

Harmful flame retardant found in electronic cigarette aerosol

CHUNG, Shan Shan; Zheng, Jin Shu; Kwong, Antonio C.S.; Lai, Vienna W.Y.

Published in:
Journal of Cleaner Production

DOI:
[10.1016/j.jclepro.2017.09.286](https://doi.org/10.1016/j.jclepro.2017.09.286)

Published: 10/01/2018

[Link to publication](#)

Citation for published version (APA):
CHUNG, S. S., Zheng, J. S., Kwong, A. C. S., & Lai, V. W. Y. (2018). Harmful flame retardant found in electronic cigarette aerosol. *Journal of Cleaner Production*, 171, 10-16. <https://doi.org/10.1016/j.jclepro.2017.09.286>

General rights

Copyright and intellectual property rights for the publications made accessible in HKBU Scholars are retained by the authors and/or other copyright owners. In addition to the restrictions prescribed by the Copyright Ordinance of Hong Kong, all users and readers must also observe the following terms of use:

- Users may download and print one copy of any publication from HKBU Scholars for the purpose of private study or research
- Users cannot further distribute the material or use it for any profit-making activity or commercial gain
- To share publications in HKBU Scholars with others, users are welcome to freely distribute the permanent publication URLs

1 **Harmful flame retardant found in electronic cigarette aerosol**

2

3 Shan-Shan Chung,^a Jin-Shu Zheng,^a Antonio CS Kwong,^b Vienna WY Lai^b

4 ^aDepartment of Biology, Hong Kong Baptist University, Kowloon Tong, Hong Kong

5 ^bHong Kong Council on Smoking and Health, Wan Chai, Hong Kong

6

7 **Correspondence to**

8 Dr Chung Shan Shan, Department of Biology, Hong Kong Baptist University,

9 Kowloon Tong, Hong Kong. sschung@hkbu.edu.hk; tel: +852-34117741; fax:

10 +852-34117743

11

12 **Key Words**

13 electronic cigarette; brominated flame retardant; PBDEs.

14

15 **Word Count** 5250 words

16

17 **Harmful flame retardant found in electronic cigarette aerosol**

18 **Abstract**

19 Studies on the health impact of vaping so far have largely ignored the fact that
20 electronic cigarette (e-cigarette) is essentially an electronic product which is likely to
21 contain a group of endocrine disrupting flame retardants, namely, polybrominated
22 diphenyl ethers (PBDEs) as additives in the combustible components of the product.
23 Thus, the conclusion that e-cigarette is less harmful to health than tobacco smoking
24 may be based on incomplete information. This study reports moderate to elevated
25 levels of PBDEs in 5 out of the 13 samples of e-cigarettes. This finding is suggestive
26 of the continued use of PBDEs in the manufacturing of e-cigarette atomizers and the
27 associated protective casing. This study is unique as it confirms the existence of this
28 developmental neurotoxicant in e-cigarette aerosols. In view of the significant levels
29 of PBDEs and other known carcinogens (polycyclic aromatic hydrocarbons and
30 formaldehydes) in e-cigarette aerosol, there is an urgent need for conducting a
31 thorough review of the health risks of vaping by relevant professionals. A further
32 lesson learnt from this study is that policy makers and relevant product manufacturers
33 should be aware of the possible presence of PBDEs in the aerosol of body care and
34 medical electrical devices such as face steamers, inhalators and nebulizers, especially
35 when PBDEs are utilized in the combustible components of these devices.

36 **Harmful flame retardant found in electronic cigarette aerosol**

37 **1. Introduction**

38 An electronic cigarette (e-cigarette) is a battery driven device mimicking tobacco
39 cigarettes and is being marketed as a less harmful alternative to tobacco cigarette or as
40 an aid for smoking cessation (Pearson, et al., 2012). Other than the lithium-ion battery,
41 an e-cigarette comprises of a light-emitting diode light, an atomizer, a microprocessor,
42 and a cartridge containing a liquid solution generally referred to as e-liquid (Lerner, et
43 al., 2015). The atomizer assembly mounting base is usually made of rigid plastics
44 containing holes for housing wires extending through the base (Politics and
45 Government Week, 2016). During use, the battery heats up the liquid in the cartridge
46 while the atomizer vaporizes the liquid, emitting it as a mist or aerosol that users
47 inhale. An e-cigarette is essentially an electronic product that is designed to generate
48 aerosols that are directly inhaled by vapers (smokers of e-cigarettes).

49 Though there is a lack of complete statistics on global e-cigarette consumption, it
50 may be noted that e-cigarette use is growing or persisting across many countries. In
51 the US, the number of adult vapers doubled between 2010 and 2013 (King et al.,
52 2015). Dramatic increases in e-cigarette use among young people in the US have been
53 found since 2013 (US Department of Health and Human Services, 2016). In EU,
54 though the percentage of vaper has remained at 2%, 15% of the population has tried

55 e-cigarette at some point (European Commission, 2017). The growth in the popularity
56 of e-cigarettes is presenting two major research challenges. The first challenge is
57 about the health risk of vapers and the associated indirect aerosol receivers. The
58 second is about the environmental impact of e-cigarette manufacturing and disposal
59 (Lerner et al., 2015). So far, most of the e-cigarette research attention has been
60 directed towards the human health risks associated with vaping (Lerner, et al., 2015).
61 A growing body of literature directed toward comparing the health impacts of vaping
62 and smoking is also noted. One view is that there are health benefits in vaping as
63 opposed to smoking. The Royal College of Physicians (2016) offered the view that
64 vapers were more likely to successfully quit smoking and that e-cigarettes were
65 “popular with smokers and offer a viable harm-reduction option” owing to reduced
66 bodily absorption of benzene, tobacco-specific nitrosamines and PAHs in vaping than
67 tobacco smoking. Parker and Rayburn (2017) showed that the leachate from one type
68 of e-cigarette was about ten times less toxic on embryos than tobacco cigarette butts.
69 Also by testing one brand of e-cigarette, Azzopardi et al. (2016) demonstrated that
70 aerosol from e-cigarette was significantly less cytotoxic than cigarette smoke.
71 However, some studies showed that nicotine exposure of vapers was not significantly
72 different from smokers (Göney, et al., 2016) and the flavorings in e-liquid was
73 causing significant levels of aldehydes to be formed in e-cigarette aerosols (Khlystov

74 and Samburova, 2016).

