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perception of liquid electric fragrance diffusers

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An assessment of VOC emissions and human strength perception of liquid electric fragrance diffusers†

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Fragrance products are commonplace in everyday life and their air quality effects extensively studied. In this study the use of multiple plug-in diffusers (liquid electricals/LEs) was assessed by quantifying air concentrations in controlled test rooms ('toilet booth' and 'large booth') with up to 5 LEs of known formulation in concurrent use. Olfactive strength tests were completed under the same conditions in blind assessments. Air samples were analysed using thermal desorption – (TD) gas chromatography (GC) coupled to flame ionisation (FID) and mass spectrometry (MS) detectors. Significant positive linearities were found for several VOCs (*e.g.* α-pinene toilet booth and large booth $R = 1$ and $p = 0.0028$, *p*-cymene toilet booth $R = 0.94$ and $p = 0.017$, large booth $R = 0.89$ and $p = 0.033$), with α-pinene presenting the highest measurable gas-phase concentration (mean 25 μg m^{-3}, toilet booth with 5 LEs present). All measurable linearities for fragrance species in toilet booths were significant. However, olfactive intensity assessment showed a plateauing in fragrance perception after the addition of 2 LEs. Only very volatile fragrance ingredients such as α-pinene and benzaldehyde could be detected in the gas phase, but at ambient concentrations that were always lower than literature values for their individual odour detection thresholds. The plateauing of the perception of fragrance strength may aid in limiting potential off-instruction use, thereby limiting end-user exposure to potentially high concentrations of emitted VOCs. The drivers of human fragrance perception here appeared to be lower vapour pressure constituents of fragrance formulations like sesquiterpenes. Moderation of the concentration of monoterpenes used in room fragrance formulations may be a practical solution in limiting possible air quality impacts of product use, whilst still maintaining end-user fragrance perception.

Environmental significance

An assessment of human fragrance strength perception and atmospheric concentrations has been made using commercially available liquid electrical (LE) fragrance diffusers in climate-controlled rooms. VOC concentrations increased linearly as up to 5 additional LEs were added to each room but with reported fragrance perception plateauing after the addition of two devices. Monoterpene concentrations remained below odour detection thresholds even for 5 LE devices, suggesting olfactory perception was a result of less volatile ingredients. A lack of perceived increase in fragrance perception may potentially lead to a self-limitation in product use in indoor settings.

1. Introduction

Fragrances have been used by humans since antiquity and are well-documented in many different cultures and regions through history, and the use of fragranced products is commonplace in 21st century life.[1–4] Common gas phase emissions from fragranced products include monoterpenes, alcohols and esters.[5,6] Monoterpenes are also emitted from many other sources indoors including from flowers, fruit, cooking and cleaning.[7–10] Since they are relatively reactive to oxidation indoors they are potential precursors to the production of secondary organic aerosols (SOAs).[9,11–13] Monoterpenes are typically included in fragrance formulations for their individual fragrance notes and are known to be important in the perception of fragrance.[14,15] Through electroencephalography, common emissions from fragrance products such as limonene and terpinolene can be linked to

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changes in brain function, including an increased perception of relaxation and pleasure in humans.[16–18]

Monoterpenes are only one of many classes of compound found in fragranced products,[19–21] however they are often some of the most volatile present and hence generate some of the highest gas-phase indoor concentrations indoors when a product is used. They are more amenable to analysis by methods such as thermal desorption gas chromatography mass spectrometry (TD-GC-MS) or on-line mass spectrometry than less volatile fragrance components such as terpene oxides and sesquiterpenes. Some monoterpenoids, such as geraniol and borneol, while commonly described as 'volatile', have vapour pressures comparative with sesquiterpenes and as such are often not easily quantified. There is an understandable attraction in air quality science for using gas-phase measurements of monoterpenes as a proxy for the presence of fragrance, despite many non-fragrance sources of these species also existing indoors. Those species which likely impact substantially on human perception of fragrance are often not detectable in ambient air due to very low gas-phase concentrations and vapour pressures.

Fragrance diffusers belong to a group of home fragrance products that actively emit into an air space. This is commonly through passive diffusion, *e.g.* using wooden sticks/reeds to enhance evaporation, or using an electrically powered element (commonly called 'plug ins', referred to commercially as liquid electricals/LEs). LEs can be further differentiated into those that use a heated wick to actively diffuse fragrance into the airspace, or so-called 'nebulisers' which aerosolises fragrance oil. LEs are designed to deliver a constant rate of emission of volatile organic compounds (VOCs) and semi-volatile organic compounds (SVOCs) associated with the scent.[22] The concentration in air of each VOC/SVOC emitted is dependent on the fragrance formulation and compound vapour pressure.[10,23] There has been some research into the differences in VOC emissions arising from passive and plug-in fragrance diffusers; the formulation of fragrance generally having more influence than the diffuser type.[22,24] Using a constant-emission fragrance device within an airspace leads to an initial increase in VOC concentration before reaching a steady state concentration that is determined by the balance of emission with loss processes including fresh air ventilation rate, surface deposition and in-room oxidation.[23,24]

Evaluation of the indoor air effects of LE devices has predominantly been conducted in small laboratory test chambers; these generate gas phase concentrations much higher than are seen in real-world home settings.[22,25–27] Test chambers allow for greater experimental control, such as chamber air exchange rate (AER) and chemical composition of diluent air.[28] Evaluating effects of LEs on indoor air quality and in human perception of fragrance is complicated by pre-existing VOCs from other sources and widely different product use behaviours and varying air exchange rates.[26,29–31] Warburton *et al.* (2023)[26] showed that incremental indoor VOC increases from using a single LE in real homes were difficult to discern and that air ventilation and location in the home (and associated properties such as room size) were key factors.[32–34] Ventilation in homes is highly variable but is generally reducing in residential

properties as they are made more energy efficient. Fragranced products including LEs must perform well from an environmental and human perception perspective in both old and new/retrofitted housing stock.

Any solvent-containing product (*e.g.* paints, glues, aerosols, adhesives) used in a manner that deviates substantially from its intended method of application has the potential for health harms.[35–39] This might occur, for example if users do not follow labelling instructions for frequency/duration of use, quantity/amount of product intended to be used, or ventilation requirements. Theoretically high concentrations of fragranced-derived VOCs could be generated indoors if numerous products (or multiples of the same product) were used off-instruction, such as in small rooms that had poor ventilation. Fragrance species are one constituent within a complex matrix of chemicals within product formulations however, and concentrations of each must be borne in mind while assessing safety of use.

