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► To cite this version:

Guillaume Karr, Etienne Quivet, Martine Ramel, Mélanie Nicolas. Sprays and diffusers as indoor air fresheners: Exposure and health risk assessment based on measurements under realistic indoor conditions. *Indoor Air*, 2022, 32 (1), pp.e12923. 10.1111/ina.12923 . hal-03328669

HAL Id: hal-03328669

<https://hal.science/hal-03328669>

Submitted on 30 Aug 2021

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

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

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Data availability statement

The data that support the findings of this study will be made available online in a public report (PRESSENS project - <https://www.ademe.fr/> - abstract in English) before March 2021.

24

25 **Acknowledgments**

26 The authors would like to thank the French Agency for Ecological Transition (ADEME, in the
27 framework of the CORTEA research call through the research project PRESSENS) and the French
28 Ministry of Environment (Ministère de la Transition Écologique) for their financial support;
29 Nathalie VELLY (Ineris) for her technical support; Isabelle AUG EVEN-BOUR (ADEME) and
30 Souad BOUALLALA (ADEME) for their benevolent supervision of the PRESSENS project.

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

Abstract

Noncombustible air fresheners are indoor air emission sources of concern. The associated health risks should be better understood. Based on 15 products (4 sprays, 6 passive diffusers and 5 active diffusers), the health risk assessment (HRA) approach was applied to a national use survey in France and to concentrations measured in an experimental house. The targeted substances included volatile organic compounds (VOCs), carbonyls, and fine particles (PM_{2.5}). Mean-use and reasonable worst-case generic scenarios were designed. No situation of concern occurred regarding chronic exposure associated with the mean use. Under the reasonable worst-case scenarios, the chronic risk could exceed selected health reference standards, mainly for acrolein (average inhaled concentration (AIC) up to 3.5 µg/m³), benzene (AIC up to 4 µg/m³), and limonene (AIC up to 8 mg/m³). The acute exposure, defined as a 1-hour exposure, could exceed selected health standards, primarily for acrolein (up to 23 µg/m³) and formaldehyde (up to approximately 370 µg/m³). Furthermore, the 1-hour average PM_{2.5} concentration, including ultrafine particles, could exceed 100 µg/m³, typically for sprays. These results suggest that the highest exposures should be reduced and, as such, that the emissions of the highest-emissivity products should be lowered.

Keywords

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

Practical implications

- This study provides evidence that exposure to noncombustible air fresheners may lead to situations of concern.
- The highest risk may be associated with exposure to acrolein, benzene, limonene, formaldehyde, and fine particles (PM_{2.5}).

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

Introduction

In temperate climates, most people spend nearly 90% of their time in indoor environments, mainly at home. Consequently, the indoor air quality is a topic of major importance for public health.¹⁻³ Noncombustible air fresheners, such as sprays and diffusers (i.e., not burning incenses or scented candles), are commonly applied consumer products. These products can be specific indoor emission sources of air pollutants, e.g., formaldehyde, particles, and terpenes,⁴⁻⁷ and are a subject of concern.⁶⁻⁹ The associated risks should be better understood.^{7,10} In particular, only a few health risk assessment (HRA) studies have been conducted, generally studying a limited number of products and substances based on models and laboratory measurements carried out in emission test chambers.^{6,11-15} This study aimed to contribute to a better characterization of the exposure and risk levels associated with the household use of noncombustible air fresheners: an HRA was conducted on fifteen noncombustible air fresheners of various types based on measurements under realistic indoor conditions. In particular, this HRA aimed to identify the emitted substances of the highest concern and to assess the associated health issues.

Methods

Experiments under realistic indoor conditions

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

The emissions from the above selected products were measured in the Mechanized house for Advanced Research on Indoor Air (MARIA; Scientific and Technical Center of Building (CSTB), Marne-la-Vallée, France).¹⁶ The room where the measurements were carried out corresponded to a 32-m³ bedroom (See details in Supplementary Information (SI) “Room description”). This room did not contain furniture, and the finishing coatings were minimal: raw concrete floor, painted plasterboard walls, and painted concrete ceiling. All experiments were conducted under typical indoor environmental parameters. The indoor air conditions of the room, e.g., the temperature (17 ± 1)°C and relative humidity (38 ± 3)%, were continuously monitored. The air change rate (ACR) was controlled and maintained by the means of mechanical ventilation system and the resulting ACR was (0.45 ± 0.03) /h.

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

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

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

- Volatile organic compounds (VOCs), including benzene, toluene, ethylbenzene, styrene, xylenes, naphthalene, d-limonene, and alpha-pinene;
- Carbonyl compounds, including formaldehyde, acetaldehyde, acetone, propionaldehyde, benzaldehyde, and acrolein;
- Particulate matter, including particles with an aerodynamic diameter of less than 10 microns (PM₁₀) and fine particles (PM_{2.5}).

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

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

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

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

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

Design of the generic exposure scenarios

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

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

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

When the national survey did not provide enough information to select a value for certain components of the considered scenarios, the values determined in the European EPHECT project^{18,19} were selected, e.g., volume, air change rate, and daily presence in the rooms of the considered house.