75 Although there is no consensus as yet on the health risks of vaping, the World
76 Health Organization (WHO) has recommended that e-cigarettes should not be used in
77 work places or public areas in view of the harmful substances known to be emitted
78 with the aerosol (WHO, 2014). Despite this recommendation, controversies on the
79 merits and demerits of e-cigarettes continue. In line with the WHO recommendation,
80 the Hong Kong Special Administrative Region (HK) government has proposed,
81 among others, to ban the import, manufacture, sale, distribution and advertising of
82 e-cigarettes in the city. Despite the large number of studies on the health risk
83 associated with e-cigarette use, the potential toxicities of inhaling or absorbing the
84 substances and additives used in the e-liquid, atomizer and protective casing continue
85 to be incompletely understood owing to the lack of comprehensive knowledge and
86 evaluation of its benefits and harms. To provide objective scientific evidence for the
87 ban, the Hong Kong Council on Smoking and Health (COSH) commissioned a study
88 to test the concentrations of harmful substances in e-cigarettes available in HK in
89 2015-2016. The results reported in this paper form part of the COSH study.
90 Substances tested in the aerosols of e-cigarettes include, among others, polycyclic
91 aromatic hydrocarbons (PAHs), nicotine, formaldehydes and polybrominated diphenyl
92 ethers (PBDEs). This paper reports only on the concentrations of PBDEs in

93 e-cigarette aerosol because the concentrations of nicotine, PAHs and formaldehydes in
94 e-cigarette aerosols have already been reported comprehensively in previous studies
95 (e.g., Cheng, 2014).

96 PBDEs are flame retardants widely used in many products such as building
97 materials, textiles, cars and electronic and electrical equipment. They are applied to
98 combustible components of target products, usually plastics. PBDEs tend to be easily
99 leached out or carried away from the host products (Besis et al., 2014). Coupled with
100 the widespread use of PBDEs, this phenomenon makes contamination ubiquitous.
101 Though not all congeners of PBDEs are harmful to humans, at least one is an
102 endocrine disruptor (BDE-47) while others have been shown to cause cancer in high
103 doses. They exhibit developmental and reproductive toxicity and damage the central
104 nervous system (Schechter et al., 2006). As a result, PBDEs have been recognized as
105 hazardous substances and their use in electrical and electronic products is restricted in
106 the European Union (Directive 2011/65/EU). Besides, they have been listed as
107 persistent organic pollutants under the Stockholm Convention (the Secretariat of the
108 Stockholm Convention, 2009). PBDEs are bio-accumulative and can be biomagnified.
109 Although PBDEs have been found to be ubiquitous in food, Schechter et al. (2006)
110 argued that dietary exposure alone could not explain the high human body burden.
111 Other sources of exposure include ingestion of dust in workplaces and even

112 households as well as indoor and outdoor air inhalation (Ni et al., 2013).
113 Environmental exposure, resulting from the industrial application of PBDEs in
114 electronic and electrical products is another reason for the high body burden noted.
115 This paper aims to i) enrich the present knowledge on the health risks of vaping
116 versus smoking by presenting the levels of PBDEs in thirteen e-cigarette and two
117 tobacco cigarette samples and calculating the “safe” doses of e-cigarette with respect
118 to specific PBDE congeners; and ii) discuss the implications of such findings on
119 environmental sustainability, public health and clean production.

120

121 **2. Materials and methods**

122 *2.1. Samples and sampling*

123 A total of six different brands of e-cigarettes (A-F) were included in the study by
124 means of convenience and judgement sampling. A convenience sample is a sample
125 obtained by using convenience sampling method which is a type of non-probability
126 sampling. Samples are made up of easy-to-reach individuals. Judgement sampling is
127 also a type of non-probability sampling and selects samples based on expert (the
128 vendors) advice. Five brands of e-cigarettes and e-liquids were chosen based on
129 market observations at the time of the study and the results of a 2014 COSH study on
130 the promotion and availability of e-cigarettes in HK. One additional brand (A) was

131 also included based on a supplier's recommendation of its popularity. In total, a
 132 sample of thirteen e-cigarettes, four filled with e-liquids and nine connected with
 133 e-capsules from six brands were procured through online platforms and normal retail
 134 outlets in HK in February 2014 and in June to July 2015. As for the tobacco cigarette
 135 samples (RC1 and RC2), two packets of a common commercial brand (G) were
 136 acquired. Table 1 lists the types, flavors and nicotine information shown on the
 137 samples' labels.

138 **Table 1**

139 Nicotine and other information on the labels of e- and tobacco cigarette samples

Sample	Types	Brand	Flavors	Indication of nicotine levels on label
EC1	e-liquid	A	Tobacco	May contain nicotine
EC2		B	Mint	Nicotine (unless zero) ^a
EC3		C	Mint	No mention of nicotine
EC4		C	Cola	content
EC5	original e-cigarette	D	Cigarette brand flavor ^a	0 mg nicotine
EC6		D	Cigarette brand flavor ^a	
EC7		D	Strawberry	
EC8	cartridge	E	Tobacco	
EC9		E	Chocolate	
EC10		F	Tobacco	0 mg nicotine & tar
EC11		F	Mint	
EC12		E	Tobacco	
EC13		F	Mint	0 mg nicotine
RC1	Tobacco cigarette pack 1	G	n.a.	n.a.
RC2	Tobacco	G	n.a.	n.a.

