Building and Environment 255 (2024) 111314

Contents lists available at ScienceDirect

Building and Environment

journal homepage: www.elsevier.com/locate/buildenv

Real-time evaluation of terpene emissions and exposures during the use of scented wax products in residential buildings with PTR-TOF-MS

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ARTICLE INFO

Keywords:
Indoor air quality
Volatile organic compounds
Chemical emissions
Human exposure
Indoor chemistry

ABSTRACT

Scented wax products, such as candles and wax warmers/melts, are popular fragranced consumer products that are commonly used in residential buildings. As scented wax products are intentionally fragranced to produce pleasant smellscapes for occupants, they may represent an important source of volatile organic compounds (VOCs) to indoor atmospheres. The aim of this study is to evaluate terpene emission factors (EFs) and inhalation intake fractions (iFs) for scented wax products to better understand their impact on indoor chemistry and chemical exposures. Full-scale emission experiments were conducted in the Purdue zEDGE Test House using a variety of scented candles ($n = 5$) and wax warmers/melts ($n = 14$) under different outdoor air exchange rates (AERs). Terpene concentrations were measured in real-time using a proton transfer reaction time-of-flight mass spectrometer (PTR-TOF-MS). PTR-TOF-MS measurements revealed that scented candle and wax warmer/melt products emit a variety of monoterpenes ($C_{10}H_{16}$) and oxygen-containing monoterpenoids ($C_{10}H_{14}O$, $C_{10}H_{16}O$, $C_{10}H_{18}O$, $C_{10}H_{20}O$), with peak concentrations in the range of 10^{-1} to 10^{2} ppb. Monoterpene EFs were much greater for scented wax warmers/melts ($C_{10}H_{16}$ EFs $\sim 10^{2}$ mg per g wax consumed) compared to scented candles ($C_{10}H_{16}$ EFs $\sim 10^{-1}$ to 10^{0} mg per g wax consumed). Significant emissions of reactive terpenes from both products, along with nitrogen oxides (NO, NO_2) from candles, depleted indoor ozone (O_3) concentrations. Terpene iFs were similar between the two products (iFs $\sim 10^{3}$ ppm) and increased with decreasing outdoor AER. Terpene iFs during concentration decay periods were similar to, or greater than, iFs during active emission periods for outdoor AERs ≤ 3.0 h^{-1}. Overall, scented wax warmers/melts were found to release greater quantities of monoterpenes compared to other fragranced consumer products used in the home, including botanical disinfectants, hair care products, air fresheners, and scented sprays.

1. Introduction

The manufacture and use of fragranced consumer products, such as air fresheners, personal care products, cleaning agents, and surface disinfectants, is known to release volatile organic compounds (VOCs) to the atmosphere [1–5]. More than 2600 chemical ingredients are documented in fragrance production [6]. Major identified chemical substances from the use of fragranced consumer products include a variety of VOCs, such as alcohols, aldehydes, acetates, and terpenes [7–10]. According to the U.S. Federal Food, Drug, and Cosmetic Act (FFDCA), ingredients in fragrances are not required to be listed on the product labels for both regulated and unregulated consumer products [11]. A headspace analysis revealed that less than 4% of identified VOC species were present on fragranced consumer product labels [12]. Meanwhile, manufacturers of consumer products are not required to disclose all ingredients to consumers [13], and VOC emission rates under realistic

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https://doi.org/10.1016/j.buildenv.2024.111314
Received 12 November 2023; Received in revised form 28 January 2024; Accepted 15 February 2024

indoor conditions are seldom reported to the public. Under these circumstances, potential indoor VOC emissions and exposures during the use of fragranced consumer products remains unknown.

Scented wax products, such as scented candles and wax warmers (also known as wax melts), are popular fragranced consumer products that are used to produce pleasant aromas in homes, offices, and other indoor environments [14–18]. The use of scented candles is associated with emissions of VOCs and adverse impacts on indoor air quality [19, 20]. However, there are no prior investigations into VOC emissions from scented wax warmers, which are growing in popularity and are marketed as a non-combustion alterative to candles. Prior research into the identification and characterization of VOC emissions from the use of scented candles has predominantly focused on conducting chamber experiments with offline sample analysis via gas chromatography-mass spectrometry (GC-MS) [7,14,19,21–23]. Such studies have detected a wide range of VOCs with molecular weights from 30 to 240 g mol^{-1} [21, 22], including numerous terpenes that are associated with essential oils incorporated into the wax of the candle [24–27]. Monoterpenes are composed of two isoprene units [28–30]. Monoterpenes, such as D-limonene and α-pinene, and oxygen-containing monoterpenoids, such as linalool and eucalyptol, are common terpene species present in essential oils [29,31–34]. It has been demonstrated that household exposure to D-limonene may increase the prevalence of asthma among the general population [35]. Given the strong fragrances attributed to wax warmers, it is expected that they represent an important, yet poorly characterized, indoor terpene source.

Considering the massive market demand for scented wax products [36], the emission of terpenes from these products can potentially affect indoor and outdoor atmospheric chemistry as terpenes have been identified as reactive precursors to the formation of gas-phase carbonyls and secondary organic aerosol (SOA) [37–48]. It has been reported that the ozonolysis of monoterpenes can form carbonyls, such as formaldehyde and acids, and other low volatility products that can nucleate and form sub-3 nm nanocluster aerosol [39,49,50]. Exposure to these secondary products can also cause adverse health impacts. A major health outcome observed from ozone-terpene reactions was respiratory irritation in the upper airways [51]. In addition, sub-3 nm nanocluster aerosol are associated with high respiratory tract deposited dose rates and have been reported to have an impact on heart rate variability in healthy individuals [52]. Therefore, it is important to investigate terpene emissions and exposures from scented wax products via a material balance framework to better understand their impact on the chemical composition of indoor air and the potential health risks associated with their use in buildings.