VOCs associated with fragrance possess an odour detection threshold (ODL). This is defined as the minimum concentration required for a human to reliably perceive the presence of the VOC. Any human-based olfactive assessment has the potential for uncertainties given person-to-person variability, and as such ODLs can change between studies.[40–42] The ODL of a compound depends greatly on its shape, size, vapour pressure, polarity and reactivity.[43,44] Certain malodours are detectable in low atmospheric mixing ratios, while some commonly used ingredients such as α-pinene have ODLs orders of magnitude above these thresholds.[45,46] There has additionally been shown to be some moderate discrepancies in ODL between younger and older adults, with younger adults tending to have lower VOC-specific ODLs.[47] While ODLs may differ and vary person-to-person, they provide a useful benchmark for assessing the impact of VOC-releasing products on user perception. However, the use of ODLs in comparison to indoor VOC concentrations originating from product emissions remains relatively understudied.

1.1 Objectives

This study aims to uniquely combine the indoor air quality effects arising from using a well-controlled LE source in realistic but controlled rooms and the olfactive strength of that source. It remains a possibility that the non-standard, off-instruction and excessive use of any VOC-containing products may lead to unintentional harm in the home, with the most significant hazards arising where there is poor or no olfactory detection of the emitted VOCs. The indoor concentrations of odourless VOCs such propane or butane might reach many hundreds of parts per million before being perceived. In these worst cases, there is a risk of fatality with longer-term chronic exposure potentially leading to sub-acute effects.

Fragrance VOCs and SVOCs can be perceived at low ambient concentrations,[48,49] and so here we assess the VOC emissions from LEs coupled with fragrance strength perception by trained sensory panellists. It is hypothesised that excessive and/or off-instruction use of LEs, such as using multiple LEs simultaneously in rooms with low AERs, is less likely to occur than for other household VOC-containing products, since usage may be

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attenuated by human tolerance of the resulting odours created. This was evaluated using controlled test booths of different sizes for in-room monitoring of VOC concentrations in addition to human olfactory testing. A comparison to real world indoor air studies measuring VOC contributions from LEs was also conducted to assess the ability to apply these artificial room studies as a model for estimating normal consumer home environments. The combination of indoor air quality measurements with olfactory evaluation is undocumented in research literature to our knowledge. Assessing personal responses to VOCs is technically challenging owing to person-to-person variance in perception and odour detection threshold, different effects of fragrances to physiological responses, pre-existing expectations of product performance, and time required in establishing robust methodology to account for these possible person-to-person discrepancies.[17,50–52] Here we combine state of the art fragrance industry testing protocols, a trained sensory panel and indoor air quality measurements to assess strength perception of a typical fragrance releasing household product.

2 Methodology

The following sections give detailed explanations of the methodological processes followed in this study, while summary flowcharts can be found in the ESI Fig. S1.†

2.1 Canister preparation

Whole air samplers were used to collect in-room samples using 6 L vacuum-intake stainless steel canisters internally treated with silica (Entech, CA, USA) using flow-restrictive inlets (Entech, CA, USA) resulting in samples being drawn over 72 hours, found by Heeley-Hill *et al.* to be linear over the first 48 hours with a reducing flow rate over the last 24 hours.[29] Prior to deployment, canisters were evacuated to 0.01 Pa, or 29.9 Hg vacuum (gauge). Canister valves were checked prior to deployment to qualitatively assess valve seal integrity by attaching a vacuum gauge atop the closed canister valve and leaving for at least 2 hours. Failed valves were replaced like-for-like, and the evacuation and valve integrity process repeated.

2.2 Sample collection and preparation for analysis

Evacuated canisters were placed in one of two laboratory test rooms ('booths') at Givaudan Ltd, Ashford, UK, either designed to mimic a small bathroom (W 1.92 m $\times$ D 1.90 m $\times$ H 2.6 m) or a medium-sized living room (W 3.0 m $\times$ D 4.45 m $\times$ H 2.49 m). These were mechanically ventilated with outdoor air with an air exchange rate of 7.5 h^{-1}. Top-down schematics of the large booth and the toilet booth are given in the ESI Fig. S2(a) and (b),† respectively. Between 1 and 5 identical LEs of known formulation were placed together in each of the rooms. Canisters for sampling were placed at identical distances from the LEs. LEs were turned on 24 hours before the sampling period began to aid equilibration of output and for mixing of the airspace; after 24 hours the canisters would begin sampling. After 72 hours had elapsed, the valves were closed, and when

sampling had finished across all booths, the canisters were returned to University of York for analysis.

LE emissions were measured through oil mass loss over the sampling period. LEs were weighed both pre- and post-sampling. Mass loss ranged between 1.8 g and 3.7 g with a standard deviation of 0.490 g. Mean oil mass loss was 2.66 g, and median oil mass loss was 2.54 g. Commercial comparable LEs draw between 2 and 4 W of power, and the LE used here drew 2.8 W (UK 230/240 V, AC current with a frequency of 50 Hz).

An equal number of samples were taken for each LE multiple (between 1 and 5). Controls were taken of empty booths over the same time-period to allow for background (fresh air ventilation) air composition to be quantified.

2.3 Sample analysis

Filled canisters were pressurised to 1 bar (gauge) with highly purified air free of VOCs, generated from a custom-built thermal catalytic oxidiser with compressed ambient air passed over platinum beads at 400 °C to fully oxidise any VOCs in the compressed air (hereafter referred to as 'blank gas'). Samples were analysed following the method detailed in Warburton *et al.*[26] In brief, 500 mL of canister air was drawn through a 16-port Valco electrically-actuated multi-position valve (VICI Valco Instruments Co. Inc., TX, USA) into a custom-built thermal desorption unit (TDU), comprised of (sequentially) a water trap, a sample pre-concentration trap and finally a pre-injection focus trap. The first GC column was a 60 m long, 150 μm internal diameter (ID) VF-WAX column with a film thickness of 0.50 μm (Agilent Technologies, CA, USA) at a flow rate of 1.6 mL min^{-1} (carrier gas pressure of 35 psi). The early eluting unresolved analytes (C_2 to C_6 hydrocarbons) were passed to an Na_2SO_4-deactivated Al_2O_3 porous-layer open tubular (PLOT) column (50 m $\times$ 320 μm ID, with a film thickness of 5 μm, Agilent Technologies, CA, USA), through a Deans switch (Agilent Technologies, CA, USA) with detection *via* flame ionisation detection (FID). After 8.3 minutes, the Deans switch diverted the analyte flow through a section of fused silica (2 m $\times$ 150 μm ID) to both balance column flows at the Deans switch and split analyte flow between the second FID and the quadrupole mass spectrometer (QMS) for simultaneous detection, through sections of 150 μm ID fused silica of length 0.91 m and 2.1 m, respectively. GC elution data was acquired through MassHunter Qualitative Analysis (Agilent Technologies, CA, USA). The QMS ionisation was achieved through electron impact (EI) ionisation, with the QMS scanning for ions with an m/z between 30 and 150 units. Ion source temperature was 230 °C and an electron energy of 70 eV, with a quadrupole temperature of 150 °C.

