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1 **Sprays and diffusers as indoor air fresheners: exposure and health risk assessment based on**  
2 **measurements under realistic indoor conditions**

3

4 *Short running title* - Air fresheners: HRA for sprays and diffusers

5

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21 **Data availability statement**

22 The data that support the findings of this study will be made available online in a public report

23 (PRESSENS project - <https://www.ademe.fr/> - abstract in English) before March 2021.

24

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31

32 **Conflict of interest disclosure**

33 The authors declare that they have no conflicts of interest.

34

35 **CRedit authorship contribution statement**

36 Guillaume KARR: Conceptualization, Formal Analysis, Methodology, Original Draft Preparation,  
37 Writing – Review & Editing. Etienne QUIVET: Methodology, Investigation, Validation, Data  
38 Curation, Writing – Review & Editing. Martine RAMEL: Conceptualization, Methodology,  
39 Validation, Writing – Review & Editing, Project Administration. Mélanie NICOLAS:  
40 Methodology, Validation, Data Curation, Writing – Review & Editing, Funding Acquisition,  
41 Project Administration.

42

## 43 **Abstract**

44 Noncombustible air fresheners are indoor air emission sources of concern. The associated health  
45 risks should be better understood. Based on 15 products (4 sprays, 6 passive diffusers and 5 active  
46 diffusers), the health risk assessment (HRA) approach was applied to a national use survey in  
47 France and to concentrations measured in an experimental house. The targeted substances included  
48 volatile organic compounds (VOCs), carbonyls, and fine particles (PM<sub>2.5</sub>). Mean-use and  
49 reasonable worst-case generic scenarios were designed. No situation of concern occurred regarding  
50 chronic exposure associated with the mean use. Under the reasonable worst-case scenarios, the  
51 chronic risk could exceed selected health reference standards, mainly for acrolein (average inhaled  
52 concentration (AIC) up to 3.5 µg/m<sup>3</sup>), benzene (AIC up to 4 µg/m<sup>3</sup>), and limonene (AIC up to 8  
53 mg/m<sup>3</sup>). The acute exposure, defined as a 1-hour exposure, could exceed selected health standards,  
54 primarily for acrolein (up to 23 µg/m<sup>3</sup>) and formaldehyde (up to approximately 370 µg/m<sup>3</sup>).  
55 Furthermore, the 1-hour average PM<sub>2.5</sub> concentration, including ultrafine particles, could exceed  
56 100 µg/m<sup>3</sup>, typically for sprays. These results suggest that the highest exposures should be reduced  
57 and, as such, that the emissions of the highest-emissivity products should be lowered.

58

## 59 **Keywords**

60 air fresheners; health risk assessment; indoor air; sprays; diffusers; emission sources

61

## 62 **Practical implications**

- 63 ● This study provides evidence that exposure to noncombustible air fresheners may lead to  
64 situations of concern.
- 65 ● The highest risk may be associated with exposure to acrolein, benzene, limonene,  
66 formaldehyde, and fine particles (PM<sub>2.5</sub>).

- 67 • Chronic and acute calculated exposures are provided for these priority substances of  
68 concern.
- 69 • Exposures may be highly reduced by reasonable use, room ventilation after use, and direct  
70 inhalation avoidance.
- 71 • The obtained results suggest that the emissions of high-emissivity products should be  
72 reduced to ensure safe use.

73

## 74 **Introduction**

75 In temperate climates, most people spend nearly 90% of their time in indoor environments, mainly  
76 at home. Consequently, the indoor air quality is a topic of major importance for public health.<sup>1-3</sup>

77 Noncombustible air fresheners, such as sprays and diffusers (i.e., not burning incenses or scented  
78 candles), are commonly applied consumer products. These products can be specific indoor  
79 emission sources of air pollutants, e.g., formaldehyde, particles, and terpenes,<sup>4-7</sup> and are a subject  
80 of concern.<sup>6-9</sup> The associated risks should be better understood.<sup>7,10</sup> In particular, only a few health  
81 risk assessment (HRA) studies have been conducted, generally studying a limited number of  
82 products and substances based on models and laboratory measurements carried out in emission test  
83 chambers.<sup>6,11-15</sup>

84 This study aimed to contribute to a better characterization of the exposure and risk levels associated  
85 with the household use of noncombustible air fresheners: an HRA was conducted on fifteen  
86 noncombustible air fresheners of various types based on measurements under realistic indoor  
87 conditions. In particular, this HRA aimed to identify the emitted substances of the highest concern  
88 and to assess the associated health issues.

89

## 90 **Methods**

### 91 *Experiments under realistic indoor conditions*

92 Fifteen noncombustible air fresheners were selected from different manufacturers and sellers. All  
93 products are available and can be used by consumers. The selected products included 4 sprays,  
94 6 passive diffusers, and 5 active diffusers. In this study, passive and active indicate the absence and  
95 use, respectively, of a source of energy.