140 ^a exact label of the e-liquid container/e-cigarette cartridge, unclear/not informative

141 *2.2. Experimental set up for testing e-cigarette and tobacco cigarette aerosol*

142 Our experimental set-up was similar to the Sparging Apparatus used by US Food
143 and Drug Administration (2009) for testing of e-cigarettes. It consisted of two 50 mL
144 polypropylene conical test tubes connected together with glass tubes and flexible
145 silica tubings which were further connected to a pump (Model DQA-P104-AA
146 Volts:115 Amps:4.2 HZ 60 USA connecting with CT-1000AC-AC Converter 1000
147 Watt 50/60 HZ) to suck air from the e- and tobacco cigarette samples. To set the
148 correct puff velocity, the researcher began with the lowest velocity and slowly raised
149 it to a level where it was strong enough to light the e-cigarette. Upon activation of the
150 pump, aerosol from the samples passed through two tubes of solvent mixture. A valid
151 “puff” is indicated by illumination of the LED indicator at the front of the e-cigarette
152 sample and bubbling of the solvent mixture. In each bout, a sample was “lit” for 12
153 min with each “puff” (by activating the pump) lasting about 4 s to 5 s with an
154 inter-puff time of 2 s. This is translated into about 111 puffs per bout. While the 12
155 min duration for a bout was generally longer than a normal smoking activity and the
156 puff regimes deployed in other e-cigarette studies, the intention was to maximize the
157 collection of target analytes so as to ensure that no harmful substances, not known to
158 the researchers, escape notice. A case in point is Khlystov and Samburova (2016) who

159 admitted that “the small number of puffs” in their test regime was likely the cause of
160 non-detection of target analytes in some samples. In their study, two puffs (each lasts
161 4 s) were sampled after 15 warm-up puffs (Khlystov and Samburova, 2016).
162 Goniewicz et al. (2014) similarly admitted that since their puff regime was likely to
163 be shorter (1.8 s) than actual situations, their findings might have understated actual
164 quantities of harmful substances inhaled by vapers. Hence, using a longer puff regime
165 (4.5 s) is a sensible approach in the present case. This approach is applied in Health
166 New Zealand Ltd. (2008) and Burstyn (2014) as well.

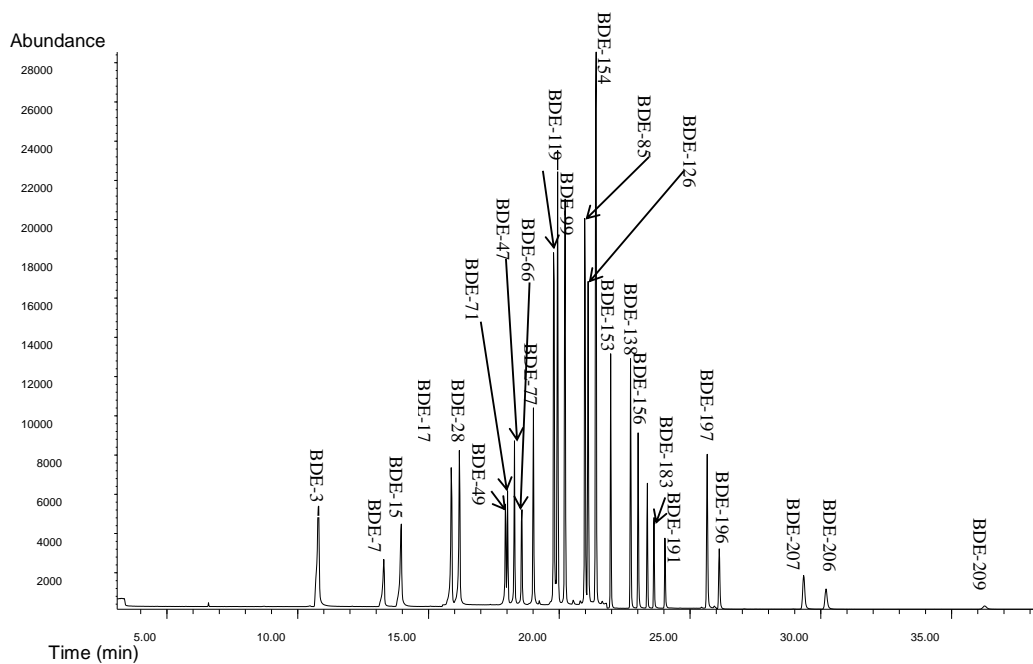
167 *2.3. Extraction and analysis of PBDEs*

168 The solvents used in the two polypropylene conical test tubes consisted of 12.5
169 mL of dichloromethane and 12.5 mL of hexane in each tube, i.e., 25 mL in each tube.
170 After 12 min of exposure to e- or tobacco cigarette aerosol, the solvents in the two
171 tubes were combined and concentrated by a rotary pressure reduced evaporator to
172 around one mL which was then transferred to specialized 1.5 mL vials for determining
173 PBDEs.

174 Subsequent analytical procedures used in this study were adopted from Liu et al.
175 (2005). The presence of PBDEs was detected by a gas chromatography-mass
176 spectrometer (GC-MS) from Agilent Technologies (7890A GC system) with a 7683B
177 series injector for sample injection. An Agilent Technologies 5975C inert MSD with