A material balance model can enable the calculation of speciated emission factors (EFs), emission rates (ERs), and inhalation intake fractions (iFs) for VOCs emitted into an indoor environment [53–62]. These parameters can enable the quantification of VOC emissions and human exposures for different products and can provide a useful means for choosing products considering their impact on indoor air quality [58, 63–66]. Nevertheless, the calculation of emission factors and intake fractions require instruments that enable continuous monitoring of indoor air, such as a high-resolution proton transfer reaction time-of-flight mass spectrometer (PTR-TOF-MS) [67–72]. In recent years, PTR-TOF-MS has been deployed for the real-time characterization of indoor VOC emissions from human activities, including the use of fragranced consumer products [72–82]. Terpenes, including monoterpenes, monoterpenoids, sesquiterpenes, and other terpene derivatives, can be detected by PTR-TOF-MS with mass-to-charge ratios (m/z) ranging from 67 to 221 [69,83–88].

To date, there is limited research on online PTR-TOF-MS measurements of VOC emissions from scented candle use and no research on VOC emissions from scented wax warmer use [7,77]. Also, there are no prior reports of the real-time evaluation of terpene EFs, ERs, and iFs during scented wax product use in residential buildings. A comparison of terpene emissions between scented wax products with and without combustion, as well as the impact of building air exchange rates (AERs) on indoor terpene exposures, is needed given the growing popularity of wax warmers in home environments. This study integrates real-time PTR-TOF-MS measurements with a material balance model to address knowledge gaps related to the impact of different scented wax products on indoor air quality. The objectives of this study are to: (a) characterize terpene emissions during the use of scented wax products with PTR-TOF-MS; (b) compare the gas-phase emission profiles for different scented candles and wax warmers featuring a variety of fragrances; (c) evaluate terpene emission potentials and associated human exposures for each product by use of a material balance model; and (d) investigate the impact of outdoor AERs on human exposure to terpenes during active emission and concentration decay periods.

2. Materials and methods

2.1. Study site: Purdue zEDGE Test House

Indoor VOC emission measurements were conducted in a mechanically ventilated residential architectural engineering laboratory – the Purdue zero Energy Design Guidance for Engineers (zEDGE) Test House located on the Purdue University campus in West Lafayette, Indiana, U.S.A. Exterior and interior photos of the Purdue zEDGE Test House are shown in Fig. S1. zEDGE is a single zone residential building built on a mobile trailer according to the guidelines established by the Recreational Vehicle Industry Association (RVIA) and holds a National Organization of Alternative Housing (NOAH) certificate. The interior volume of zEDGE is 60.35 m^3. A single-zone ductless heating and cooling system (FTX12NMVJU, Daikin North America LLC, Houston, TX, U.S.A.) was adopted to maintain a nominal indoor air temperature of 20 °C (68 °F). The outdoor AER of zEDGE was controlled by a combination of a powered ventilator with two MERV 13 filters to supply filtered outdoor air to indoors, a variable-speed bathroom exhaust fan (FV–0511VKS2, Panasonic Eco Solutions of North America, Newark, NJ, U.S.A.), and a portable air conditioner with an exhaust duct (QPCA08JAMWG1, Haier, Louisville, KY, U.S.A.). Detailed ventilation conditions for each AER setting and their associated pressure differentials are presented in Text S1. The layout of the ventilation equipment within zEDGE is illustrated in Fig. S2.

2.2. Instrumentation and measurements

2.2.1. Real-time measurement of indoor VOCs with PTR-TOF-MS

The compounds of interest in this study are monoterpenes (chemical formula: $C_{10}H_{16}$; detected at m/z 81 and 137, representing a known fragment $[C_6H_9]^+$ and a protonated mass $[(C_{10}H_{16})H]^+$, respectively) and monoterpenoids (chemical formula: $C_{10}H_{14}O$, $C_{10}H_{16}O$, $C_{10}H_{18}O$, $C_{10}H_{20}O$; detected at m/z 151, 153, 155, and 157, representing the protonated mass of $[(C_{10}H_{14}O)H]^+$, $[(C_{10}H_{16}O)H]^+$, $[(C_{10}H_{18}O)H]^+$, and $[(C_{10}H_{20}O)H]^+$, respectively). Mixing ratios of VOCs were measured at 1 Hz with an inlet sampling flow rate of 100 sccm by a PTR-TOF-MS (PTR-TOF 4000, Ionicon Analytik Ges.m.b.H., Innsbruck, Austria) using hydronium (H_3O^+) as the reagent ion. VOCs with proton affinities greater than water were ionized by H_3O^+ through a proton transfer reaction in the drift tube (Eq. (1)) [83,89–91]:

$$H_3O^+ + VOC \rightarrow VOCH^+ + H_2O \tag{1}$$

The ionized molecules ($VOCH^+$) are then separated and detected by the TOF-MS and m/z 30 to 450 are recorded. The ionization field energy (E/N) of the PTR-TOF-MS was maintained at approximately 139 Td with the operational pressure, voltage, and temperature of the drift tube set at 2.2 mbar, 600 V, and 70 °C, respectively. A perfluoroalkoxy (PFA) sampling line (3/8 in. OD) was located in the center of zEDGE. A polytetrafluoroethylene (PTFE) membrane filter (1 μm pore size) was installed at the intake of the sampling line to remove particles. The PTFE membrane filter was replaced daily.

The PTR-TOF-MS was calibrated daily with two VOC gas standard mixtures. The first one contained approximately 2 ppm of 16 different compounds and the second one contained approximately 200 ppb of 5 compounds including a cyclic monoterpene – D-limonene ($C_{10}H_{16}$). A list of compounds and their mixing ratios in the calibration gas standard mixtures is provided in Table S1. A liquid calibration unit (LCU, Ionicon Analytik Ges.m.b.H., Innsbruck, Austria), which features inlets for compressed gases, was adopted to dilute the two gas standards to 2 – 40 ppb by ultra-zero air. For VOCs not available in the gas standards, the mixing ratios of these compounds were calculated based on proton transfer theory [91,92].