Sample calibration was achieved by using a thirty-component mix of non-methane hydrocarbons (NMHCs) in nitrogen, with each gas at a mole fraction of approximately 4 ppb, provided by the National Physical Laboratory, Teddington, UK (cylinder number D933515, hereafter referred to as 'NPL 30') for VOCs contained therein, with remaining VOCs calibrated using equivalent carbon responses (ECN), using *n*-heptane as the proxy for ECN calculations. Blank gas was sampled three

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times after each canister sequence to confirm complete sample transfer and to allow for any artefacts to be corrected. After the blank gas samples, five NPL 30 calibrations were run, followed by three carrier gas/internal samples ('no flow blanks'). The no flow blank method resembled a regular canister sample method, however no sample was drawn (sample fill volume was set to 0 mL), but the carrier gas flow rate and flow time remained unchanged, resulting in an equal volume of carrier gas flowing through the TDU, and subsequently the GC, as would occur in a regular sampling method. Combined, this allowed for canister samples to be corrected for blank gas diluent contamination (none seen) and carrier gas, or system, contamination (consistently 0.95 µg m^{-3} benzene only). Canisters would then be re-evacuated according to the previously described method. Canisters were randomly selected to check for contamination by filling from fully evacuated to 1 bar (gauge) with blank gas and run according to the previous method. Instrument limits of detection (LOD) and quantification (LOQ) for the six fragrance-originating VOCs in this paper are given in ESI Table S1.† LODs were calculated using a signal-to-noise ratio (SNR) of 3 : 1, and LOQ an SNR of 10 : 1.

Chromatogram peak integration was completed using *GCWerks* (GC Soft Inc., CA, USA). Over 120 VOC species were identified and included in the automated analysis. Only the VOCs associated with the LE formulation are reported in this paper to simplify the presentation of results, however the entire VOC dataset is open-access from the Centre for Environmental Data Analysis (CEDA) repository at **https://www.ceda.ac.uk/**.

2.4 Data visualisation and statistical analysis

All data analysis and manipulation were conducted using the R language, through RStudio software. The *tidyverse* package was used in all data processing. Data plotting used *ggplot2* for all figures except quantile–quantile plots (qq-plots) for data normality which used *ggpubr*. Boxplots show values in the order of (from bottom-to-top): lower outliers, 5th percentile, 25th percentile, median value, 75th percentile, 95th percentile, and upper outliers. Regression statistics were calculated using Spearman's rho using *stat_cor()* from the *ggpubr* package. A confidence interval of 95% ($\alpha = 0.05$) was used to indicate significance. Diffuser increment plots were produced using the *raster* and *contour_filled* functions in *ggplot2*, with additional contour lines and contour line labels added using the *contour2* and *geom_text_contour* functions in the *metR* package.

For statistical analysis of VOC concentrations difference arising from different numbers of LE used in each booth, all data were first transformed by natural logarithm. Following this a Kruskal–Wallis test was completed on the transformed data with a *post hoc* Dunn tests for species which returned a positive result for significance following the initial Kruskal–Wallis test, indicated by a *p*-value lower than the $\alpha = 0.05$ level. Statistical analysis methodology for the olfactive results is given later.

2.5 Olfactory methodology

Human olfactory testing was completed using a group of trained panellists from a sensory panel at Givaudan, UK. The members of the panel were selected based on their olfactory sensory acuity and then trained for a period of 4–6 months to enable them to discriminate between products and score consistently. The sensory panellist training programme at Givaudan Inc., UK consists of several stages including: (1) discrimination through triangle tests, to enhance skills for differentiating between different odour types or odour intensities, (2) ranking, to develop skills for comparing the relative strength of sample, (3) scaling, to develop the ability to quantify differences between samples and (4) scoring against a control, to further develop the ability to use the scale. The trained panellists assessed the strength of the LEs from 10 m^3 toilet booths, following the same LE usage as with VOC sampling.

Samples were monadically assessed blind and sequentially. Panellists were not aware of the purpose of the investigation. The trained panellists were asked to rate the overall perceived intensity of the fragrance inside the 10 m^3 toilet booths using a linear 0–100 scale. Sample orders were randomised using a Latin square design to control experimental error. Replicates were included within the sample set, and panellists were not told there were replicates within the assessments. Sampled panellist data was assessed for reliability using published methods in Talsma (2016).[53] Statistical analysis was completed using analysis of variance (ANOVA) followed by a Benjamini–Hochberg *post hoc* assessment to observe significance between levels. The ANOVA was additionally used to assess and ensure no significant interaction between products and replicates. As with sample VOC analysis, a confidence interval of 95% ($\alpha = 0.05$) was used to indicate significance. Panellists sampled the LE scent through a porthole from the chamber so as to not interfere with booth airspace composition, and panellists were unaware of the number of LEs present within the booth.

2.6 VOC metrics and modelling

In this paper, the metric "TVOC" is defined as the sum of all quantifiable VOCs within a sample, making it an operationally defined term specific to the analytical methods used. Additionally, we introduce the metric "fragrance TVOC," which is operationally defined as the sum of the concentrations of six VOCs: α-pinene, β-pinene, γ-terpinene, benzaldehyde, *p*-cymene, and eucalyptol. While this metric does not represent the entire fragrance formulation, it includes all fragrance-origin VOCs that were sampled and resolved using our methodology. The "fragrance TVOC" metric was adopted for brevity when describing the behaviour of VOCs originating from the LE fragrance. In specific cases, discussed later, individual fragrance VOCs were highlighted, particularly when some compounds were missing from samples or fell below detection limits.

Warburton *et al.* (2023)[26] reported a simple method for determining a steady-state increment of a species with a known emission rate, shown in eqn (1)

$$C = \left(\frac{q}{A_{\mathrm{H}} V}\right) 10^6 \qquad (1)$$

where C is the concentration of the species within an airspace (μg m^{-3}), q is the emission rate of the species (g h^{-1}), A_H is the air exchange rate (h^{-1}) and V is the room volume (m^3). This equation assumes a steady-state airspace in a well-mixed, one-box compartment with no chemical loss. In this study a more complete model was used which solves the ordinary differential equation (ODE) given in Carslaw (2007),[54] shown in eqn (2), with some term symbols changed to match those used in eqn (1). For the analysis in this study, α-pinene was chosen to model concentrations, as it represented a considerable portion of the volatile fraction within the LE oil itself.

