the indoor and outdoor environments in Alexandria, Egypt. Results show that  
521 PDBEs are present everywhere at elevated concentrations relative to NFRs targeted here. The  
522 exposure assessment study indicated that inhalation of volatile flame retardants is a more  
523 important exposure route for adults than dust ingestion. Accordingly, gaseous concentrations of  
524 flame retardants should be included in future studies. For that purpose, LDPE could be used as  
525 an effective indoor sampling technique in human exposure studies. Inclusion of the outdoor  
526 samples indicated that: (i) indoor flame retardants are mainly emitted from the existing indoor  
527 sources (higher concentrations in the indoor samples) and (ii) elevated fine dust samples are less  
528 influenced by contamination from outdoor dust compared to the floor dust samples. Thus,  
529 outdoor samples should be considered in future studies especially at locations lacking emission  
530 inventory database. Concentrations of flame retardants in the elevated fine dust were higher than  
531 those for the floor dust samples, which could result in underestimated exposures when relying on  
532 floor dust. Nevertheless, estimated daily doses from all exposure routes for some of the target  
533 analytes were much lower than available reference doses. However, the small number of floor

534 dust samples analyzed in the current study is limiting conclusions. Our results could represent an  
535 important baseline database for developing response actions to limit the exposure to the  
536 organohalogenated flame retardants in the atmospheric environment of Alexandria, Egypt

537

### 538 **Supplementary data**

539 Details on the sampling, chemical and statistical analysis, estimation of gaseous concentrations  
540 and human exposure assessment are given in Appendix A.

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