All raw signals from the PTR-TOF-MS were processed by Ionicon Data Analyzer (IDA, Ionicon Analytik Ges.m.b.H., Innsbruck, Austria). Raw signals were converted to mixing ratios for VOCs in parts per billion (ppb) based on Eq. (2).

$$VOC\ (ppb) = 1.657 \bullet 10^{-11}\frac{1}{k} \bullet \frac{U_{drift} \bullet T_{drift}^{2}}{P_{drift}^{2}} \bullet \frac{I_{VOCH^{+}}}{I_{H_3O^{+}}} \bullet \frac{TR_{H_3O^{+}}}{TR_{VOCH^{+}}} \tag{2}$$

Where P_{drift} (mbar), T_{drift} (K), and U_{drift} (V) are the pressure, temperature, and voltage of the drift tube as measured by the instrument. I_{VOCH^+} (cps) and $I_{H_3O^+}$ (cps) are the respective ion count rates of the protonated VOC and hydronium. $TR_{H_3O^+}$ and TR_{VOCH^+} are the transmission values of hydronium and the protonated VOC which are obtained from the fitted transmission curve. The transmission curves were determined by PTR-MS Viewer (Ionicon Analytik Ges.m.b.H., Innsbruck, Austria) using the daily calibration signals. An example of a transmission curve acquired during the measurement campaign is provided in Fig. S3, and the transmission values of each experiment are provided in Table S2. The reaction rate constant (k) of selected compounds was assumed to be 2×10^{-9} cm^3 s^{-1} [93]. Table S3 lists the additional information including the associated m/z and tentatively identified isomers for identified terpenes. In this study, we were unable to separate isomers, thus, the targeted monoterpenoids may share the $[C_6H_9]^+$ (m/z 81) and $[(C_{10}H_{16})H]^+$ (m/z 137) ions with monoterpenes at a high % abundance [85,86]. To conduct real-time separation of isomers, a PTR-TOF-MS configured with a fast gas chromatograph (fastGC) at the inlet is required [94]. Therefore, the mixing ratios derived from the PTR-TOF-MS measurement in this study are the sum of all isomers with the same chemical formula.

2.2.2. Real-time measurement of indoor trace gases

Indoor mixing ratios of nitrogen oxides (NO, NO_2) were measured by a chemiluminescence NO-NO_2-NO_x analyzer (Model 42C, Thermo Electron Corp., Waltham, MA, U.S.A.). Indoor mixing ratios of ozone (O_3) were measured with a photometric analyzer based on non-dispersive ultraviolet (UV) absorption of O_3 at 254 nm (Serinus 10, ACOEM Ecotech, Melbourne, Australia). The NO-NO_2-NO_x and O_3 analyzers were calibrated before the measurement campaign.

2.3. Experimental approach

In total, five scented candle products (A to E) and fourteen scented wax warmer products (A to N) were tested under different outdoor AERs during the measurement campaign. The scented wax products were purchased from a local retail store and represent a variety of manufacturers and fragrances. The fragrance for each product and associated experimental information are summarized in Table 1. The experimental AER and the calculated loss rate for $C_{10}H_{16}$ for each product are provided in Table S4. The source event period of the scented wax warmer experiments was designed to last longer than the scented candle experiments because the wax warmers required a longer period to reach a complete melt. The decay period of each experiment was adequate for the decrease in the indoor VOC concentration to reach steady-state (approximately 95% decrease from the peak VOC mixing ratios).

2.3.1. Experimental sequence for scented candle products

For experiments using scented candle products, three emission experiments were completed for each of the products. The experimental sequences are illustrated in Fig. 1a and 1b, and a detailed experimental protocol is elaborated in Text S2. For experiments using products B to E, the outdoor AER of zEDGE was controlled at 6.5 h^{-1}, and experiments were conducted with 20 min of background, a 20 min steady burning event, and a 1 h concentration decay period. Additionally, experiments using candle product A with an AER at 0.5 h^{-1} were conducted to provide an overview of the emission profile to compare the mixing ratios and human exposures between high and low AERs. These additional experiments were conducted with 20 min of background, a 20 min steady burning event, and a 2 h concentration decay period.

2.3.2. Experimental sequence for scented wax warmer products

For experiments using scented wax warmer products A to N, one

Table 1

Summary of tested scented wax products and associated experimental information.

Scented Candle Product	Fragrance(s)	No. of Products	AER (h^{-1})	Source (min)	Decay (min)	Scented Wax Mass Loss (g)
A	Citrus, berry	4	0.5	20	120	4.2
B	Rose, peach, floral	4	6.5	20	60	4.0
C	Bergamot, amber, woods, musk	2	6.5	20	60	9.0
D	Raspberry	4	6.5	20	60	6.8
E	Blue ferns, citrus	2	6.5	20	60	3.2
Scented Wax Warmer Product	**Fragrance(s)**	**No. of Products**	**AER (h^{-1})**	**Source (min)**	**Decay (min)**	**Scented Wax Mass Loss (g)**
A	Papaya	2 cubes	6.5	50	60	0.5
B	Balsam, cedar	2 cubes	6.5	50	60	0.4
C	Pineapple, vanilla	2 cubes	6.5	50	60	0.2
D	Tangerine	2 cubes	6.5	50	60	0.1
			3.0	50	60	0.4
E	Lime, eucalyptus	2 cubes	6.5	50	60	0.2
F	Lemon	2 cubes	6.5	50	60	0.3
G	Patchouli, musk	2 cubes	6.5	50	60	0.4
H	Bergamot, sage	2 cubes	6.5	50	60	0.5
			3.0	50	60	0.4
I	Lemon, mandarin	2 cubes	6.5	50	60	0.4
			3.0	50	60	0.5
J	Lemon	2 cubes	6.5	50	60	0.4
K	Peppermint	2 cubes	6.5	50	60	0.3
L	Bergamot, amber, woods, musk	2 cubes	6.5	50	60	0.3
M	Bamboo	2 cubes	6.5	50	60	0.4
N	Lemon grass, ginger	2 cubes	6.5	50	60	0.5