96 The emissions from the above selected products were measured in the Mechanized house for  
97 Advanced Research on Indoor Air (MARIA; Scientific and Technical Center of Building (CSTB),  
98 Marne-la-Vallée, France).<sup>16</sup> The room where the measurements were carried out corresponded to  
99 a 32-m<sup>3</sup> bedroom (See details in Supplementary Information (SI) “Room description”). This room  
100 did not contain furniture, and the finishing coatings were minimal: raw concrete floor, painted  
101 plasterboard walls, and painted concrete ceiling. All experiments were conducted under typical  
102 indoor environmental parameters. The indoor air conditions of the room, e.g., the temperature ( $17$   
103  $\pm 1$ )°C and relative humidity ( $38 \pm 3$ )%, were continuously monitored. The air change rate (ACR)  
104 was controlled and maintained by the means of mechanical ventilation system and the resulting  
105 ACR was ( $0.45 \pm 0.03$ ) /h.

106 Indoor air samples were collected at the extraction equipment. Consequently, the corresponding  
107 air is assumed to reflect the average concentration in the room.

108 A detailed physicochemical characterization of the emissions was performed using online  
109 instrumentation and off-line chemical analysis of the gaseous phase (See details in SI “Sampling”  
110 and “Analysis”).

111 After reviewing the scientific literature and pretesting in emission test chambers, the selected  
112 targeted substances included:

- 113 • Volatile organic compounds (VOCs), including benzene, toluene, ethylbenzene, styrene,  
114 xylenes, naphthalene, d-limonene, and alpha-pinene;
- 115 • Carbonyl compounds, including formaldehyde, acetaldehyde, acetone, propionaldehyde,  
116 benzaldehyde, and acrolein;
- 117 • Particulate matter, including particles with an aerodynamic diameter of less than  
118 10 microns (PM<sub>10</sub>) and fine particles (PM<sub>2.5</sub>).

119 Semi-volatile organic compounds (SVOCs) were not in the scope of this HRA.

120 Volatile organic compounds (VOCs) and carbonyls compounds, including benzene, toluene,  
121 styrene, formaldehyde, acetaldehyde, acetone, and acrolein were monitored online using a proton  
122 transfer reaction-time of flight-mass spectrometer (PTR-ToF-MS 8000, Ionicon Analytik GmbH).

123 Other carbonyl compounds, including propionaldehyde and benzaldehyde, were collected on  
124 DNPH coated cartridges (Waters) and quantified using HPLC-DAD (Alliance, Waters) after liquid  
125 extraction with 5 mL of acetonitrile. Other VOCs, including ethylbenzene, xylenes, naphthalene,  
126 d-limonene, and alpha-pinene, were collected on Tenax TA sorbent tubes (Perkin Elmer), extracted  
127 by automated thermal desorption (ATD Turbomatrix, Perkin Elmer), and analyzed using GC/MS  
128 (Perkin Elmer Clarus / Turbomass) for identification and quantification).

129 Particles were monitored using a scanning mobility particle sizer (SMPS, model 3936, TSI - from  
130 10 nm to 807 nm) and an optical particle counter (OPC, model 1.109, GRIMM Aerosol Technik  
131 GmbH - from 260 nm to 32 µm).

132 All the results obtained are available in SI “Field measurement data”.

133

#### 134 *Design of the generic exposure scenarios*

135 A survey on the household use of sprays and diffusers was conducted in September 2017 by the  
136 French Ministry of Environment.<sup>17</sup> The survey consisted of interviews carried out via self-

137 administered online questionnaires. These interviews were conducted on a sample of  
138 approximately 1,500 people, representative of the French population over the age of 18. This  
139 representativeness was obtained by the quota sampling method (sex, age, and occupation) after  
140 stratification by region and by the urban area category.

141 Based on the results of this national survey, two types of generic exposure scenarios were designed.  
142 The mean-use scenarios aimed to characterize a user with common practices (i.e., frequency and  
143 duration of use) and environmental characteristics (i.e., room volume, air change rate), based on  
144 the average of the observed practices and characteristics. Typically, the components of these  
145 scenarios were chosen from the 50<sup>th</sup> percentiles of the responses collected in the national survey.  
146 The reasonable worst-case scenarios aimed to characterize a user whose practices and  
147 environmental characteristics reasonably increase the average exposure. Typically, the components  
148 of these scenarios were chosen from the 90<sup>th</sup> percentiles of the responses collected in the national  
149 survey.

150 When the national survey did not provide enough information to select a value for certain  
151 components of the considered scenarios, the values determined in the European EPHECT  
152 project<sup>18,19</sup> were selected, e.g., volume, air change rate, and daily presence in the rooms of the  
153 considered house.

154 Each scenario was defined by several components: frequency of use in and volume of each room  
155 where noncombustible air fresheners are used (units: /d and m<sup>3</sup>, respectively), air change rate (/h),  
156 daily presence in the rooms where the products are used (h/d), duration of exposure over a lifetime  
157 (years), potential manual ventilation of the room (i.e., a user opens a window to temporarily  
158 increase the ventilation), during or after air freshener use (yes/no), and duration of use (for sprays:  
159 number of sprayings, dimensionless, and duration of each spraying, s; for diffusers used  
160 continuously: months/year; for diffusers used sporadically: min per use).