$$\frac{dC_S}{dt} = -V_d\left(\frac{A}{V_C}\right)C_S + \lambda f C_{out} - \lambda C_S + \frac{q_S}{V_C} + \sum_{j=1}^{n} R_{ij} \qquad (2)$$

where C_s is the concentration of the species within the airspace (molecules per cm^3), t is time (s), V_d is the deposition velocity of the species (cm s^{-1}), A is the surface area of the room (cm^2), V_C is the volume of the room (cm^3), λ is air exchange rate (s^{-1}), f is the outdoor-to-indoor penetration factor (dimensionless), q_s is the species emission rate (molecules per s), and R_{ij} is the rate of reaction between species i and j (molecules per cm^3 per s). In this study, it was assumed there was no surface deposition, that the outdoor-to-indoor penetration factor was equal to 1 as in Dimitroulopoulou *et al.* (2001),[55] and chemical removal was *via* oxidation reactions with OH and NO$_3$ radicals, as well as with O$_3$. Reaction rates were calculated for the oxidation of α-pinene to form APINAO2, APINBO2, APINCO2, NAPINAO2, NAPINBO2, APINOOA, and APINOOB, rate constants for which were obtained using the Master Chemical Mechanism (MCM) *via* http://www.mcm.york.ac.uk.[56,57] The ozone fraction was originally set to 10 ppb to give a model for α-pinene increment expected in residential homes, but was set to 35 ppb to be reflective of the air make-up within the booths used in this study. OH and NO$_3$ radicals were given maximum concentrations of 2×10^6 molecules per cm^3 and 9.2×10^8 molecules per cm^3 respectively, in a sinusoidal circadian rhythm, with OH concentrations peaking at 12 pm and NO$_3$ concentrations peaking at 2 am. Low concentrations used for OH and NO$_3$ were 1×10^5 molecules per cm^3 and 1×10^6 molecules per cm^3, respectively. NO$_3$ radicals, while important in VOC oxidation, do not typically see very high indoor concentrations and are normally not a major consideration for indoor modelling.[54] However, in this study the booths were supplied with outdoor air throughout, and additionally were allowed to equilibrate for one day before sampling began. As such the air makeup in the booths over the sampling period was treated as if it were outdoor air and subject to circadian outdoor behaviours.

The rooms are based at a large industrial facility in Ashford, UK where similar sensory science investigations take place and were compared against available data for comparable locations and times of year when samples were obtained, with ozone fractions taken from http://www.gov.uk/government/statistical-data-sets/env02-air-quality-statistics, and OH and NO$_3$ radical concentrations used here were influenced by various studies.[58–62] This model, which assumes a well-mixed one-box compartment, was run for each emission rate with a 5 second

resolution over one day of constant emission and was then iterated over a variety of air exchange rates and volumes to give an array of simulations for each LE emission load. As each simulation progressed, the room concentration of α-pinene reached a constant concentration, indicating the simulation had reached a steady-state. The simulated steady-state concentrations were then plotted as a z-axis colour contour against room volume and air exchange rate.

Using eqn (2) to model indoor VOC concentrations provided a more comprehensive prediction than eqn (1), as it accounted for VOC loss through various pathways, most importantly oxidation. Monoterpenes and monoterpenoids, which are highly reactive indoors, underwent oxidation particularly with ozone, as well as with nitrate and hydroxyl radicals. This oxidation served as a substantial sink for these compounds. For upper-bound estimates, eqn (1) provided a straightforward method for approximating concentrations; however, for this more detailed analysis of one specific VOC, eqn (2) yielded a fuller representation.

3 Results and discussion

3.1 TVOC and fragrance species concentrations

Boxplots of fragrance TVOC concentration at discrete levels of LEs (0–5) are shown in Fig. 1. As would be expected, the VOC concentrations in the smaller booths were higher than in the larger room, however both increased linearly as the number of LE devices increased. Fig. 2(a) shows the change in mean individual species concentrations and linear regression statistics for the large room as number of LE devices increases, with Fig. 2(b) showing the same for the small booth. Spearman's Rho was used in the calculation of regression statistics, and R and p-values are given on each plot. Significant linearities were found for α-pinene and p-cymene for the large booth, and all quantifiable species in the toilet booth. The effect of additional LEs in the large booth was more scattered and at lower concentrations than the toilet booth. Neither showed any indication of concentration plateauing at a high number of concurrent LEs. Note that significance in linearity does not reflect significance in the pairwise comparison of concentrations across LE levels, which is discussed later on. Benzaldehyde 0 LE concentrations in the large booth appeared to be a potential outlier, as this higher-than-expected concentration skewed the linearity significantly. Ignoring the 0 LE result, LE additions resulted in a linear and roughly stepwise trend. Replacing this outlier with the 0 LE benzaldehyde concentration from toilet booth data resulted in a significant linearity with $R = 1$ and $p = 0.0028$ (ESI Fig. S3†).

It has been previously reported that VOCs with lower vapour pressures tend to have lower human odour detection limits (ODLs).[63,64] Value-based definitions of VOCs vary, but vapour pressure-based definitions separating VOCs from lower-vapour pressure species is generally between 0.075 mmHg and 0.1 mmHg.[65,66] Tamas *et al.* (2006)[67] demonstrated a plateauing of perception of limonene (vapour pressure/V_p = 1.5 mmHg) within test chambers up to 115 ppb, or approximately 641 μg m^{-3}. The same study also reported an ODL for limonene of

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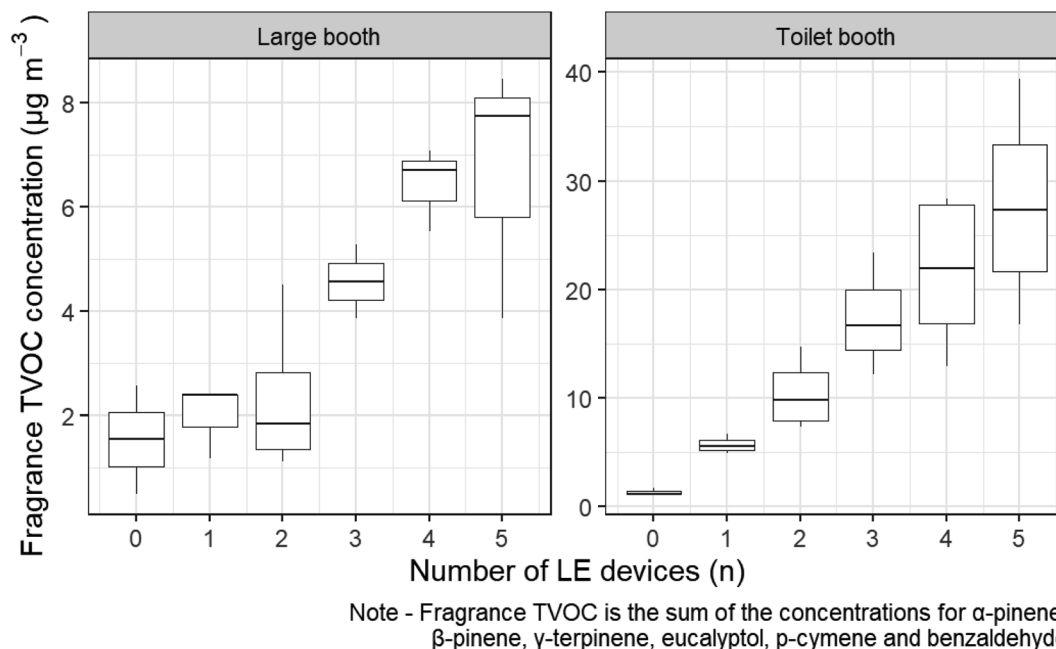*Environ. Sci.: Adv.*, 2025, **4**, 739–752 | **743**