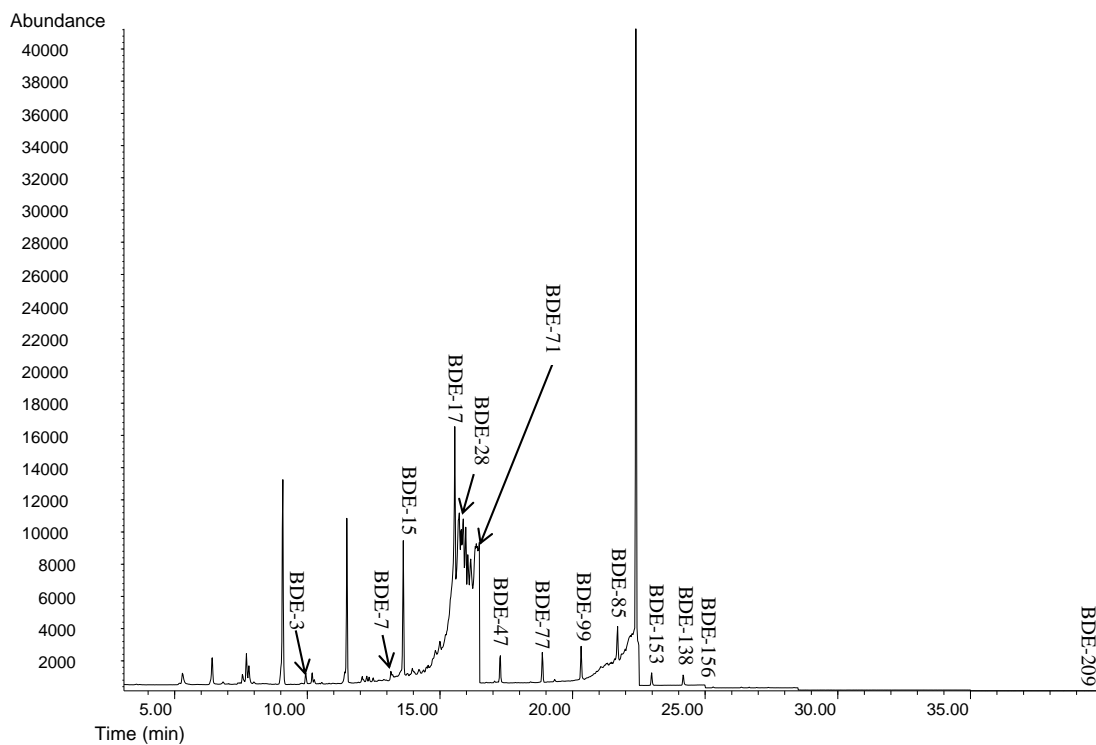
178 triple-axis detector was deployed for compound identification and quantity calculation.
179 A 30m HP-5 mass fused silica capillary column (0.25 mm diameter and 0.25 μm film
180 thickness; 95% dimethyl-polysiloxane) was used for the GC oven. The
181 chromatographic separation was achieved by setting the oven at 80°C for the first two
182 min. The temperature was then increased at a rate of 8°C/min to 280°C and was held
183 there for 15 min. Helium was used as the carrier gas at a flow rate of 1.9 mL/min. The
184 injector temperature was set at 280°C in the splitless-injection mode. The mass spectra
185 of targeted analytes were obtained in EI mode (70 eV). After scanning in the range of
186 50–800 m/z with a cycle time of 1s, the MSD could identify compounds by
187 comparing the m/z of unknown compounds against the default NIST-library database.
188 Finally the determination of PBDEs was confirmed by standards.

189 For quantitative analysis, a series of standard solutions at 8 different levels: 0, 5,
190 10, 20, 50, 100, 250 and 500 ng/mL were prepared and injected into the GC-MS for
191 producing the standard calculation curves (accepted $R^2=0.999$ or above). All
192 parameters obtained were then analyzed by the computer. The internal standard,
193 Mass-Labelled Polybrominated Diphenyl Ethers (MBDE-MXE)¹³ C-PBDE
194 (Wellington Laboratories Inc., 345 Dr. Guelph ON N1G 3M5, Canada), was spiked
195 into the samples and used with the standard calculation curves for quantitative
196 analysis. Fig. 1a and 1b show the chromatograms of the internal standard and EC6.



197

198 Fig. 1a Chromatogram of the internal standard



199

200 Fig. 1b Chromatogram of EC6

201 2.4. Quality control

202 The determination limits for PBDEs were set at 0.1 ng/mL for individual

203 congener and 1.0 ng/mL for total PBDEs (Table 3). Recovery was between 74-85%
 204 for PBDE congeners from BDEs 3 to 191, and 54%-75% for high molecular BDEs
 205 196, 197, 206, 207 and 209. Our recovery rates were on par with Liu et al. (2005) and
 206 Oros et al. (2005). Blank tests were also conducted with each e-cigarette aerosol test.
 207 Laboratory air was taken as a blank. Table 2 states the ion mass of the 27 PBDE
 208 congeners.

209 **Table 2**
 210 BDE – MXE selected ion mass of 27 PBDEs

Congeners	Ion	Congeners	Ion
BDE-3	248.0	BDE-126	565.7
7	168.1	154	483.7
15	327.9	153	483.7
17	246.0	138	483.7
28	245.9	156	483.7
49	325.9	184	563.7
71	325.9	183	561.7
47	325.9	191	563.7
66	325.9	197	641.7
77	485.7	196	641.7
100	403.8	207	719.5
119	403.8	206	719.5
99	405.8	209	799.5
85	405.8		

211

212 *2.5. Reference doses*

213 In line with other studies (e.g. Wang et al., 2011), the reference doses (RfDs)
 214 established by USEPA (2014) for some congeners of PBDEs were used in this study
 215 for comparison purposes (second last row of Table 3).

216 **3. Results**

217 *3.1. Congener Profiles*

218 The unadjusted mean concentrations from two replications of analyses of the 27
219 PBDE congeners, expressed as ng/mL of concentrated solvent, detected in the aerosol
220 of samples are stated in Table 3. Adjustment is not recommended because the
221 recovery rates achieved here are typical of PBDE determination. As all the PBDEs
222 from one bout are believed to be dissolved in the 25 mL solvent which was then
223 concentrated to one mL for determination, the concentrations of PBDEs found in each
224 sample of e-cigarette aerosol could also be understood as Σ_{27} PBDEs absorbable by a
225 vaper in one bout of e-cigarette smoking.