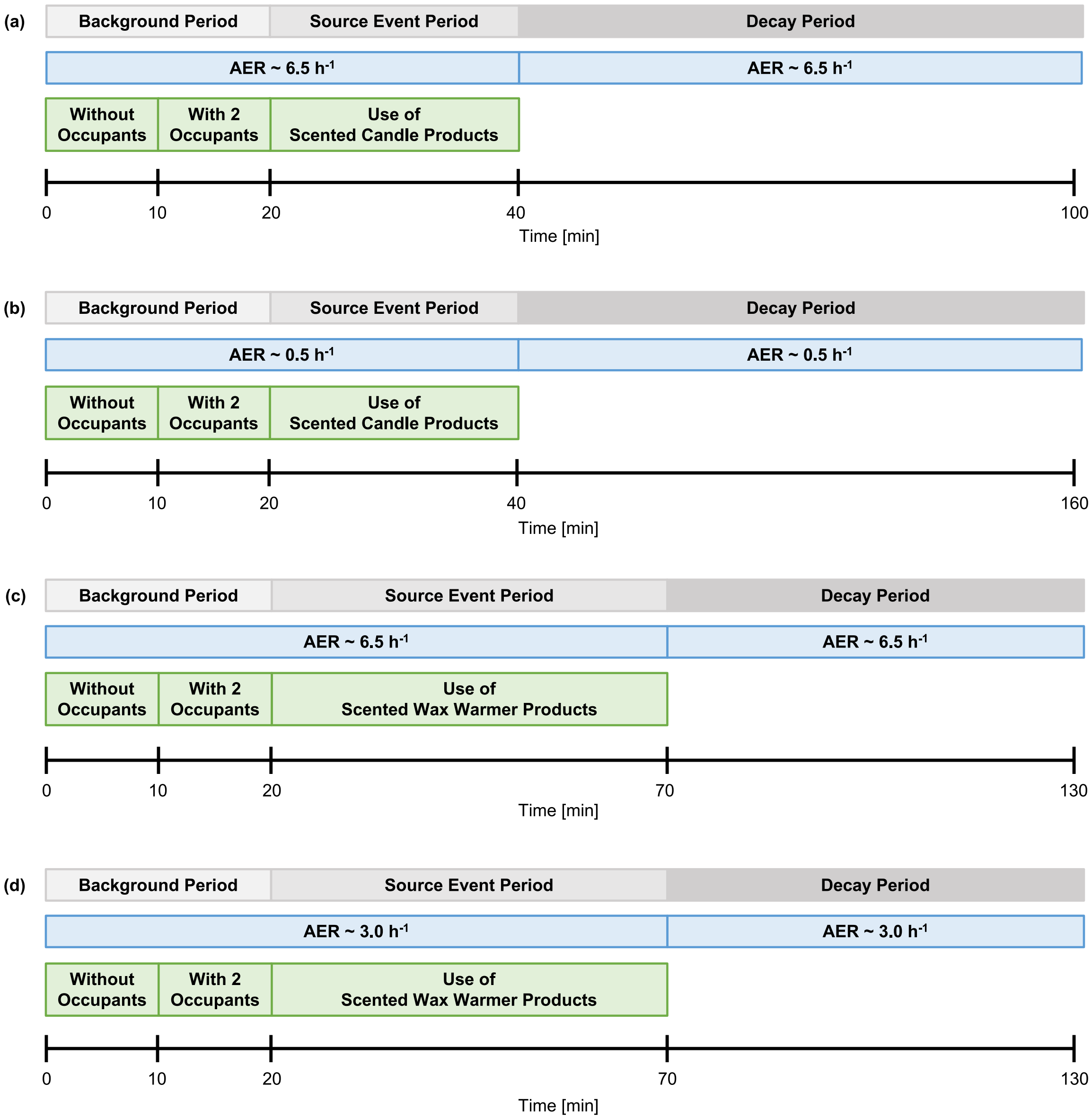

Fig. 1. Experimental sequences for scented wax product emission experiments. (a) Sequence for scented candle emission experiments with AER ~ 6.5 h^{-1}. (b) Sequence for scented candle emission experiments with AER ~ 0.5 h^{-1}. (c) Sequence for scented wax warmer emission experiments with AER ~ 6.5 h^{-1}. (d) Sequence for scented wax warmer emission experiments with AER ~ 3.0 h^{-1}.

emission experiment was completed for each of the products. The experimental sequences are illustrated in Fig. 1c and 1d, and a detailed experimental protocol is elaborated in Text S3. All products were tested under an outdoor AER at 6.5 h^{-1}, and products D, H, and I were chosen to be repeated with a lower AER at 3.0 h^{-1} for comparison. All experiments were conducted with 20 min of background, a 50 min source event, and a 1 h concentration decay period.

2.4. Calculation of terpene emission factors and inhalation intake fractions

EFs and iFs of the targeted monoterpenes and monoterpenoids were calculated based on a material balance model [69,70,72,95]. A schematic of the material balance model is illustrated in Fig. S4. For scented wax products, the EF can be expressed as the mass of VOC emitted per mass of wax consumed and the iF can be expressed as the mass of VOC inhaled by an individual per mass of VOC emitted. For scented candle products, the reported EFs were the mean value of the three identical experiments with an error bar indicating the standard deviation of the three experiments. For the scented wax warmer experiments, the reported EFs were the mean value of the individual experiment with an error bar representing the standard deviation of a single experiment. The duration of the source event period and decay period, along with the associated wax mass loss of each scented wax product after completion of the source event period, are provided in Table 1. To apply the material balance model, zEDGE was treated as a completely mixed flow reactor (CMFR) with four fans to promote adequate indoor air mixing.

To define the matrices mentioned above for each scented wax product, we shall first evaluate indoor VOC emissions by determining the overall VOC source and loss rates. The procedure for the application of the material balance model in this study can be described in six steps: (1) determine the overall loss rate of each terpene during the decay period; (2) determine the overall source rate of each terpene using the calculated loss rate; (3) normalize the overall source rate of each terpene by the wax mass loss of each product to obtain the EF and by the duration of the source event period to obtain the ER; (4) determine the inhalation intake of each terpene; (5) determine the total terpene inhalation intake (sum of all terpenes) and the total terpene emission (sum of all terpenes); and (6) determine the overall terpene iF (sum of all terpenes) using the total terpene inhalation intake and emission.

A general material balance model treating zEDGE as a CMFR can be expressed as Eq. (3):

$$V\frac{dC_{m,in}(t)}{dt}=S_m(t)-L_mC_{m,in}(t)V \tag{3}$$

where m represents the compound of interest, $C_{m,in}(t)$ is the indoor mass concentration of compound m at time t (mg m^{-3}), $S_m(t)$ is the overall source rate at time t (mg h^{-1}), V is the interior volume of zEDGE (m^3), and L_m is the overall loss rate (h^{-1}), which is assumed to be approximately constant over time.