161

162 ***Exposure assessment***

163 The results of the national survey revealed that noncombustible air fresheners were used in several  
164 rooms at home. Consequently, a house with seven types of rooms was considered in the HRA:  
165 kitchen, bedroom, bathroom, living/dining room, restroom (WC), entrance hall, and storeroom.  
166 The corresponding considered characteristics (Table 1) approximately conform to those adopted in  
167 other European studies, including the HRA of the EPHECT project (considered population groups:  
168 housekeepers and retired people).<sup>19,20</sup>

169 The results of the field measurements consisted of the average concentration during several  
170 successive periods, depending on the type of emission pattern considered, but typically during the  
171 3 h 30 min following the beginning of use (See details in SI “Sampling”). These concentrations  
172 were extrapolated to daily average concentrations based on the following hypotheses: (i) for mean-  
173 use scenarios, 10 min of manual ventilation renews the air in a room<sup>21</sup> (the measured concentrations  
174 are considered until the manual ventilation); (ii) for reasonable worst-case scenarios, the  
175 concentration in a room is supposedly decreasing only with the selected air change rate (no manual  
176 ventilation ; mass balance approach). Under each scenario and for each emitted substance, these  
177 hypotheses allowed us to extrapolate the measured concentrations to concentrations at any time of  
178 the day in any room. The daily average concentrations were then calculated for each room.

179 Sorption/desorption phenomena and migration within the house were not quantified, assuming the  
180 associated variability to be much lower than the uncertainties associated with the other assumptions  
181 of the HRA.

182 Based on the above daily average concentration data, the average inhaled concentration (AIC) was  
183 calculated by considering the components of each generic exposure scenario: frequency of use,  
184 daily presence in each room, duration of use, etc. For each emitted substance, the AICs of each

185 room were summed to obtain the characteristic AIC of the average daily presence in the considered  
186 house ( $AIC_{\text{house}}$  - 24 h time-weighted-average) (Table 1). These calculated  $AIC_{\text{house}}$  values  
187 characterized the chronic inhalation exposure.

188 The acute exposure level was also assessed. In general, this type of exposure corresponds to periods  
189 that may vary between one hour and a few days. Considering the uses described in the national  
190 survey, an exposure duration of one hour was adopted in the HRA. Therefore, the acute exposure  
191 level was characterized by the maximum 1-hour average concentration ( $HAC_{\text{max}}$ ), combining the  
192 maximum measured (sprays) or expected (diffusers) concentrations with the characteristics of the  
193 considered house (volumes and daily presences detailed in Table 1).

194

#### 195 *Selection of the toxicity values*

196 Regarding both chronic and acute effects, a choice of toxicological reference values (TRVs) was  
197 made for each tested substance from among the TRVs reported in common reference databases,  
198 such as the United States Environmental Protection Agency (US EPA), World Health Organization  
199 (WHO), Agency for Toxic Substances and Disease Registry (ATSDR), French Agency for Food,  
200 Environmental and Occupational Health & Safety (ANSES), Health Canada, Dutch National  
201 Institute for Public Health and the Environment (RIVM) and California Office of Environmental  
202 Health Hazard Assessment (OEHHA). This choice was made in regard to the threshold effect  
203 ( $TRV_T$  -  $\mu\text{g}/\text{m}^3$ ), nonthreshold effect ( $TRV_{NT}$  -  $/(\mu\text{g}/\text{m}^3)$ ) and acute effect ( $TRV_{\text{acute}}$  -  $\mu\text{g}/\text{m}^3$ ).  
204 Considering that  $HAC_{\text{max}}$  was selected to characterize the acute exposure level, the TRVs  
205 associated with a 1-hour exposure duration were preferentially chosen.

206

207 ***Health risk assessment***

208 Based on the exposure levels calculated from the concentrations measured under realistic indoor  
209 conditions and with the generic exposure scenarios designed from the national survey, an HRA  
210 was conducted.

211 The scope of this HRA only included the inhalation route of exposure. The chronic threshold and  
212 nonthreshold risks, as well as the acute risk, were quantified by the following risk indicators:  
213 threshold effect ratio (TER), individual excess risk (IER) and acute risk ratio (ARR), respectively.

214 
$$TER = \frac{AIC_{house}}{TRV_T} \quad (1)$$

215 
$$IER = \frac{AIC_{house} \times TRV_{NT} \times \text{Years of exposure}}{\text{Lifetime of 70 years}} \quad (2)$$

216 
$$ARR = \frac{HAC_{max}}{TRV_{acute}} \quad (3)$$

217  
218 To be conservative, in coherence with the results of the national survey, the HRA considered a  
219 lifetime exposure (i.e., a ratio ‘Years of exposure / Lifetime of 70 years’ equal to 1).