Fig. 1 Boxplots of the spread of fragrance TVOC concentrations in the large booth (left panel) and toilet booth (right panel). From bottom to top, each boxplot shows 5th percentile, 25th percentile, 50th percentile/median, 75th percentile, 95th percentile. Any outliers are given as single dots above or below the 5th/95th whisker.

around 40 ppb, or approximately 220 μg m^{-3}. Yoshio *et al.* (2003)[46] reported a limonene ODL of approximately 40 ppb also, and an α-pinene (V_p = 4.75 mmHg) ODL of 18 ppb, or roughly 101 μg m^{-3}, with a β-pinene (V_p = 2.93 mmHg) ODL of

approximately 33 ppb, or roughly 184 μg m^{-3}. Yoshio *et al.* (2003)[46] also report the ODL for the sesquiterpenoid geosmin (V_p = 0.003 mmHg) at 6.5 ppt, or 0.05 μg m^{-3}, and Schoenauer and Schieberle (2016)[68] reporting an ODL for grapefruit
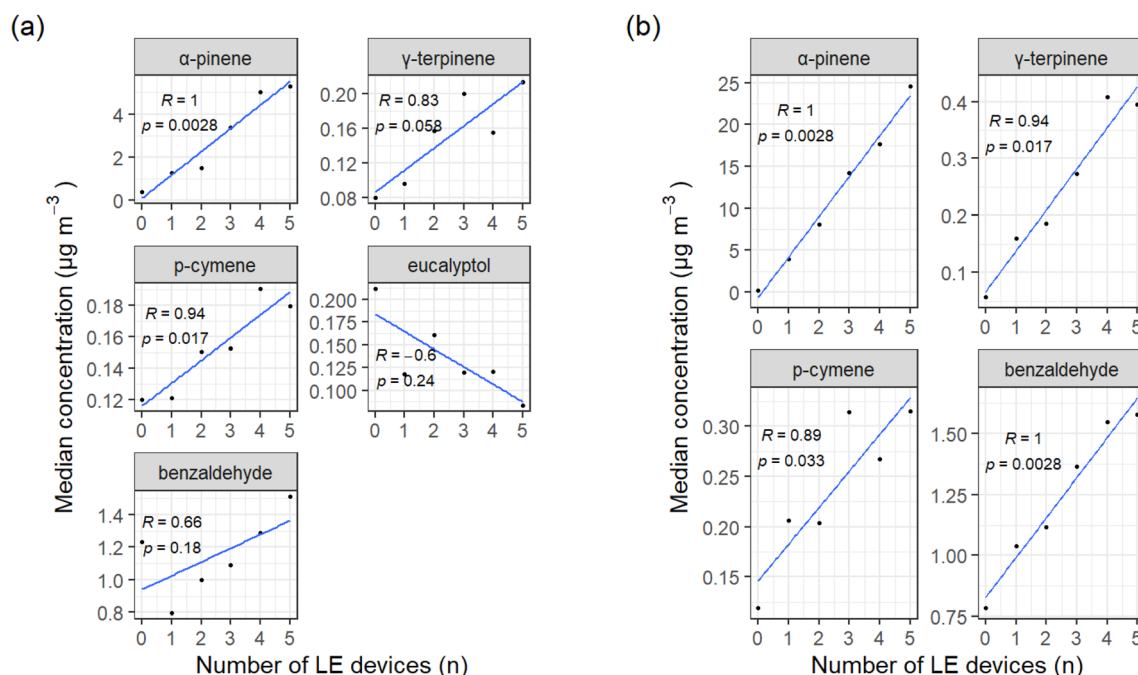

Fig. 2 (a) Scatter plot showing median concentration results for fragrance species at each LE level (0 to 5) for the large booth, and (b) the same as (a) but for toilet booth results. Concentrations are given in μg m^{-3}. Regression statistics were calculated using Spearman's Rho. Note – β-pinene plots are missing from both (a) and (b) as all values were below the LOQ for β-pinene. Eucalyptol is missing from (b) due to only having values above LOQ for LE levels 5 and 6, resulting in an incomplete plot.

mercaptan (V_p = 0.1 mmHg) of 0.0049 ppt, or approximately 0.000034 µg m^{-3}. All vapour pressure values were obtained from The Good Scents Company (http://www.thegoodscentscompany.com/). As described in later sections of this paper, toilet booths were perceived by sensory panellists to have detectable fragrance odour when any number of LEs (1–5) were present, however the 'measurable' VOC component of the fragrance (monoterpenes and other higher vapour pressure species) were at concentrations below their individually reported ODLs, suggesting that the detectable odour arose from less volatile species from the fragrance formulation such as γ-undecalactone and β-caryophyllene – likely present at ppt and sub-ppt mixing ratios and below measurable limits.

3.2 Comparison of LE emission rates

LEs were weighed before and after sample collection to determine oil mass loss over the sampling period. Median diffuser mass loss was 2.54 g, average mass loss was 2.67 g, with a standard deviation of 0.50 g. Using mean mass loss, this resulted in an overall LE oil emission rate of 0.028 g h^{-1}. Using the known fragrance formulation this gave an α-pinene emission rate of 1.60×10^{-3} g h^{-1}. The LE emission rate was compared against available literature, shown in Table 1. There was little literature available which disclosed LE emission rates directly or which contained data from which an emission rate could be calculated. Values were taken from Warburton *et al.* (2023),[26] Angulo-Milhem *et al.* (2023)[24] and Angulo-Milhem *et al.* (2021).[23] In Angulo-Milhem *et al.* (2021)[23] it was noted the LE was placed on a medium intensity, resulting in an emission rate of 0.044 g h^{-1}. It is common for commercial LE devices to have both variable and fixed output settings, however the LE used in this study had a fixed output. Given the lack of available emission rate statistics of LEs in literature, it was not possible to contextualise these emission rates, however the impact of a higher oil emission rate would be seen in both elevated concentrations of emitted VOCs, as well as increased fragrance perception. Section 3.4 discusses the estimation of indoor concentrations from known emission rates and sources.

3.3 Perceived intensity of LEs

Trained sensory panellists were asked to give an overall perceived intensity rating from 0 to 100 for the toilet booths, containing between 0 and 5 LEs. Panellists were blind to the number of LEs present in the room during each assessment. Fig. 3 gives mean results from the difference in perceived fragrance intensity as pairwise values. A stepwise increase in LE
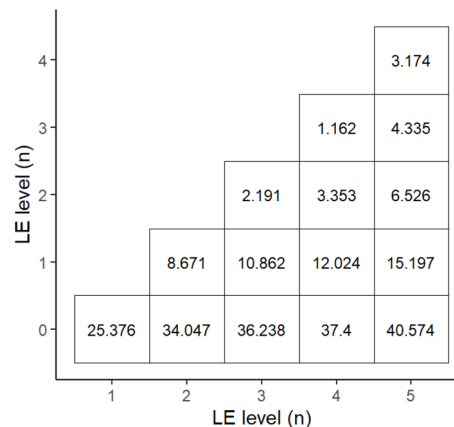

Fig. 3 a matrix which shows the mean change in perceived fragrance intensity at each combination of LE level from a scale of 0 to 100, 0 being the lowest/no change in perceived intensity and 100 being the highest.

number, *i.e.* from 1 to 2 to 3 *etc.*, resulted in increasing small perceived differences in odour intensity once the first LE was added. The most marked increase in step-wise perception of change in fragrance odour was between rooms containing 0 and 1 LE. Testers identified limited differences in their perception of the fragrance intensity between 2 and 3 or more devices.