In this study, the primary loss mechanism for the various terpenes is the ventilation of indoor air to the outdoors through the bathroom exhaust fan and portable air conditioner. Other minor factors that need to be considered include the sorption of the compound to interior surfaces and reactions between the terpene and indoor O_3. Reactions between terpenes and indoor hydroxyl radicals (OH) are assumed to be negligible as indoor OH concentrations are typically low at 10^4 to 10^5 molecule cm^{-3} [96,97]. Thus, the overall loss rate can be expressed as Eq. (4):

$$L_m=AER+k[O_3](t)+k_{m,s} \tag{4}$$

where AER is the outdoor AER for zEDGE (h^{-1}), k is the reaction rate constant between compound m and O_3 (cm^3 molecule^{-1} s^{-1}), $[O_3](t)$ is the indoor concentration of O_3 at time t (molecule cm^{-3}), and $k_{m,s}$ is the sorption rate coefficient of compound m to indoor surfaces (h^{-1}). Although $k[O_3](t)$ is a function of time, L_m is considered as approximately constant over time due to the small variation in $[O_3]$ during the experiments.

To calculate the loss rate (L_m) for the targeted terpenes, an analytical equation (Eq. (5)) was derived from Eq. (3). L_m can be determined by fitting Eq. (5) using a non-linear least squares method in MATLAB (The MathWorks Inc., Natick, MA, U.S.A.):

$$C_{m,in}(t)=\left(C_{m,in}(0)-C_{m,ss}\right)e^{-L_mt}+C_{m,ss} \tag{5}$$

where $C_{m,in}(0)$ is the indoor mass concentration for compound m at the beginning of the decay period and $C_{m,ss}$ is the approximate steady-state indoor mass concentration for compound m at the end of the decay period [69]. In this study, we use the data 10 min after the peak VOC mixing ratio to fit the decay curve and $C_{m,ss}$ is assumed to be 98% of the concentration values at the end of the decay. Fig. S5 illustrates the non-linear curve fitting for an experiment using scented wax warmer product A as an example.

Three sources were considered for the calculation of the overall source rate in zEDGE: emissions from the scented wax product due to volatilization of VOCs from the melted wax, ventilated air from outdoors to indoors via the powered ventilator, and desorption from the interior surfaces of zEDGE. The overall source rate can be expressed as Eq. (6):

$$S_m(t)=E_m(t)+AER\times C_{m,out}(t)\times V+S_{m,d}(t) \tag{6}$$

where $E_m(t)$ is the emission rate for the scented wax product (mg h^{-1}), $C_{m,out}(t)$ is the outdoor mass concentration for compound m at time t (mg m^{-3}), and $S_{m,d}(t)$ is the desorption rate of compound m from indoor surfaces at time t (mg h^{-1}). Given the significant elevation in indoor terpene mixing ratios observed during the source event periods beyond those measured during the background periods, it is assumed that the latter two source processes of outdoor air ($AER\times C_{m,out}(t)\times V$) and desorption from the interior building surfaces ($S_{m,d}(t)$) are negligible in magnitude compared to emissions from the scented wax products ($E_m(t)$) from t_1 to t_2, thus: $E_m(t)\gg AER\times C_{m,out}(t)\times V+S_{m,d}(t)$ during the emission period [69]. Thus, $E_m(t)\sim S_m(t)$.

To calculate the emission of targeted terpenes, an integration equation (Eq. (7)) was derived from Eq. (3). Here, the overall loss rate (L_m) calculated from Eq. (5) was considered constant throughout the experiment. The emission of targeted terpenes can be determined by integrating the overall source rate from the source event start time to the end time before the concentration decay period:

$$\int_{t_1}^{t_2}S_m(t)dt=V\left[C_{m,in}(t_2)-C_{m,in}(t_1)\right]+L_mV\int_{t_1}^{t_2}C_{m,in}(t)dt \tag{7}$$

where t_1 and t_2 represent the start time and end time of the source event period (burning candle or melting wax warmer products), respectively, and $\int_{t_1}^{t_2}S_m(t)dt$ is the amount of compound emitted from t_1 to t_2 (mg). Normalizing $\int_{t_1}^{t_2}S_m(t)dt$ by the recorded wax mass loss of the scented wax product (W_L, g) after the source event will give us the EF of a compound in mg g^{-1} (Eq. (8)):

$$EF\left(mg\,g^{-1}\right)=\frac{mass\ of\ VOC\ emitted\ to\ the\ environment\ (mg)}{wax\ mass\ loss\ (g)}=\frac{1}{W_L}\int_{t_1}^{t_2}S_m(t)\,dt \tag{8}$$

The ER of a compound can also be determined by normalizing $\int_{t_1}^{t_2}S_m(t)dt$ by the time of source event period (t_s, min) (Eq. (9)):

$$ER\left(mg\,min^{-1}\right)=\frac{mass\ of\ VOC\ emitted\ to\ the\ environment\ (mg)}{time\ of\ source\ event\ (min)}=\frac{1}{t_s}\int_{t_1}^{t_2}S_m(t)\,dt \tag{9}$$

The inhalation intake of a compound by an occupant during the experimental period for each scented wax product can be determined by Eq. (10):

$$mass\ of\ VOC\ inhaled\ by\ an\ occupant\ (mg)=Q\int_{t_1}^{t_2}C_{m,in}(t)\,dt \tag{10}$$

where Q is the inhalation rate (m^3 h^{-1}), and t_1 and t_2 represent the start and end time of a specific exposure period. The inhalation rate for an adult engaged in light activity (1.25 m^3 h^{-1}) was adopted for the calculation of the iFs and assumed to be constant over time. Dividing $Q\int_{t_1}^{t_2}C_{m,in}(t)\,dt$ by the emission of the compound calculated from Eq. (7) gives a ratio which is the iF of that compound (Eq. (11)):

$$iF=\frac{mass\ of\ VOC\ inhaled\ by\ an\ occupant\ (mg)}{mass\ of\ VOC\ emitted\ to\ the\ environment\ (mg)}=\frac{Q\int_{t_1}^{t_2}C_{m,in}(t)\,dt}{\int_{t_1}^{t_2}S_m(t)\,dt} \tag{11}$$

In this study, we reported an overall terpene iF for a full source period and a full decay period for each product. The overall terpene iF can be represented by $\frac{Q\int_{t_1}^{t_2}\sum(C_{m,in}(t))\,dt}{\int_{t_1}^{t_2}\sum(S_m(t))\,dt}$, which is the total mass of $C_{10}H_{16}$, $C_{10}H_{14}O$, $C_{10}H_{16}O$, $C_{10}H_{18}O$, and $C_{10}H_{20}O$ inhaled by an occupant divided by the total emission of $C_{10}H_{16}$, $C_{10}H_{14}O$, $C_{10}H_{16}O$, $C_{10}H_{18}O$, and $C_{10}H_{20}O$ emitted to indoor air.