220

221 **Results**

222 ***Elaboration of the generic exposure scenarios***

223 The main general findings of the national survey on household exposure include the following: *The*  
224 *use of noncombustible air fresheners is widespread: more than two-thirds of French people use*  
225 *sprays in their homes, almost half use passive diffusers and more than one-third use active*  
226 *diffusers; uses are well anchored in usual practices; 13% of spray users and 37% of diffuser users*  
227 *report that children may be present in the rooms where noncombustible air fresheners are used;*  
228 *65% of French people report that the use of noncombustible air fresheners improves or does not*

229 affect the indoor air quality; 40% of spray users, 40% of passive diffuser users and 50% of active  
230 diffuser users apply these products aiming to clean the indoor air.

231 The national survey also provided detailed results that enabled us to elaborate two generic exposure  
232 scenarios for each type of noncombustible air freshener considered in the HRA (Table 2).

233

### 234 *Exposure assessment*

235 Under each exposure scenario, a  $AIC_{house}$  was calculated for each quantified substance emitted by  
236 each tested product (Tables S1 and S2). Table 3 presents a overview of the obtained results  
237 (Minimum, Maximum, Median and Average data) for a selection of usual substances of interest.

238 Furthermore, a  $HAC_{max}$  was calculated for each quantified substance emitted by each tested  
239 product. Table 4 provides a overview of the obtained results for a selection of substances known  
240 for their potential acute effects.

241

### 242 *Selection of the toxicity values*

243 For each substance tested in the experimental house, TRV values were selected. Table 5 presents  
244 the TRVs adopted for a selection of substances of interest and their associated critical effects.

245 No TRV was available for limonene and alpha-pinene in the consulted databases. Consequently,  
246 in regard to chronic exposure, the derived European lowest concentrations of interest (EU-LCI),  
247 determined with a method similar to the one used for the TRV values, were considered for  
248 informative purposes only. These EU-LCIs are equal to  $2.5 \text{ mg/m}^3$  (alpha-pinene) and  $5 \text{ mg/m}^3$   
249 (limonene).

250 No TRV was also available for  $PM_{10}$  and  $PM_{2.5}$  in the consulted databases. Consequently, WHO  
251 ambient air quality guideline values were considered for the chronic and acute effects, i.e., 20 and

252 50  $\mu\text{g}/\text{m}^3$ , respectively, for  $\text{PM}_{10}$ ; 10 and 25  $\mu\text{g}/\text{m}^3$ , respectively, for  $\text{PM}_{2.5}$ . However, these  
253 guidelines are not TRVs: they are considered for informative purposes only.

254

### 255 *Chronic risk assessment*

256 For each substance emitted by each tested product and under each generic exposure scenario, TER  
257 and IER values were calculated when  $\text{TRV}_T$  and  $\text{TRV}_{NT}$  values, respectively, were available. The  
258 main obtained results are presented below.

259

### 260 *Mean-use scenarios*

261 For each emitted substance considered individually, no TER nor IER value exceeded the usual  
262 reference values of 1 and  $10^{-5}$  (1-in-10,000 lifetime cancer risk), respectively. Furthermore, for  
263 each product, the multisubstance sums of the TER and IER values did not exceed these usual  
264 reference values.

265

### 266 *Reasonable worst-case scenario – spray users*

267 The acrolein TER value generally exceeded the reference value, ranging from 1.1 to 24.  
268 Furthermore, several  $\text{PM}_{2.5}$  exposure levels exceeded the WHO guideline ( $10 \mu\text{g}/\text{m}^3$ ), ranging from  
269 30 to  $60 \mu\text{g}/\text{m}^3$ .

270 Several benzene IER values exceeded the reference value, ranging from  $4.0 \cdot 10^{-5}$  to  $7.2 \cdot 10^{-5}$ .

271

### 272 *Reasonable worst-case scenario – passive diffuser users*

273 Several acrolein TER values exceeded the reference value, ranging from 1.3 to 1.5. One benzene  
274 IER value exceeded the reference value, equal to  $2.5 \cdot 10^{-5}$ .

275

276 *Reasonable worst-case scenario – active diffuser users*

277 One acrolein TER value exceeded the reference value, equal to 3.6. Furthermore, two PM<sub>2.5</sub>  
278 exposure levels exceeded the WHO guideline (10 µg/m<sup>3</sup>), equal to 13 and 24 µg/m<sup>3</sup>, and several  
279 limonene exposure levels exceeded the corresponding EU-LCI, up to a factor of 1.6.  
280 Several benzene IER values exceeded the reference value, ranging from 7.3.10<sup>-5</sup> to 1.0.10<sup>-4</sup>.

281

282 *Acute risks assessment*

283 For each substance emitted by each tested product, an ARR value was calculated when TRV<sub>acute</sub>  
284 was available. The main obtained results are presented below.

285

286 *Spray users*

287 Two acrolein ARR values exceeded the reference value, equal to 2.6 and 3.3, while several  
288 formaldehyde ARR values exceeded the reference value, ranging from 1.7 to 2.3. Furthermore,  
289 several PM<sub>2.5</sub> exposure levels exceeded the WHO guideline value (25 µg/m<sup>3</sup>, 24h-exposure),  
290 ranging from 130 to 180 µg/m<sup>3</sup>.