For olfactive results, pairwise analysis was completed on difference in perceived intensity using an Analysis of Variation (ANOVA) test, with a Benjamini–Hochberg *post hoc* assessment, *p*-values for which are shown as a matrix in Fig. 4(a). These results confirmed a step-wise incremental increase in significance for the addition of LEs up to 2, after which the significance of the difference in perceived intensity drops and did not give rise to further significant increase in perceived fragrance intensity.

Fragrance TVOC concentrations were assessed across LE levels using a Kruskal–Wallis test, which returned a positive stochastic result for significance. All samples were used in this assessment, and the data were transformed by natural logarithm prior to Kruskal–Wallis analysis. A *post hoc* Dunn test was then performed, which gave the *p*-value results shown in Fig. 4(b). Pairwise analysis of fragrance TVOC concentrations yielded insignificant results when comparing a stepwise increase in LE number, with significant pairwise increases in fragrance TVOC concentration only found when multiple LEs were added into the airspace at once.

The lack of significance in perception of change in intensity may lead to a real-world consequence where the use of LEs in

Table 1 Comparison of LE emission rates and booth properties between this study and several other studies available in literature

	This study	Warburton *et al.* (2023)[26]	Angulo-Milhem *et al.* (2023)[24]	Angulo-Milhem *et al.* (2021)[23]
Diffuser emission rate (g h^{-1})	0.028	0.024	0.053	0.044
Chamber volume (m^3)	33.73 (large booth) 9.49 (toilet booth)	—	40	1
Air exchange rate/AER (h^{-1})	7.5	—	0.3 ± 0.1	0.3 ± 0.05

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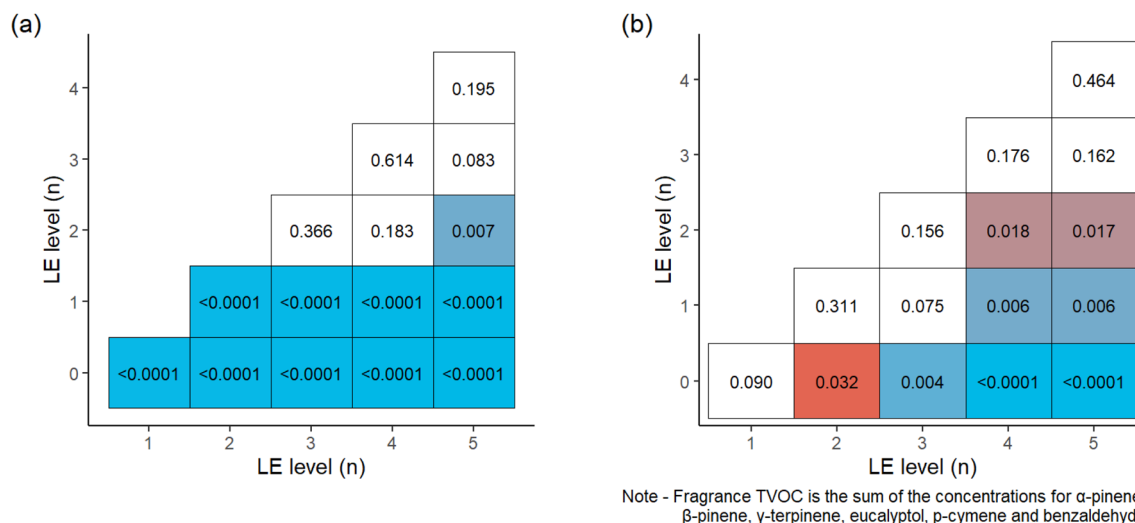*Environ. Sci.: Adv.*, 2025, **4**, 739–752 | **745**

Fig. 4 (a) Matrix showing p-value results for each LE level pair following ANOVA and Benjamini–Hochberg *post hoc* analysis for fragrance perception, and (b) a matrix which shows p-value results for each LE level pair following Kruskal–Wallis and Dunn *post hoc* analysis for fragrance TVOC concentrations. Non-significant results are given in white, and results of significance graduate from red ($p = 0.05$) to blue ($p \to 0$).

a single room is to a degree self-limiting because of limited perception of further benefit to the user if additional LEs are added. We however note that other factors such as user tolerance and fragrance acceptability will also contribute to end-user-based limitation of LE use, and future work would be required to identify whether LE use could be labelled as self-limiting. In the larger room and with the maximum of 5 LEs operating, a median α-pinene concentration of 5 µg m^{-3} and an upper α-pinene concentration of 7 µg m^{-3} was generated. This is a surprisingly low concentration given the number of LEs present. The concentration of α-pinene in the larger room was typical of concentrations (within the 50$^{\text{th}}$ percentile) found in

real-world homes with comparable room sizes in the work of Heeley-Hill *et al.* (2021)[29] and within the 60$^{\text{th}}$ percentile of homes in Warburton *et al.* (2023).[26] Monoterpene emissions are undoubtedly associated with the use of fragranced products and are often species with the higher emission fractions within product formulations such as in air fresheners, surface cleaners and shampoos.[69,70] However even in an exaggerated product perturbation experiment the concentrations of monoterpenes generated in realistically sized and ventilated rooms are low, hence their presence or absence may not be a good indicator for whether a room would have perceptible fragrance odour.
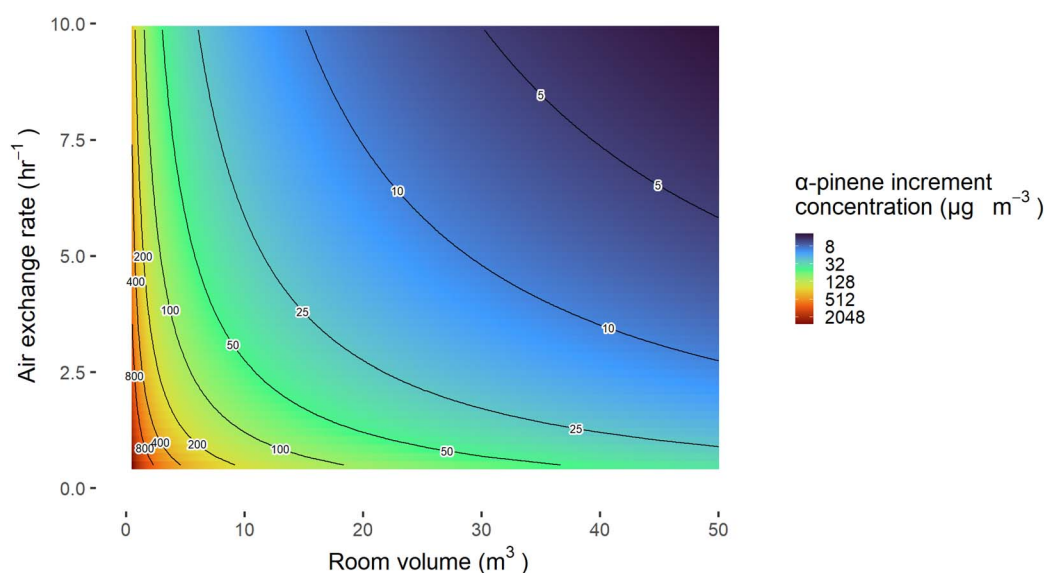