226 Based on the PBDEs concentration, the e-cigarette samples were grouped into
227 three tiers. Tier 1 samples, namely, EC1-3, 5, 8, 10, 12 and 13, had low to very low
228 levels of PBDEs despite the use of a more intense and long puff regime. Tier 2
229 samples (EC4, 7, 9 and 11) had moderate levels of PBDEs. EC6 had the highest
230 concentration of PBDEs, about 8-10 times Tier 2 samples and formed a tier (Tier 3)
231 on its own.

Table 3

Mean concentrations of PBDEs in the aerosol of e-cigarette and tobacco cigarette samples (ng/mL) (replicate = 2)

	BDE-47	BDE-85	BDE-99	BDE-153	BDE-209	Total (Σ_{27}) PBDEs mean (sd)
EC1	0.2	0.2	0.2	0.3	0.2	2.5 (0.28)
EC2	0.1	0.2	0.1	0.2	0.1	2.1 (0.07)
EC3	1.5	1.8	0.7	0.7	0.5	9.0 (0.28)
EC4	31.4	22.4	34.7	19.8	2.8	191.9 (0.07)
EC5	1.8	0.7	0.4	1.2	0.2	7.2 (0.57)
EC6	182.7	185.5	272.6	257.9	11.5	1490.0 (7.07)
EC7	14.9	24.3	19.7	17.8	7.4	149.3 (3.82)
EC8	8.4	7.7	9.1	6.8	2.4	46.8 (0.28)
EC9	23.5	21.9	21.5	16.3	3.6	145.7 (4.24)
EC10	1.1	0.4	0.5	0.5	0.3	3.5 (0.57)
EC11	22.7	18.6	19.4	21.6	11.6	158.1 (2.97)
EC12	0.4	0.1	0.0	0.9	0.1	2.3 (0.07)
EC13	0.5	0.2	0.3	0.0	0.0	1.7 (0.07)
RC1	1.4	0.7	0.8	0.9	0.0	6.3 (0.00)
RC2	1.2	0.5	0.4	0.6	0.1	5.6 (0.42)
Control	0.0	0.0	0.0	0.0	0.0	0.0 (0.00)
RfDs (mg/kg/d)	0.0001	na	0.0001	0.0002	0.007	na
Detection limit			0.1			1.0

Notes: sd, standard deviation; na, not available; RC, tobacco cigarette; EC, e-cigarette.

232 **4. Discussion**

233 The views on e-cigarette continue to be polarized with each side having its own
234 arguments. Soon after WHO's report, Financial Times reported that 53 public health
235 academics from 15 countries appealed to WHO to not regulate e-cigarette in the same
236 way as tobacco cigarettes since they saw e-cigarette as a solution to tobacco addiction
237 (Duncan, 2014) with the major argument being e-cigarettes tested so far were less
238 harmful than conventional cigarettes (McNeill et al., 2014). This was then followed
239 by the release of The Royal College of Physicians' report (2016).

240 As far as harm reduction is concerned, it appears that the e-cigarette industry has
241 significantly reduced the level of nicotine in e-cigarette (compare Table 1 with
242 Goniewicz et al., 2013) rendering vaping less addictive. However, in view of the
243 newly discovered group of manufacturing additive, PBDEs, and together with
244 Khlystov and Samburova's (2016) findings, it is no longer appropriate to encourage
245 its use even if it is later proven that e-cigarette is a solution to tobacco addiction.
246 Neither is it correct to argue that the levels of toxins from e-cigarettes are much lower
247 than tobacco cigarettes before a systematic and comprehensive study on the
248 concentrations of PBDEs, aldehydes (as revealed in Khlystov and Samburova, 2016)
249 and possibly other newly discovered harmful substances in e-cigarette aerosols has
250 been carried out.

251 While low levels of PBDEs were found in the aerosol of Tier 1 samples and
 252 tobacco cigarette (Table 3), moderate to high levels of PBDEs were found in Tiers 2
 253 and 3 e-cigarettes which were available from retail outlets in HK. As no datum
 254 regarding the level of PBDEs in e-cigarette aerosols was available from the existing
 255 body of literature for comparison, concentrations of PBDEs in food and various
 256 environmental settings were cited instead (Table 4). Although direct comparison is
 257 difficult owing to the diverse nature of data in Table 4, in general, it is reasonable to
 258 conclude that the levels of PBDEs in some of our samples were many orders of
 259 magnitude higher than those in ambient air and food. The levels were comparable to
 260 those in dust particles. Besides, people do not regularly ingest or inhale dust
 261 contaminated with such levels of PBDEs.

262 **Table 4**

263 Concentrations of PBDE in food and various environmental settings

Nature of sample	Total PBDEs	Year & place	Reference
Air conditioner filter dust	1271 ng/g	Greece, 2014	Besis et al. (2014)
Indoor dust in workplace	138-477 ng/g	Belgium, 2010	Pearson et al. (2012)
Outdoor air	195-1450 pg/m ³	Shunde, China, 2009	Besis and Samara (2012)
Indoor air (homes)	1.3-3980 pg/m ³	Various places, 2004-2011	Besis and Samara (2012)
Pork	41 pg/g wet weight		
Farmed Salmon fillet	1590-1919 pg/g wet weight	USA, 2004	Schechter et al., (2006)
Cow's milk	7.9 pg/g wet weight		
Various freshwater fish	1.4-5.3 ng/g wet weight	Hong Kong, 2011	Wang et al. (2011)
E-cigarette aerosol	1.7-1490 ng/mL	Hong Kong, 2015	This study.

264 Since PBDEs (mostly BDE-209) are only physically coated on the plastic

265 materials in an e-cigarette atomizer, they can easily migrate onto the aerosol and the
266 external environment. The use of PBDEs as a flame retardant in e-cigarette atomizer
267 and protective casing was suspected to be the main source of PBDEs in e-cigarette
268 aerosol for Tiers 2 and 3 samples.