291

292 *Passive diffuser users*

293 Two formaldehyde ARR values exceeded the reference value, equal to 3.0 and 3.7.

294

295 *Active diffuser users*

296 Two formaldehyde ARR values exceeded the reference value, equal to 2.3 and 2.9. Furthermore,  
297 several PM<sub>2.5</sub> exposure levels exceeded the WHO guideline value (25 µg/m<sup>3</sup>, 24h-exposure),  
298 ranging from 35 to 190 µg/m<sup>3</sup>.

299 Furthermore, for all tested products, the maximum alpha-pinene HAC<sub>max</sub> value was 2.8 mg/m<sup>3</sup>, and  
300 the maximum limonene HAC<sub>max</sub> value was 52 mg/m<sup>3</sup>, which are below the acute critical exposure  
301 limits (CELs) determined in the EPHECT project,<sup>11</sup> i.e., at 45 and 90 mg/m<sup>3</sup>, respectively.

302

## 303 **Discussion**

### 304 *Elaboration of the generic exposure scenarios*

305 The national survey allowed us to establish generic exposure scenarios for the considered  
306 noncombustible air fresheners. However, a substantial variety of products exists within the spray,  
307 passive diffuser, and active diffuser general categories. For certain tested products, the description  
308 and recommendation of the manufacturers did not precisely fit the established scenarios. For  
309 example, the use could be associated with a laptop USB port, which does not fit well with the use  
310 in a bathroom or restroom (WC). Additionally, recommendations could strongly focus on sporadic  
311 use, e.g., “30 min before your child goes to bed”, which does not suitably conform to the continuous  
312 use under the reasonable worst-case scenario established for active diffusers. Consequently, when  
313 reasonably feasible, the generic exposure scenarios were adapted in this study for certain products  
314 to compensate for the above limitation.

315

### 316 *Exposure assessment*

317 To our knowledge, this HRA is the first to combine measurements under realistic indoor conditions  
318 and the results of a national survey on the use of noncombustible air fresheners.

319 The exposure levels were calculated from the concentrations measured under realistic indoor  
320 conditions over approximately two hours following the beginning of use. These field  
321 concentrations allowed us to consider the secondary substances formed in the indoor air during the  
322 measurement periods. This secondary formation could represent a major contribution to the

323 measured concentrations, as observed for the secondary formation of formaldehyde<sup>5,22,23</sup> that  
324 occurs indoors through chemical reactions between, for example, ozone and terpenes. The  
325 measured field concentrations also allowed us to avoid the difficulties of the simulation of real  
326 emissions processes in a laboratory test chamber. These difficulties include the assumption of  
327 various parameters, e.g., humidity rate (water content of air, %RH), oxygen rate (oxygen content  
328 of air, %), air flow rate (air volume entering chamber per time), and temperature.<sup>24</sup> Furthermore,  
329 the Surface/Volume ratio could favor sorption phenomena and could lead to underestimated  
330 concentrations.

331 The main limitations associated with the AIC determination consist of the rather simple  
332 assumptions chosen to extrapolate the measured concentrations. However, this approach has been  
333 applied in previous studies.<sup>5</sup>

334 The exposure levels associated with the use of noncombustible air fresheners have been assessed  
335 in other studies.<sup>6,14,15,25-30</sup> These studies were based on other hypotheses, other types of  
336 measurements and other methods. The main identified differences are related to the use of emission  
337 data measured in test chambers or under realistic indoor conditions, the use of models to estimate  
338 exposure, the selected products (a great variability in emissions has been observed within the same  
339 type of noncombustible air freshener),<sup>8,10</sup> the selected air change rate and volume of the room where  
340 noncombustible air fresheners are used, the location of the measuring instrumentation, e.g., the  
341 distance to the tested product, and the availability of detailed information on the practices of the  
342 considered users.

343 Consequently, the corresponding exposure levels could not be directly compared to those  
344 determined in this study. However, no obvious inconsistency was identified. Furthermore, the  
345 determined maximum chronic and acute exposure levels frequently exceeded the values reported



346 in the scientific literature, especially for formaldehyde (the acute exposure level is below 100  $\mu\text{g}/\text{m}^3$   
347 in the scientific literature).

348

### 349 *Chronic and acute risk characterization*

350 The obtained results under the mean-use scenarios suggest that the chronic exposure levels  
351 associated with the most common use patterns are not of concern, with regard to the targeted  
352 substances, the selected products and the assumptions made in this HRA.

353 The exceedances determined under the reasonable worst-case scenarios suggest that the highest  
354 chronic exposure levels should be reduced, especially for acrolein and benzene. However, the  
355 associated maximum chronic exposure levels — 3.5 and 4  $\mu\text{g}/\text{m}^3$ , respectively — correspond to  
356 background concentrations reported in certain private dwellings.<sup>31,32</sup>

357 Moreover, the exceedances determined for the acute exposure to acrolein and formaldehyde  
358 suggest a need to reduce the emissions of the highest-emissivity products.