Each scenario was defined by several components: frequency of use in and volume of each room where noncombustible air fresheners are used (units: /d and m³, respectively), air change rate (/h), daily presence in the rooms where the products are used (h/d), duration of exposure over a lifetime (years), potential manual ventilation of the room (i.e., a user opens a window to temporarily increase the ventilation), during or after air freshener use (yes/no), and duration of use (for sprays: number of sprayings, dimensionless, and duration of each spraying, s; for diffusers used continuously: months/year; for diffusers used sporadically: min per use).

Exposure assessment

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

The results of the field measurements consisted of the average concentration during several successive periods, depending on the type of emission pattern considered, but typically during the 3 h 30 min following the beginning of use (See details in SI “Sampling”). These concentrations were extrapolated to daily average concentrations based on the following hypotheses: (i) for mean-use scenarios, 10 min of manual ventilation renews the air in a room²¹ (the measured concentrations are considered until the manual ventilation); (ii) for reasonable worst-case scenarios, the concentration in a room is supposedly decreasing only with the selected air change rate (no manual ventilation ; mass balance approach). Under each scenario and for each emitted substance, these hypotheses allowed us to extrapolate the measured concentrations to concentrations at any time of the day in any room. The daily average concentrations were then calculated for each room.

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

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

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

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

Selection of the toxicity values

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

Health risk assessment

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

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

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

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

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

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

Results

Elaboration of the generic exposure scenarios

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

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

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

Exposure assessment

Under each exposure scenario, a AIC_{house} was calculated for each quantified substance emitted by each tested product (Tables S1 and S2). Table 3 presents a overview of the obtained results (Minimum, Maximum, Median and Average data) for a selection of usual substances of interest. Furthermore, a HAC_{max} was calculated for each quantified substance emitted by each tested product. Table 4 provides a overview of the obtained results for a selection of substances known for their potential acute effects.

Selection of the toxicity values

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

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

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

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

Chronic risk assessment

For each substance emitted by each tested product and under each generic exposure scenario, TER and IER values were calculated when TRV_T and TRV_{NT} values, respectively, were available. The main obtained results are presented below.

Mean-use scenarios

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

Reasonable worst-case scenario – spray users

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

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

Reasonable worst-case scenario – passive diffuser users

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

Reasonable worst-case scenario – active diffuser users

One acrolein TER value exceeded the reference value, equal to 3.6. Furthermore, two PM_{2.5} exposure levels exceeded the WHO guideline (10 µg/m³), equal to 13 and 24 µg/m³, and several limonene exposure levels exceeded the corresponding EU-LCI, up to a factor of 1.6. Several benzene IER values exceeded the reference value, ranging from 7.3.10⁻⁵ to 1.0.10⁻⁴.

Acute risks assessment

For each substance emitted by each tested product, an ARR value was calculated when TRV_{acute} was available. The main obtained results are presented below.

Spray users

Two acrolein ARR values exceeded the reference value, equal to 2.6 and 3.3, while several formaldehyde ARR values exceeded the reference value, ranging from 1.7 to 2.3. Furthermore, several PM_{2.5} exposure levels exceeded the WHO guideline value (25 µg/m³, 24h-exposure), ranging from 130 to 180 µg/m³.

Passive diffuser users

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

Active diffuser users

Two formaldehyde ARR values exceeded the reference value, equal to 2.3 and 2.9. Furthermore, several PM_{2.5} exposure levels exceeded the WHO guideline value (25 µg/m³, 24h-exposure), ranging from 35 to 190 µg/m³.

Furthermore, for all tested products, the maximum alpha-pinene HAC_{max} value was 2.8 mg/m³, and the maximum limonene HAC_{max} value was 52 mg/m³, which are below the acute critical exposure limits (CELs) determined in the EPHECT project,¹¹ i.e., at 45 and 90 mg/m³, respectively.

Discussion

Elaboration of the generic exposure scenarios

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

Exposure assessment

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

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

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

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

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

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

in the scientific literature, especially for formaldehyde (the acute exposure level is below 100 µg/m³ in the scientific literature).

Chronic and acute risk characterization

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

The exceedances determined under the reasonable worst-case scenarios suggest that the highest chronic exposure levels should be reduced, especially for acrolein and benzene. However, the associated maximum chronic exposure levels — 3.5 and 4 µg/m³, respectively — correspond to background concentrations reported in certain private dwellings.^{31,32}

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

The acrolein acute exposure levels (up to 23 µg/m³) can also be compared to the OEHHA acute reference exposure level (2.5 µg/m³, 1 h, respiratory and eye irritation, 2008) and to the acute critical exposure limit established in the EPHECT project (21 µg/m³, 30 min, subjective eye irritation, 2015).