3. Results and discussion

3.1. Contribution of terpenes to total VOC emissions during the use of scented wax products

Among all detected VOCs, monoterpenes (detected at m/z 81 and $m/$

z 137) were identified as the most abundant VOCs for every scented wax product tested in this study. Fig. 2 illustrates the contribution of VOCs detected by the PTR-TOF-MS during the use of scented candle product C and scented wax warmer product B. For scented candle product C (Fig. 2a), terpenes (including $C_{10}H_{16}$, $C_{10}H_{14}O$, $C_{10}H_{16}O$, $C_{10}H_{18}O$, and $C_{10}H_{20}O$) contributed 42% of the total VOC emissions (by mixing ratio). Besides terpenes, eight VOCs were identified, including C_6H_{10} (detected at *m/z* 83, possibly cyclohexene and hexadiene) [98], C_7H_6 (detected at *m/z* 91, possibly a xylene fragment) [85], C_7H_8 (detected at *m/z* 93, possibly toluene) [99], C_7H_{10} (detected at *m/z* 95, possibly 1-methyl cyclopentene and 2-norbornene) [100,101], C_7H_{12} (detected at *m/z* 97, possibly norbornane and cycloheptene) [102], C_8H_8 (detected at *m/z* 105, possibly styrene) [67], $C_{10}H_{14}$ (detected at *m/z* 135, possibly m-cymene, diethyl benzene, and butylbenzene) [99], and $C_{10}H_{12}O_2$ (detected at *m/z* 165, possibly phenethyl acetate) [103]. For scented wax warmer product B (Fig. 2b), terpenes contributed 78% of the total VOC emissions (by mixing ratio). Four VOCs were identified besides terpenes, including C_8H_{10} (detected at *m/z* 107, possibly xylene) [104], C_8H_{12} (detected at *m/z* 109, possibly 2-methylenenorbornane) [105], $C_8H_8O_2$ (detected at *m/z* 137, possibly methyl benzoic acid and methyl benzoate) [67,103], and $C_9H_{14}O$ (detected at *m/z* 139, possibly limona ketone) [106,107]. The emission of non-terpene VOCs could be attributed to other fragrance additives incorporated into the wax or terpene derivatives via reactions with indoor O_3/NO_x. For instance, $C_9H_{14}O$ was reported as a secondary ozonide that is produced via the ozonolysis of D-limonene [107]. All unidentified *m/z* values and unknown fragment ions are categorized as “others”. As terpene emissions were significant during the use of scented wax products, it is important to further evaluate the five terpene species to explore their impact on the indoor atmospheric environment.

3.2. Temporal variations in indoor monoterpene, monoterpenoid, NO/NO_2, and O_3 concentrations during the use of scented wax products in the Purdue zEDGE Test House

Real-time indoor VOC measurements via PTR-TOF-MS enabled the evaluation of temporal emission profiles for selected monoterpenes and monoterpenoids while using scented wax products. Table S3 reports the precise *m/z* of each terpene and the possible compounds associated with their chemical formulas. Fig. 3a and 3b are representative examples of time-dependent changes in the chemical composition of indoor air during the use of scented candle products B and C under AER ~ 6.5 h^{-1}. Panel A illustrates the mixing ratios of identified terpenes. Panel B illustrates the mixing ratios of nitrogen oxides (NO, NO_2). Panel C illustrates the mixing ratio of indoor ozone (O_3). Fig. 4a and 4b are representative examples of terpene emission profiles for scented wax warmer product B (AER ~ 6.5 h^{-1}) and product H (AER ~ 3.0 h^{-1}). Panel A illustrates the mixing ratios of identified terpenes and Panel B illustrates the mixing ratio of indoor O_3. NO and NO_2 emissions were not observed for scented wax warmer products as there was no combustion (NO + NO_2 ~ 0 ppb), thus, NO and NO_2 emission profiles are not provided for these two representative examples.

As shown in Figs. 3 and 4, the mixing ratios of terpenes emitted from both scented candles and wax warmers increased gradually over time, with small fluctuations attributed to temperature-dependent volatilization of terpenes from the wax during the melting process. According to Table 2, monoterpenes were detected in all tested products and were revealed as the dominant terpene species with 16 out of 19 products exhibiting peak $C_{10}H_{16}$ mixing ratios >20 ppb. The peak mixing ratio of monoterpenes varied significantly between tested products. For scented candles ($n = 5$), product C had the highest peak mixing ratio at 100.42 ppb and product D had the lowest at 5.64 ppb. For scented wax warmers ($n = 14$), product I had the highest peak at 258.18 ppb, and product C had the lowest at 7.60 ppb. Compared with the emissions of monoterpenes, the emissions of the four monoterpenoids were comparatively low, with peak mixing ratios mostly between 10^{-1} to 10^{1} ppb. For scented candles, only products A and C had emissions for all terpenes. Monoterpenoids $C_{10}H_{14}O$ and $C_{10}H_{20}O$ were not detected in products B, D, and E. For scented wax warmers, seven products had emissions for all terpenes. Monoterpenoids $C_{10}H_{16}O$, $C_{10}H_{18}O$, and $C_{10}H_{20}O$ were detected for more than 13 products. For both types of scented wax products, $C_{10}H_{14}O$ was the least detected monoterpenoid species during the source event (only detected in two scented candles and six scented wax warmers).