359 The acrolein acute exposure levels (up to 23  $\mu\text{g}/\text{m}^3$ ) can also be compared to the OEHHA acute  
360 reference exposure level (2.5  $\mu\text{g}/\text{m}^3$ , 1 h, respiratory and eye irritation, 2008) and to the acute  
361 critical exposure limit established in the EPHECT project (21  $\mu\text{g}/\text{m}^3$ , 30 min, subjective eye  
362 irritation, 2015).

363 Furthermore, since the indoor air samples were collected with air extraction equipment in the test  
364 room, the measured concentrations were lower than those occurring close to the products.

365 Consequently, the actual acute exposure levels may be much higher than those assessed in this  
366 HRA, thus supporting the identified need to reduce the emissions of high-emissivity products.

367 In the scientific literature, few HRAs have been conducted targeting noncombustible air  
368 fresheners.<sup>6,14,30</sup> These assessments have concluded that no situation of concern is typically  
369 expected. This difference with the conclusions of this HRA can be explained by the differences in

370 the selected toxicity values and by the higher maximum exposure levels determined in the present  
371 study. However, other types of studies have concluded that exposure to noncombustible air  
372 fresheners could be of concern: the measured concentrations could exceed the thresholds associated  
373 with the exacerbation of existing symptoms in people with asthma,<sup>25,33,34</sup> and certain  
374 epidemiological studies have identified a significant association between the use of some air  
375 fresheners and respiratory disorders during the first years of life,<sup>35</sup> a higher risk of breast cancer<sup>36</sup>  
376 and a short-term decrease in lung ventilation capacity,<sup>37</sup> while toxicological studies have reported  
377 a significant association between the application of certain air fresheners and inflammatory  
378 phenomena in rats,<sup>38</sup> neurobehavioral effects in rats,<sup>39</sup> and adverse effects on the liver cells in  
379 young rats.<sup>40</sup> A literature review demonstrated a significant association between the use of  
380 noncombustible air fresheners and certain health effects (disorders of the respiratory system) but  
381 no causal relation was identified,<sup>23</sup> and other reviews indicated that the available data are  
382 insufficient to draw conclusions on the potential health effects associated with aerosol sprays<sup>41</sup> and  
383 essential oil air fresheners,<sup>42</sup> while a European collective expert assessment concluded that certain  
384 noncombustible air fresheners may cause or aggravate symptoms in highly sensitive persons and  
385 may be of concern in children.<sup>8</sup> Surveys conducted in five industrialized countries (the United  
386 States of America, United Kingdom, Germany, Australia and Sweden) indicated that 15% to 20%  
387 of people have reported health effects, e.g., headaches or breathing difficulties, associated with the  
388 use of noncombustible air fresheners.<sup>7,43,44</sup>

389

### 390 ***Priority substances of interest***

391 Based on all the calculated risk indicators, the substances of highest interest identified in the HRA  
392 are acrolein, benzene, PM<sub>2.5</sub>, formaldehyde, and, to a lesser extent, limonene.

393 These substances are not specific to the emissions of noncombustible air fresheners. Other emission  
394 sources are commonly present in indoor environments, e.g., furniture, cleaning products, tobacco  
395 smoke, construction products, paints, and cooking fumes. Consequently, cumulative exposure is  
396 expected, which could lead to higher risks than those identified for each source, considered  
397 individually. Background exposure levels in European private dwellings are available for several  
398 of the above identified priority substances:<sup>19,31,45</sup> formaldehyde (minimum: 7  $\mu\text{g}/\text{m}^3$ ; maximum:  
399 57  $\mu\text{g}/\text{m}^3$ ; mean: 22  $\mu\text{g}/\text{m}^3$ ); benzene (minimum: 0  $\mu\text{g}/\text{m}^3$ ; maximum: 32  $\mu\text{g}/\text{m}^3$ ; mean: 3  $\mu\text{g}/\text{m}^3$ );  
400 limonene (minimum: 0  $\mu\text{g}/\text{m}^3$ ; maximum: 493  $\mu\text{g}/\text{m}^3$ ; mean: 29  $\mu\text{g}/\text{m}^3$ ). The health reference  
401 values selected in this HRA, and those considered in the Discussion section, relate to the total  
402 indoor air concentration: they do not relate to the concentration attributable to the use of  
403 noncombustible air fresheners only. This suggests a need to limit the emissions of noncombustible  
404 air fresheners, especially for the identified priority substances of interest, until the corresponding  
405 concentrations are much lower than the selected health values.

406

#### 407 *Emitted particles*

408 The chronic and acute  $\text{PM}_{2.5}$  exposure levels could exceed the WHO guideline values, especially  
409 for sprays, which may indicate potential situations of concern. However, since the composition of  
410 the particles emitted by noncombustible air fresheners likely greatly differs from the composition  
411 of the ambient air particles considered in the WHO guideline values, the associated risks cannot be  
412 precisely characterized. Further studies are required to evaluate whether situations of concern are  
413 expected.