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

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

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

the selected toxicity values and by the higher maximum exposure levels determined in the present study. However, other types of studies have concluded that exposure to noncombustible air fresheners could be of concern: the measured concentrations could exceed the thresholds associated with the exacerbation of existing symptoms in people with asthma,^{25,33,34} and certain epidemiological studies have identified a significant association between the use of some air fresheners and respiratory disorders during the first years of life,³⁵ a higher risk of breast cancer³⁶ and a short-term decrease in lung ventilation capacity,³⁷ while toxicological studies have reported a significant association between the application of certain air fresheners and inflammatory phenomena in rats,³⁸ neurobehavioral effects in rats,³⁹ and adverse effects on the liver cells in young rats.⁴⁰ A literature review demonstrated a significant association between the use of noncombustible air fresheners and certain health effects (disorders of the respiratory system) but no causal relation was identified,²³ and other reviews indicated that the available data are insufficient to draw conclusions on the potential health effects associated with aerosol sprays⁴¹ and essential oil air fresheners,⁴² while a European collective expert assessment concluded that certain noncombustible air fresheners may cause or aggravate symptoms in highly sensitive persons and may be of concern in children.⁸ Surveys conducted in five industrialized countries (the United States of America, United Kingdom, Germany, Australia and Sweden) indicated that 15% to 20% of people have reported health effects, e.g., headaches or breathing difficulties, associated with the use of noncombustible air fresheners.^{7,43,44}

Priority substances of interest

Based on all the calculated risk indicators, the substances of highest interest identified in the HRA are acrolein, benzene, PM_{2.5}, formaldehyde, and, to a lesser extent, limonene.

These substances are not specific to the emissions of noncombustible air fresheners. Other emission sources are commonly present in indoor environments, e.g., furniture, cleaning products, tobacco smoke, construction products, paints, and cooking fumes. Consequently, cumulative exposure is expected, which could lead to higher risks than those identified for each source, considered individually. Background exposure levels in European private dwellings are available for several of the above identified priority substances:^{19,31,45} formaldehyde (minimum: 7 µg/m³; maximum: 57 µg/m³; mean: 22 µg/m³); benzene (minimum: 0 µg/m³; maximum: 32 µg/m³; mean: 3 µg/m³); limonene (minimum: 0 µg/m³; maximum: 493 µg/m³; mean: 29 µg/m³). The health reference values selected in this HRA, and those considered in the Discussion section, relate to the total indoor air concentration: they do not relate to the concentration attributable to the use of noncombustible air fresheners only. This suggests a need to limit the emissions of noncombustible air fresheners, especially for the identified priority substances of interest, until the corresponding concentrations are much lower than the selected health values.

Emitted particles

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

However, a portion of the PM_{2.5} particles emitted by the tested noncombustible air fresheners were submicron particles (PM₁), even ultrafine particles (PM_{0.1}), which is consistent with the results of other studies^{23,46-48} and certain descriptions of manufacturers. For example, a spray and active

diffuser yielded a 1-hour PM₁ concentration in excess of 3 µg/m³; singularly among the tested products, one active diffuser (atomizer) resulted in a 1-hour PM₁ concentration of approximately 88 µg/m³. These submicron particles are a subject of concern because of their high surface reactivity and ability to penetrate the pulmonary system.^{49,50}

Conclusions

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

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

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

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

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

The acquired results under the reasonable worst-case scenarios suggest that the highest chronic exposure levels should be reduced, especially for acrolein (up to 3.5 µg/m³) and benzene (up to 4 µg/m³).

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

The emitted $\text{PM}_{2.5}$ particles could also be of concern because the acute exposure level reached 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.

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

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Tables

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

Rooms	Volume (m ³)	Daily presence (h/d)
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

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 _{2.5}	-	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 _{2.5}	-	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 _T (µg/m ³)	Org.	Date	TRV _{NT} (µg/m ³) [†]	Org.	Dated	TRV _{acute} (µg/m ³)	Org.	Date	Critical effects
50-00-0	Formaldehyde	-	-	-	-	-	-	1.0.10 ²	WHO	2010	Acute: Subjective and objective eye irritation
75-07-0	Acetaldehyde	1.6.10 ²	ANSES	2014	-	-	-	3.0.10 ³	ANSES	2014	Threshold: Degeneration of the olfactory epithelium Acute: Bronchoconstriction in individuals with asthma
107-02-8	Acrolein	1.5.10 ⁻¹	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 ⁴	ANSES	2018	-	-	-	2.0.10 ⁴	ANSES	2018	Threshold: Neurological effects (color vision disorders) Acute: Neurological effects
91-20-3	Naphthalene	1.10 ¹	WHO	2013	5.6.10 ⁻⁶	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 ⁻³	USEPA	2017	6.0.10 ⁻⁴	US EPA	2017	-	-	-	Threshold: Increased fetal embryonic mortality Nonthreshold: Occurrence of respiratory tumors
71-43-2	Benzene	1.0.10 ¹	ANSES	2010	2.6.10 ⁻⁵	ANSES	2013	2.7.10 ¹	OEHHA	2014	Threshold: Immunological disorders Nonthreshold: Acute leukemia Acute: Reproductive disorders, aplastic anemia and acute myeloid leukemia

Notes:

- † TRV_T is the threshold toxicological reference value, for the inhalation route and chronic exposure;
- † TRV_{NT} is the nonthreshold toxicological reference value, for the inhalation route and chronic exposure;
- † TRV_{acute} 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.