The absence and low mixing ratios of monoterpenoids does not necessarily suggest the absence of emissions or low emissions as the major detected product ions by PTR-TOF-MS may not be the protonated molecule of monoterpenoids. This is because the reduction of the electric field in the PTR-TOF-MS drift tube can cause fragmentation of the protonated VOCs [108,109]. According to the ion distribution demonstrated in previous studies, the major detected ions for eucalyptol ($C_{10}H_{18}O$) by PTR-TOF-MS are the fragment ion ($[C_6H_9]^+$, *m/z* 81.07, % abundance > 44.4) and the protonated monoterpene ($[(C_{10}H_{16})H]^+$, *m/z* 137.13, % abundance > 53.6); The % abundance of the protonated eucalyptol ($[(C_{10}H_{18}O)H]^+$, *m/z* 155.14) is lower than 0.7 [85,86]. This is because oxygen-containing monoterpenoids tend to lose water from their protonated molecules to produce abundant $[(C_{10}H_{16})H]^+$ ions [85]. The $[(C_{10}H_{16})H]^+$ ions can be further ionized to $[C_6H_9]^+$, which is a stable fragment ion detected for both monoterpenes and oxygen-containing monoterpenoids [85,110]. Thus, the targeted monoterpenoids may share $[C_6H_9]^+$ and $[(C_{10}H_{16})H]^+$ ions with monoterpenes, which means the mixing ratios of monoterpenoids could be

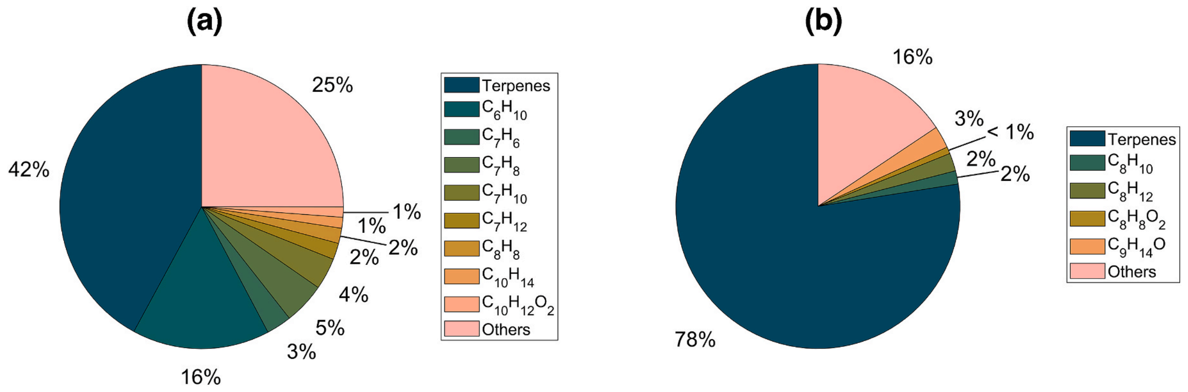

Fig. 2. Relative abundance of detected VOCs while using (a) scented candle product C and (b) scented wax warmer product B under AER ~ 6.5 h^{-1}. “Terpenes” represents the sum of monoterpenes and monoterpenoids ($\sum (C_{10}H_{16} + C_{10}H_{14}O + C_{10}H_{16}O + C_{10}H_{18}O + C_{10}H_{20}O)$). “Others” represents the sum of all unidentified *m/z* values and unknown fragments. The percentage is calculated based on the average mixing ratio for each compound during the source period of each experiment. All compounds with peak mixing ratios >1 ppb beyond background levels were considered.

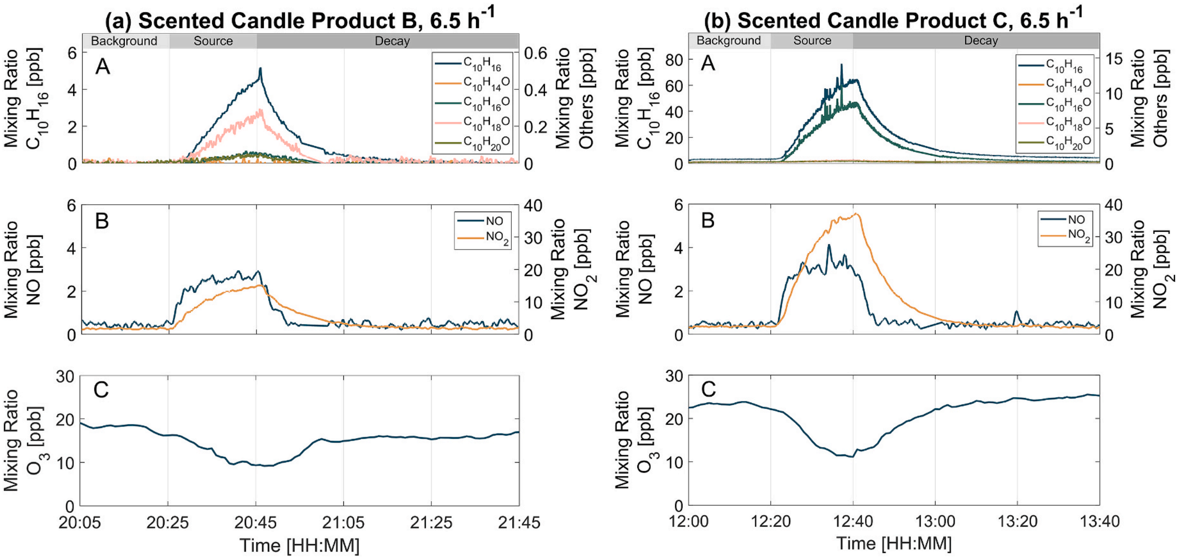

Fig. 3. (a) Time-series profiles for scented candle experiment using product B under AER ~ 6.5 h^{-1}; (b) Time-series profiles for scented candle experiment using product C under AER ~ 6.5 h^{-1}; (A) Mixing ratio of $C_{10}H_{16}$ (moderate blue line, left y-axis), $C_{10}H_{14}O$ (orange line, right y-axis), $C_{10}H_{16}O$ (teal line, right y-axis), $C_{10}H_{18}O$ (pink line, right y-axis), and $C_{10}H_{20}O$ (olive line, right y-axis). (B) Mixing ratio of NO (moderate blue line, left y-axis) and NO_2 (orange line, right y-axis). (C) Mixing ratio of O_3 (moderate blue line, left y-axis). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