414 However, a portion of the  $\text{PM}_{2.5}$  particles emitted by the tested noncombustible air fresheners were  
415 submicron particles ( $\text{PM}_1$ ), even ultrafine particles ( $\text{PM}_{0.1}$ ), which is consistent with the results of  
416 other studies<sup>23,46-48</sup> and certain descriptions of manufacturers. For example, a spray and active

417 diffuser yielded a 1-hour PM<sub>1</sub> concentration in excess of 3 µg/m<sup>3</sup>; singularly among the tested  
418 products, one active diffuser (atomizer) resulted in a 1-hour PM<sub>1</sub> concentration of approximately  
419 88 µg/m<sup>3</sup>. These submicron particles are a subject of concern because of their high surface  
420 reactivity and ability to penetrate the pulmonary system.<sup>49,50</sup>

421

## 422 **Conclusions**

423 The HRA approach was applied to better understand the health risks associated with  
424 noncombustible air fresheners, thereby identifying the emitted substances of highest concern and  
425 evaluating the associated risks.

426 Fifteen noncombustible air fresheners including 4 sprays, 6 passive diffusers and 5 active diffusers  
427 were tested in an experimental house under realistic indoor conditions.

428 Based on a national survey on air freshener use in France, conducted in 2017 by the French Ministry  
429 of Environment, two types of generic exposure scenarios were designed, namely, mean-use  
430 scenarios, and reasonable worst-case scenarios.

431 To our knowledge, this HRA is the first to combine concentrations measured under realistic indoor  
432 conditions and the results of a national use survey.

433 The obtained results under the mean-use scenarios suggest that the chronic exposure levels  
434 associated with the most common uses are not of concern, with regard to the targeted substances,  
435 the selected products and the assumptions made in this HRA.

436 The acquired results under the reasonable worst-case scenarios suggest that the highest chronic  
437 exposure levels should be reduced, especially for acrolein (up to 3.5 µg/m<sup>3</sup>) and benzene (up to  
438 4 µg/m<sup>3</sup>).

439 The obtained acute exposure (1 h) results suggest a need to reduce the emissions of the highest-  
440 emissivity products, especially acrolein (up to 23  $\mu\text{g}/\text{m}^3$ ) and formaldehyde (up to approximately  
441 370  $\mu\text{g}/\text{m}^3$ ).

442 The emitted  $\text{PM}_{2.5}$  particles could also be of concern because the acute exposure level reached  
443 180  $\mu\text{g}/\text{m}^3$ , and some of them included submicron (up to more than 3  $\mu\text{g}/\text{m}^3$ ) and ultrafine particles.

444 The obtained results provide a complementary perspective to the results of the European EPHECT  
445 project:<sup>11,19</sup> this HRA was focused on noncombustible air fresheners based on actual indoor air  
446 concentrations (experimental values instead of modeled concentrations) and considered a large set  
447 of substances. The same strategy, combining experiments under realistic conditions and a national  
448 use survey, could be implemented to assess the health risks of many other common consumer  
449 products.

450

451

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608

## 609 **Tables**

610 **Table 1.** Volume and daily presence in each room considered in the health risk assessment

<b>Rooms</b>	<b>Volume (m<sup>3</sup>)</b>	<b>Daily presence (h/d)</b>
Kitchen	30	2.75
Bedroom	45	8.5
Bathroom	24	1
Living/Dining room	90	5.75
Restroom (WC)	5	0.5
Entrance hall	12	0.5
Storeroom	10	0.5

611

**Table 2.** Generic exposure scenarios designed in the health risk assessment based on the results of a national survey on noncombustible air freshener use

Scenarios components		Spray users		Passive diffuser users		Active diffuser users	
		Reasonable Worst Case	Mean Use	Reasonable Worst Case	Mean Use	Reasonable Worst Case	Mean Use
Frequency of use (/d)	Restroom (WC)	6	1.5	Continuous	Continuous	Continuous	-
	Living/Dining room	2.5	0.1		-		0.43
	Kitchen	2.5	0.1		-		-
	Entrance hall	1.5	0.03		-		-
	Bathroom	1.5	0.03		-		-
	Bedroom(s)	1	0.01		-		-
	Other room(s)	0.43	0.01		-		-
Duration of use		Number of sprayings		Number of months per year (continuous use)		Number of months per year (continuous use)	Duration of each use (min)
		4	2				
		Duration of each spraying (s)		11	10	11	40
		2	1				
Presence in the room during and after use		Yes	Leaving the room after use, may return after 30'	yes	yes	yes	yes
Years of exposure (years)		70	70	70	70	70	70
Manual ventilation (opening to the outside)		No manual ventilation	3 times/4: 10' opening, 20' after use 1 time/4: no manual ventilation	No manual ventilation	10' each day	No manual ventilation	10' and 30' after use
Air change rate (/h)		0.35	0.35	0.35	0.35	0.35	0.35