269 Based on EPA's RfDs, using Tier 3 sample, EC6 as an example and assuming an
270 average human body weight of 60 kg, the "safe" doses for the PBDE congeners (see
271 second last row of Table 3) range from 22-47 bouts/d for BDE-47, -99 and -153 and
272 >36,500 bouts/d for BDE-209. Since EC6 has the highest PBDEs concentration
273 among all samples, it was concluded that sufficient evidence was not yet there to
274 show that the presence of PBDE in e-cigarette aerosols alone was enough to cause
275 long term health damages except among very avid vapers, i.e., those who vape more
276 than 22 bouts/d. Yet, recent preliminary study findings have shown that exposure to
277 aerosol from e-cigarettes drastically reduces sperm counts and sperm mobility in
278 juvenile mice and bring about genetic changes in the brain and alter behavior in adult
279 male and female mice (Zelikoff, 2016). With the confirmation that e-cigarette aerosol
280 also contained carcinogens such as formaldehydes and benzo(a)pyrene, the effect of
281 the additional harms from PBDEs for vapers should not be overlooked and the
282 toxicological implications of chemical mixtures in e-cigarette aerosol should be
283 studied further.

284 Given the cost-effectiveness of PBDEs as flame retardants, it would not be
285 realistic to expect the electronic and electrical products manufacturing industry to
286 totally stop their use in the near future. Yet, it is exigent that the electronic and
287 electrical industry should step up research to find a less toxic and yet cost-effective
288 substitute for PBDEs especially for electrical products or devices that generate aerosol
289 for direct human inhalation of which e-cigarette is one example. Other examples
290 include at least face steamers, nebulizers and inhalators. It is also vital that before a
291 substitute (of PBDEs) is used in production, thorough studies on how, how much and
292 in what form the chemical or additive concerned will migrate to the external
293 environment must be conducted. Information on the route of migration,
294 transformation pathways and concentrations at each stage of transformation of flame
295 retardant and/or its derivatives is essential for determining whether the use of such
296 flame retardants or additives are safe.

297 Since it has already been confirmed that e-cigarette aerosols may contain elevated
298 levels of PBDEs, more e-cigarette samples should be tested in future studies. Further,
299 the studies should adopt the international puff profile [e.g. ISO (2012)] to more
300 accurately represent human vaping behavior so that more realistic “safe” dose alerts
301 can be given to vapers.

302

303 **5. Limitations**

304 Despite the important discovery, there are limitations about this study. First, since
305 the e-cigarette samples and the control were not prepared on the same day, it is
306 possible that the quality of indoor air might have affected the results. However,
307 smoking and open burning are banned on all campuses of the University in which the
308 testing laboratory of this study was situated. As evident from the results of the
309 controls, it is believed that even if existing contamination in the air has affected the
310 results of this study, the extent is not significant. Second, the duration of each bout
311 and each puff was longer than usual, so the presently adopted bout regime may have
312 led to the occurrence of dry puffs which users avoid. Since the bout regime adopted
313 may not be realistic, the amount of PBDEs inhaled by vapers in a normal bout was
314 likely to be less than what was detected by the study. However, this does not refute the
315 presence of PBDEs in e-cigarette aerosols and the recovery rates of PBDEs range
316 between 54% and 85% only (even though these are typical recovery rates for PBDE
317 determination). The measured concentrations reported here are therefore lower than
318 what were actually present in the aerosols.

319 **6. Conclusions**

320 This study has confirmed that elevated levels of PBDEs, up to 1490 ng/bout, are
321 found in one brand of e-cigarette aerosols. Based on present findings, a person has to

322 take 22 bouts or more of such brand of e-cigarette per day to get exposed to unsafe
323 levels of PBDEs. When compared to the levels of aldehydes found (Khlystov and
324 Samburova, 2016) in flavored e-cigarette liquids with which one puff is enough to
325 take the vaper's exposure to unacceptably dangerous levels, it appears that the main
326 health concern from e-cigarette should not be PBDEs.

327 However, this study has provided several other important insights to
328 environmental sustainability, public health and clean production. This study has taken
329 the lead in alerting vapers that e-cigarette aerosols contain more harmful substances
330 than people think. Going beyond inhalation and incidental ingestion of dust and
331 consumption of normal food (Wang et al., 2013), this study fills the information gap
332 and shows that vaping is an emerging and possibly an important source of PBDEs in
333 human body. The discovery that elevated levels of PBDEs are found in e-cigarettes
334 (but not tobacco cigarettes) has additional implications. First, it provides new insights
335 into the debate between WHO and supporters of e-cigarettes, e.g. the Royal College
336 of Physicians, for the latter to re-visit their stance on e-cigarette and if e-cigarette
337 should be seen as a solution to tobacco addiction. In the interest of a complete picture
338 of the health hazards from vaping, future research studies on e-cigarette aerosols
339 should broaden the scope from focusing on the composition of e-liquid to materials
340 and additives used in e-cigarette atomizer, cartridge and even protective casing.

341 Second, this discovery is suggestive that commonly used manufacturing additives
342 such as PBDEs and possibly other undisclosed but hazardous components may exist
343 in e-cigarette aerosols as well as other electrical devices capable of generating
344 aerosols for direct human inhalation. Such devices possibly include but are not limited
345 to inhalators, face steamers and nebulizers. Exposure to PBDEs from vaping is
346 avoidable if e-cigarette manufacturers can adopt clean production principles and
347 eliminate as far as possible known hazardous substances such as PBDEs in their
348 products. This can be achieved by using less toxic flame retardants and have their
349 health risks duly assessed before extensive application so that unintended adverse
350 health and environmental impact from using these products can be much reduced.
351 Such a material change in manufacturing will also reduce the environmental loadings
352 of PBDEs in terms of industrial emission during manufacturing and end-of-life
353 e-cigarette waste. To this end, there is the obvious need for regulatory actions on the
354 manufacturing, storage, recycling and disposal of e-cigarette (Lerner, et al. 2015).