Fig. 5 Raster plot showing expected increment concentration of α-pinene in µg m^{-3}, against ventilation rates in units of h^{-1}, and room volumes in units of m^{3}. Concentrations were calculated using mean LE emission rates from this study. Increment concentrations graduate on a low-to-high colour scale from blue to red.

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3.4 Modelling α-pinene increments

Fig. 5 shows the resulting contour plot for the increment of α-pinene from the use of one diffuser. As the increment increases, the colour of the contour graduates from blue through to red. This model shows that plausibly high concentrations are possible in very small rooms combined with low AERs. Spaces in homes with this combination of low volume and low AER are likely to be rooms where occupancy is incidental and episodic however, rather than over longer periods of time, such as under-the-stairs cloakrooms or porch cupboards. The short-term (30 min) exposure limit for α-pinene in indoor settings in the UK is 45 000 µg m^{-3},[71] which may give rise to noticeable effects to the consumer's comfort. Using 1 LE with the lowest feasible model combination of room volume and AER in the above model ($V = 0.5$ m^3, $A_H = 0.5$ h^{-1}) gave an α-pinene increment of 3700 µg m^{-3}, more than an order of magnitude lower than the short-term exposure limit, and also lower than the long-term exposure limit (1 day of constant exposure) of 4500 µg m^{-3}.[71] However, spaces in homes with this combination of room volume and AER are likely small cupboards (kitchen cupboards, under-the-stairs storage cupboards) where VOC exposure would be incidental, and LE use in unlikely. A comparison of sampled concentrations from this study, along with exposure thresholds are given in Table 2.

3.5 Comparison with real-world analogues

Chambers or booths are used extensively to measure emissions of VOCs from household products and also have applications in sensory sciences.[72–74] Chambers for VOC analysis are typically of stainless steel construction with glass panels and sampling ports, and while they can range in sizes, they are typically smaller volumes for ease of use and space in laboratories. While smaller testing chambers allow for potentially better analysis of emitted VOCs through product use for instruments with higher detection limits, they do not necessarily replicate the same conditions found in real-world analogues. In this study, two sizes of booths were used, a smaller booth of approximately 10 m^3 and a larger booth of approximately 30 m^3, representing the size of a bathroom and living room, respectively. The booths used in this study additionally had more real-world representative surfaces within the volume, such as painted walls and wooden doors.

Fig. 6 shows a comparison of the concentrations of samples from this study compared with real-world concentrations of VOCs through LE use found in Warburton *et al.* (2023).[26] The same LE device and liquid formulation was used across both studies. It should be noted that the rooms used in Warburton *et al.* (2023)[26] were the main living area of the house and ranged from between 20 m^3 and 50 m^3 in volume. Whilst the data showed incremental fragrance VOC increases in the booth plot (especially so for the more confined space of the toilet booth), comparison with a cohort of real-world homes from Warburton *et al.* (2023)[26] showed little difference in both the overall VOC concentrations found, and in the concentrations of individual fragrance VOCs between the two studies. This may, of course, arise from the differences in AER levels between homes and the test booths, although comparative data for one LE in both situations shows little difference in concentrations reported. From a consumer perspective, the concentrations generated in the large booths when multiple devices were used did not lead to elevated fragrance VOC concentrations above those seen typically in homes, remaining well below the short-term and long-term exposure limit (1 day of constant exposure) as outlined by UK indoor exposure thresholds available from Public Health England (Table 2).[71] Although assessing potential health impacts is beyond the scope of this study, modelling indicated that high VOC concentrations approaching exposure limit thresholds could occur only in rooms with an unrealistic combination of low AERs and small volumes. Typically, product packaging provides user guidance on proper usage, including recommendations for adequate ventilation and suitable room size. Using the device in small, poorly ventilated spaces would therefore be considered off-label use.

Comparisons between chamber-based studies and real-world studies are complex by nature. In real-world scenarios, there is more than likely no one single point emission source of VOCs throughout the sampling period, and air makeup is highly changeable between samples.[75] Additionally, in booths there are typically in-flows and out-flows to create air exchange, which may interfere with VOC release and mixing into the volume. Such molecular-level disturbances may be difficult to detect through typical means such as the use of anemometers. However, in this study, the booths were made to be as reflective of real-world conditions as possible, given the sensory science-based nature of the regular use of the booths used in this study.

Table 2 Comparison of median sampled concentrations from this study against UK indoor exposure thresholds available from Public Health England.[71] All concentrations are shown in µg m^{-3}

Species	1 LE (small booth)	1 LE (large booth)	5 LEs (small booth)	5 LEs (large booth)	Indoor short-term limit	Indoor long-term limit
α-Pinene	3.980	1.264	24.580	5.289	45 000[a]	4500[c]
Limonene	0.162	0.038	0.947	0.117	90 000[a]	9000[c]
Acetaldehyde	4.800	4.881	4.799	5.71	1420[b]	280[c]
Benzene	0.632	0.485	0.644	0.435	0[d]	0[d]