**Table 3.** Chronic exposure levels for a selection of substances of interest - average inhaled concentration (AIC -  $\mu\text{g}/\text{m}^3$ ) under the reasonable worst-case scenarios

Substances	CAS number	Spray users				Passive diffuser users				Active diffuser users			
		Min.	Max.	Med.	Avg.	Min.	Max.	Med.	Avg.	Min.	Max.	Med.	Avg.
Formaldehyde	50-00-0	0.31	52	37	30	0.0086	57	2.2	13	0	45	1.6	8.4
Acetaldehyde	75-07-0	0.82	84	12	30	0	63	0.44	8.7	0	46	0.47	7.6
D-Limonene	5989-27-5	3.1	2220	427	591	0	97	1.1	17	0	7981	56	1006
Acetone	67-64-1	5.6	120	55	51	0	8.9	0.0062	2.1	0	40	5.3	5.1
Acrolein	107-02-8	0.17	3.5	1.1	1.3	0	0.22	0	0.047	0	0.54	0.013	0.12
PM <sub>2.5</sub>	-	0	62	19	25	0	0	0	0	0	24	0.88	4.3
Benzene	71-43-2	0	2.8	0.9	1.2	0	0.97	0	0.14	0	4	0.066	0.61

Note: Min.: minimum; Max.: maximum; Med.: median; Avg.: average; 0: below the limit of detection

**Table 4.** Calculated acute exposure levels for a selection of substances of interest - maximum 1-hour average concentration ( $\mu\text{g}/\text{m}^3$ )

Substances	CAS number	Spray users				Passive diffuser users				Active diffuser users			
		Min.	Max.	Med.	Avg.	Min.	Max.	Med.	Avg.	Min.	Max.	Med.	Avg.
Formaldehyde	50-00-0	0.92	230	110	110	0.056	370	15	84	0	290	5.8	49
Acetaldehyde	75-07-0	2.9	355	63	120	0	411	2.8	57	0	298	2.4	50
Acrolein	107-02-8	0.53	23	2.8	6.7	0	1.5	0	0.3	0	3.5	0.27	0.91
Particles PM <sub>2.5</sub>	-	0	180	76	76	0	0	0	0	0	190	5.7	39
Benzene	71-43-2	0	7.2	2.3	3	0	6.3	0	0.93	0	21	0.97	4.2

Note: Min.: minimum; Max.: maximum; Med.: median; Avg.: average; 0: below the limit of detection

**Table 5.** Values characterizing the toxicity of several substances of interest

CAS number	Substances	TRV <sub>T</sub> (µg/m <sup>3</sup> )	Org.	Date	TRV <sub>NT</sub> (µg/m <sup>3</sup> ) <sup>1</sup>	Org.	Dated	TRV <sub>acute</sub> (µg/m <sup>3</sup> )	Org.	Date	Critical effects
50-00-0	Formaldehyde	-	-	-	-	-	-	1.0.10 <sup>2</sup>	WHO	2010	Acute: Subjective and objective eye irritation
75-07-0	Acetaldehyde	1.6.10 <sup>2</sup>	ANSES	2014	-	-	-	3.0.10 <sup>3</sup>	ANSES	2014	Threshold: Degeneration of the olfactory epithelium Acute: Bronchoconstriction in individuals with asthma
107-02-8	Acrolein	1.5.10 <sup>-1</sup>	ANSES	2019	-	-	-	6.9	ATSDR	2007	Threshold: Lesions of the upper respiratory epithelium Acute: Nasal and throat irritation, decreased respiratory rate
108-88-3	Toluene	2.0.10 <sup>4</sup>	ANSES	2018	-	-	-	2.0.10 <sup>4</sup>	ANSES	2018	Threshold: Neurological effects (color vision disorders) Acute: Neurological effects
91-20-3	Naphthalene	1.10 <sup>1</sup>	WHO	2013	5.6.10 <sup>-6</sup>	ANSES	2013	-	-	-	Threshold: Lesions of the respiratory and olfactory epithelium Nonthreshold: Neuroblastomas of the olfactory epithelium
50-32-8	Benzo(a)pyrene	2.0.10 <sup>-3</sup>	USEPA	2017	6.0.10 <sup>-4</sup>	US EPA	2017	-	-	-	Threshold: Increased fetal embryonic mortality Nonthreshold: Occurrence of respiratory tumors
71-43-2	Benzene	1.0.10 <sup>1</sup>	ANSES	2010	2.6.10 <sup>-5</sup>	ANSES	2013	2.7.10 <sup>1</sup>	OEHHA	2014	Threshold: Immunological disorders Nonthreshold: Acute leukemia Acute: Reproductive disorders, aplastic anemia and acute myeloid leukemia

Notes:

- † TRV<sub>T</sub> is the threshold toxicological reference value, for the inhalation route and chronic exposure;
- † TRV<sub>NT</sub> is the nonthreshold toxicological reference value, for the inhalation route and chronic exposure;
- † TRV<sub>acute</sub> is the acute toxicological reference value for the inhalation route;
- † Date is the date of construction or the date of last revision;
- † Org. is the producing organization.