355 Third, tests should be performed on the aerosols used in other medical and beauty
356 electrical devices and determine whether PBDEs and/or other potentially hazardous
357 substances are also being generated from those devices. Appropriate regulatory action
358 should be taken on the raw materials and/or additives used in the manufacturing of

359 these electrical devices if significant levels of PBDEs and/or other potentially
360 hazardous substances are found.

361

362 **Acknowledgements**

363 This work was supported by the Council on Smoking and Health of Hong Kong.

364 The authors would like to thank Prof. Lam Tai Hing for the discussion and advice
365 during the research process.

366

367 **References**

368 Azzopardi, D., Patel, K., Jaunky, T., Santopietro, S., Camacho, O.M., McAughey, J.,

369 Gaça, M., 2016. Electronic cigarette aerosol induces significantly less

370 cytotoxicity than tobacco smoke. *Tox. Mech. Methods* 26, 477-491.

371 Besis, A., Katsoyiannis, A., Botsaropoulou, E., Samara, C., 2014. Concentrations of

372 polybrominated diphenyl ethers (PBDEs) in central air-conditioner filter dust and

373 relevance of non-dietary exposure in occupational indoor environments in Greece.

374 *Env. Pollut.* 188, 64-70.

375 Besis, A., Samara, C., 2012. Polybrominated diphenyl ethers (PBDEs) in the indoor

376 and outdoor environments – a review on occurrence and human exposure. *Env.*

377 *Pollut.* 169, 217-229.

378 Burstyn, I., 2014. Peering through the mist: systematic review of what the chemistry
379 of contaminants in electronic cigarettes tells us about health risk. BMC Public
380 Health 14(1),1-14

381 Cheng, T.R., 2014. Chemical evaluation of electronic cigarettes. Tob. Control 23,
382 ii11-ii17.

383 Directive 2011/65/EU of the European Parliament and of the Council of 8 June 2011
384 on the restriction of the use of certain hazardous substances in electrical and
385 electronic equipment, text with EEA relevance, OJ L 174, 1.7.2011, 88–110.
386 <http://eur-lex.europa.eu/legal-content/en/TXT/?uri=celex:32011L0065> (accessed
387 05.01.16).

388 Duncan, R., 2014. Academics urge WHO over e-cigarette regulations. Financial
389 Times May 29.
390 [http://www.ft.com/cms/s/0/73e3f97e-e655-11e3-bbf5-00144feabdc0.html#axzz4](http://www.ft.com/cms/s/0/73e3f97e-e655-11e3-bbf5-00144feabdc0.html#axzz40JInfQR9)
391 [0JInfQR9](http://www.ft.com/cms/s/0/73e3f97e-e655-11e3-bbf5-00144feabdc0.html#axzz40JInfQR9) (accessed 16.02.16)

392 European Commission, 2017. E-news: World No Tobacco Day: more than one in four
393 EU citizens still smoke.
394 [http://ec.europa.eu/newsroom/sante/newsletter-specific-archive-issue.cfm?newsle](http://ec.europa.eu/newsroom/sante/newsletter-specific-archive-issue.cfm?newsletter_service_id=327&newsletter_issue_id=3764&page=1&fullDate=Tue%2030%20May%202017&lang=default)
395 [tter_service_id=327&newsletter_issue_id=3764&page=1&fullDate=Tue%2030%](http://ec.europa.eu/newsroom/sante/newsletter-specific-archive-issue.cfm?newsletter_service_id=327&newsletter_issue_id=3764&page=1&fullDate=Tue%2030%20May%202017&lang=default)
396 [20May%202017&lang=default](http://ec.europa.eu/newsroom/sante/newsletter-specific-archive-issue.cfm?newsletter_service_id=327&newsletter_issue_id=3764&page=1&fullDate=Tue%2030%20May%202017&lang=default) (accessed 11.07.17)

397 Göney, G, Çok, İ., Tamer, U., Burgaz, S., Şengezer, T., 2016. Urinary cotinine levels
398 of electronic cigarette (e-cigarette) users. *Toxicol. Mech. Methods* 26, 441-445.

399 Goniewicz, M.L., Knysak, J., Gawron, M., Kosmider, L., Sobczak, A., Kurek, J.,
400 Prokopowicz, A., Jablonska-Czapla, M., Rosik-Dulewska, C., Havel, C., Jacob, P.,
401 Benowitz, N., 2014. Levels of selected carcinogens and toxicants in vapour from
402 electronic cigarettes. *Tob. Control* 23,133-9.

403 Goniewicz, M.L., Kuma, T., Gawron, M., Knysak, J., Kosmider, L., 2013. Nicotine
404 levels in electronic cigarettes. *Nicotine Tob. Res.*15, 158-66.

405 Health New Zealand Ltd. 2008. Safety Report on the Ruyan E-cigarette Cartridge and
406 Inhaled Aerosol. <http://www.healthnz.co.nz/RuyanCartridgeReport30-Oct-08.pdf>
407 (accessed 29.11.16).

408 ISO, 2012, ISO 3308 Routine Analytical Cigarette-Smoking Machine - Definitions
409 and Standard Conditions, ISO/TC 126.

410 Khlystov, A., Samburova, V., 2016. Flavouring compounds dominate toxic aldehyde
411 production during e-cigarette vaping. *Env. Sci. Technol.* 50, 13080-13085.

412 King, B.A., Patel, R., Nguyen, K.H., Dube, S.R., 2015. Trends in awareness and use
413 of electronic cigarettes among US Adults, 2010-2013. *Nicotine Tob. Res.* 17,
414 219-227.