[a] 30 min exposure. [b] 1 hour exposure. [c] 1 day exposure. [d] No safe exposure limit to benzene has been published, however England and Wales have a 5 µg m^{-3} benzene concentration target according to European (EU) ambient air quality directive 2008/50/EC.
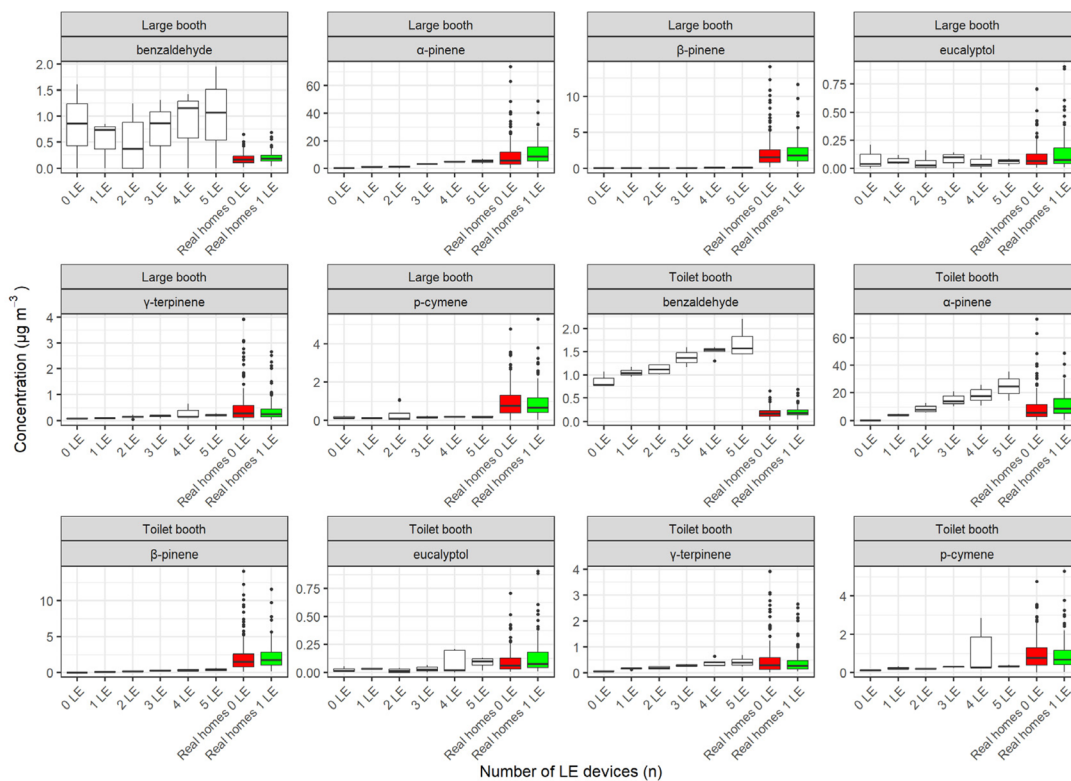
Fig. 6 Comparison of concentrations collected in this study against the concentrations found in Warburton *et al.* (2023),[26] using the same LE device and liquid formulation. All concentrations are shown in $\mu g\ m^{-3}$. Concentrations from Warburton *et al.* (2023)[26] are displayed on the plots as 'Real homes'. From bottom to top, each boxplot shows 5th percentile, 25th percentile, 50th percentile/median, 75th percentile, 95th percentile. Any outliers are given as single dots above or below the 5th/95th whisker.

4 Conclusions

In this study the olfactive perception and VOC concentrations arising from fragrance LE use in controlled but realistically sized and ventilated test rooms were assessed. As the number of LEs deployed in a room increased, concentrations of VOCs increased linearly, although individual VOC concentrations lower than 10 $\mu g\ m^{-3}$ were generated in the larger room. Based on intensity feedback reported by trained panelists, the perception of the intensity change arising from incrementally adding LEs into a room was not of significance above 2 LEs in the 10 m^3 toilet booths. The potential exists for multiple devices to be added to a single room space. In these cases, there was limited difference in fragrance intensity when 2 or more LEs were added concurrently. The lack of human perception of increased fragranced intensity with the use of multiple LEs within an airspace may however limit the potential for such off-instruction use, since little end user benefit is likely to be detected or where the consumer felt that 1 LE was performing adequately.

When 5 LEs were used in test rooms, concentrations of volatile monoterpenes remained relatively low despite testers reporting intense fragrance. α-Pinene concentrations generated in the test rooms using 5 LEs were comparable to typical concentrations found ordinarily in residential homes, with upper α-pinene concentrations comparing between the 50th to 60th percentile of real-home data in available literature. The in-room concentrations of α-pinene were below the literature reported individual ODL.

Analysis between this study and a real-world study using an LE showed generally comparable VOC release and in-room concentrations across both studies. The use of controlled but realistic booths and chambers could be a reasonable substitute to predict VOC exposure for regular LE consumer uses.

There are broader implications however for indoor air quality. Reactive VOCs can impact indoor air quality through atmospheric reactions that generate by-products such as formaldehyde, acetaldehyde and secondary organic particulate matter. Ultimately, the contribution that a fragranced-derived compound will make to increasing secondary pollutants indoors will be a combination of the amount emitted and then present available for oxidation in air, the available oxidants (*e.g.* O_3, Cl, NO_3, OH), and the bimolecular rate coefficients. It is clear that some components of the LE fragrance tested here create strong perceptible odours for humans but generate only very low concentrations in the gas phase. These were not detectable by the sampling and TD-GC-QMS methods used here, and it can be hypothesised that concentrations are likely to be in the low part per trillion range or below, rather than part per billion which was typical of more volatile species such as monoterpenes.

Much literature discussion of the possible indoor impacts of fragranced products considers the ultimate fate of the

emissions following gas phase oxidation reactions. If a fragrance contains monoterpenes and the use of the product generates in-room increments in the order of tens of μg m^{-3} in indoor air, that in turn has the potential to create comparable μg m^{-3} increments of formaldehyde (having a VOC to HCHO yield in the region 5–10%).[76]

In this study whilst monoterpenes were a substantial component of the fragrance formulation and the major type of VOC generated indoors from the use of LE products, they did not appear to be a major contributor to the perception of fragrance by human testers. Often concentrations were below their reported ODL. Reduction of monoterpene content from raw materials might be a means to reduce possible indoor air pollution in low-ventilation homes without necessarily substantially changing human-perceived product performance.

4.1 Future work

Future work aimed at identifying the olfactive intensity of fragrances in controlled settings should consider assessments under variable AERs, as this could illuminate perception gradients by varying the concentration of LE-emitted VOCs. A challenge in such studies would be the pairing of air sampling and analytical methods to resolve both the highly volatile fractions of fragrance formulations, as achieved in this study, and the less volatile fractions, which are likely present at ≤ppt gas-phase mixing ratios. However, the use of sensitive on-line analytical techniques, such as proton transfer reaction (PTR) or selected ion flow tube (SIFT) mass spectrometry, could enable assessments of the emitted concentrations of these lower volatility species, as well as offer a time series of emissions and decays of LE-originating VOCs.

Data availability

All sampled data from this study is freely available from the Centre for Environmental Data Analysis (CEDA) repository, at **https://www.ceda.ac.uk** (currently pending review at CEDA but scheduled for upload).

Author contributions

The experiments were designed by TW, ACL, NO, CJ, GA and BX. TW, JRH, SJA and AMY supported the VOC analytical measurements used in this study, NO, CJ led the olfactory testing component. TW and ACL undertook the data analysis and subsequent interpretation. All authors contributed to the drafting and reviewing of the manuscript.

Conflicts of interest

NO, CJ, and GA are employees of Givaudan UK Ltd, Givaudan Fragrances Corp. and BX is an employee of Bath & Body Works, Inc. who are industrial suppliers of chemicals and finished household and personal care products. To support independence, all analytical work and data analysis was undertaken by University of York with no restrictions placed on freedoms to publish. To support transparency in research, data collected in this study are freely available from the Centre for Environment Data and Analysis (**https://www.ceda.ac.uk**).

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