415 Lerner, C.A., Sundar, I.K., Watson, R.M., Elder, A., Jones, R., Done, D., Kurtzman, R.,

416 Ossip, D.J., Robinson, R., McIntosh, S., Rahman, I., 2015. Environmental health
417 hazards of e-cigarettes and their components: oxidants and copper in e-cigarette
418 aerosols. *Env. Poll.* 198, 100-107.

419 Liu, Y., Zheng, G.J., Yu, H.X., Martin, M., Richardson, B.J., Lam, M.H.W., Lam, P.K.
420 S., 2005. Polybrominated diphenyl ethers (PBDEs) in sediments and mussel tissue
421 from Hong Kong marine waters. *Marine Pollut. Bull.* 50, 1173-1184.

422 McNeill, A., Etter, J.F., Farsalinos, K., Hajek, P., Le Houezec, J., McRobbie, H., 2014.
423 A critique of a World Health Organization commissioned report and associated
424 paper on electronic cigarettes. *Addiction* 109, 2128-2134.

425 Ni, K., Lu, Y.L., Wang, T., Kannan, K., Gosens, J., Xu, Li., Li, Q.S., Wang, L., Liu, S.,
426 2013. A review of human exposure to polybrominated diphenyl ethers (PBDEs)
427 in China. *Int. J. Hyg. Env. Health* 216, 607-623.

428 Oros, D.R., Hoover, D., Rodigari, F., Crane, D., Sericano, J., 2005. Levels and
429 distribution of polybrominated diphenyl ethers in water, surface sediments, and
430 bivalves from San Francisco Estuary. *Env. Sci. Technol.* 39, 33-41.

431 Pearson, J.L., Richardson, A., Niaura, R.S., Vallone, D.M., Abrams, D.B., 2012.
432 E-cigarette awareness, use, and harm perceptions in US Adults. *Am J. Public*
433 *Health* 102, 1758-1766.

434 *Politics and Government Week*, 2016. Patents; "electronic cigarette and method for

435 manufacturing electronic cigarette" in patent application approval process
436 (USPTO 20160128387). 2nd June, 1403.
437 <https://search.proquest.com/docview/1791393778?accountid=11440> (accessed
438 06.07.17)

439 Parker, T.T., Rayburn, J., 2017. A comparison of electronic and traditional cigarette
440 butt leachate on the development of *Xenopus laevis* embryos. *Toxicol. Rep.* 17,
441 77-82.

442 Schecter, A., Pöpke, O., Harris, T.R., Tung, K.C., Musumba, A., Olson, J., Birnbaum,
443 L., 2006. Polybrominated Diphenyl Ether (PBDE) levels in an expanded market
444 basket survey of US food and estimated PBDE dietary intake by age and sex. *Env.*
445 *Health Perspect.* 114, 1515-8.

446 The Royal College of Physicians, 2016. Nicotine without smoke, tobacco harm
447 reduction. A report by the Tobacco Advisory Group of the Royal College of
448 Physicians. April.
449 <https://www.rcplondon.ac.uk/file/3563/download?token=uV0R0Twz> (accessed
450 21.07.16)

451 The Secretariat of the Stockholm Convention, Stockholm Convention on Persistent
452 Organic Pollutants (POPs) as amended in 2009, UNEP 2009.
453 <http://chm.pops.int/TheConvention/Overview/TextoftheConvention/tabid/2232/D>

454 [efault.aspx](#) (accessed 20.12.15).

455 USEPA, 2014. Technical fact sheet-polybrominated diphenyl ethers (PBDEs) and
456 polybrominated biphenyls (PBBs). EPA 505-F-14-006.

457 US Department of Health and Human Services, 2016. E-Cigarette Use among Youth
458 and Young Adults: A Report of the Surgeon General—Executive Summary.
459 Atlanta, GA: U.S. Department of Health and Human Services, Centers for
460 Disease Control and Prevention, National Center for Chronic Disease Prevention
461 and Health Promotion, Office on Smoking and Health.
462 [https://e-cigarettes.surgeongeneral.gov/documents/2016_SGR_Exec_Summ_508.](https://e-cigarettes.surgeongeneral.gov/documents/2016_SGR_Exec_Summ_508.pdf)
463 [pdf](#) (accessed 06.07.17)

464 US Food and Drug Administration. Evaluation of e-cigarettes 2009.
465 DPATR-FY-09-23, 2009.
466 <http://www.fda.gov/downloads/Drugs/ScienceResearch/ucm173250.pdf> (accessed
467 04.01.16).

468 Wang, H.S., Du, J., Ho, K.L., Leung, H.M., Lam, M.H.W., Giesy, J.P., Wong, C.K.C.,
469 Wong, M.H., 2011. Exposure of Hong Kong residents to PBDEs and their
470 structural analogues through market fish consumption. *J. Hazard. Mater.* 192,
471 374-380.

472 Wang, H.S., Jiang, G.M., Chen, Z.J., Du, J., Man, Y.B., Giesy, J.P., Wong, C.K.C.,

473 Wong, M.H., 2013. Concentrations and congener profiles of polybrominated
474 diphenyl ethers (PBDEs) in blood plasma from Hong Kong: Implications for
475 sources and exposure route. J. Hazard. Mater. 261,253-259.

476 WHO Framework Convention on Tobacco Control, 2014. Electronic Nicotine
477 Delivery Systems: Report by WHO. 21 July. FCTC/COP/6/10 Rev.1 Moscow.
478 http://apps.who.int/gb/fctc/PDF/cop6/FCTC_COP6_10Rev1-en.pdf (accessed
479 16.02.16).

480 Zelikoff, J., 2016. Reproductive and developmental effects of exposure to emerging
481 tobacco products. AAAS annual meeting, 12th Feb., Washington DC.
482 <https://aaas.confex.com/aaas/2016/webprogram/Paper16210.html> (accessed
483 21.07.16)