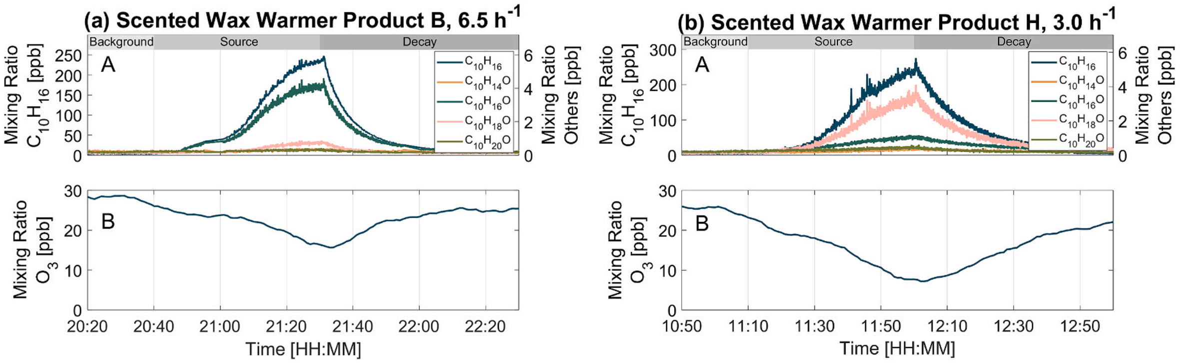

Fig. 4. (a) Time-series profiles for scented wax warmer experiment using product B under AER ~ 6.5 h^{-1}; (b) Time-series profiles for scented wax warmer experiment using product H under AER ~ 3.0 h^{-1}. (A) Mixing ratio of $C_{10}H_{16}$ (moderate blue line, left y-axis), $C_{10}H_{14}O$ (orange line, right y-axis), $C_{10}H_{16}O$ (teal line, right y-axis), $C_{10}H_{18}O$ (pink line, right y-axis), and $C_{10}H_{20}O$ (olive line, right y-axis). (B) Mixing ratio of O_3 (moderate blue line, left y-axis). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

under-estimated and the mixing ratios of monoterpenes could be over-estimated. Without a fastGC at the inlet of the PTR-TOF-MS, we are not able to separate these ions in this study.

Depletion of indoor O_3 occurred while using both scented candle and wax warmer products as shown in Figs. 3 and 4. However, NO and NO_2 emissions were only observed while using scented candle products. This is because the candle flame can reach over 1000 °C, which can initiate reactions between ambient oxygen and nitrogen to form NO and NO_2 [22,111–113]. The emission profiles of all candle experiments revealed lower NO mixing ratios compared to NO_2. The reduction in indoor O_3 mixing ratios and the lower NO mixing ratios is likely due to the reaction between the emitted NO and indoor O_3 as NO can react with O_3 to form NO_2 [43,79,114,115]. In addition to reactions between NO_x and O_3, another process that can cause the observed decrease in O_3 is the ozonolysis of the emitted terpenes. O_3 can oxidize terpenes to form carbonyl byproducts and SOA, which in turn can reduce the concentration of indoor O_3. Since NO_x emissions were absent in experiments using scented wax warmers, we believe that the decrease in O_3 during wax warmer melting events is solely due to the ozonolysis of terpenes.

3.3. Terpene emission factors and emission rates for scented wax products

Measured terpene mixing ratios varied among the tested scented candle and wax warmer products (Table 2). However, this variability is due to variable amounts of products used (number of candles or wax warmer cubes), variable amounts of essential oils added to the wax, and the outdoor AER. Thus, normalization of the observed VOC concentrations is needed to compare the indoor terpene emission potential of candles and wax warmers. Emission factors (EFs) are a generalizable metric to quantify the total amount of VOCs emitted per unit of a product used. EFs provide a useful means to predict concentrations of gas-phase pollutants in different indoor environments. Besides EFs, emission rates (ERs) are also reported to quantify the amount of terpene emitted per unit time. For consumer products, EFs can be expressed as mass emitted per mass of product used [116], and ERs can be expressed as mass emitted per time. In this study, EFs are reported in mg per g, which is the mass of terpene emitted normalized by the mass of wax consumed during the source event period (Eq. (8)). ERs are reported in mg per min, which is the mass of terpene emitted during the source event period normalized by the source event duration (Eq. (9)). Figs. 5

Table 2
Peak VOC mixing ratios of identified terpene species for each scented wax product experiment.

Scented Candle Product Experiment	Peak VOC Mixing Ratio (ppb)				
	$C_{10}H_{16}$	$C_{10}H_{14}O$	$C_{10}H_{16}O$	$C_{10}H_{18}O$	$C_{10}H_{20}O$
A1	44.84	0.59	0.52	6.00	0.94
A2	65.70	0.69	0.57	4.66	1.06
A3	46.83	0.78	0.60	4.13	6.30
B1	9.30	–	–	0.64	–
B2	8.33	–	–	0.38	–
B3	10.46	–	–	0.52	–
C1	76.81	0.54	11.16	0.56	0.43
C2	100.42	0.62	15.12	0.70	0.48
C3	59.88	0.52	8.93	0.48	0.42
D1	5.64	–	–	–	–
D2	6.67	–	–	–	–
D3	6.81	–	–	–	–
E1	24.99	–	0.65	0.43	–
E2	23.90	–	0.37	0.21	–
E3	22.07	–	0.38	0.20	–

Scented Wax Warmer Product Experiment	Peak VOC Mixing Ratio (ppb)				
	$C_{10}H_{16}$	$C_{10}H_{14}O$	$C_{10}H_{16}O$	$C_{10}H_{18}O$	$C_{10}H_{20}O$
A	36.49	5.16	0.52	1.50	7.48
B	247.86	0.44	4.61	0.88	0.42
C	7.60	–	0.17	0.30	0.25
D	59.89	–	2.25	–	1.18
E	75.18	1.10	0.51	1.66	0.51
F	21.50	–	3.93	0.58	0.36
G	52.41	–	2.08	0.70	0.26
H	190.66	0.38	0.88	2.20	0.48
I	258.18	0.61	6.03	1.94	1.01
J	69.63	–	0.56	0.34	0.88
K	64.61	5.55	28.97	8.82	–
L	42.58	–	6.08	0.51	0.33
M	48.70	–	3.63	1.89	0.33
N	170.56	0.83	11.04	2.05	0.60

and 6 provide the mean speciated EFs of target terpenes for each scented candle and wax warmer product, respectively. Table 3 provides the values of the calculated ERs of all terpenes for each scented wax product. For a clearer comparison between products, the EFs with a uniform *y*-axis are shown in Fig. S6 and the mean speciated values of EFs are provided in Table S5. As there were three identical experiments completed for each scented candle product, the EFs of terpenes for each experiment are provided in Fig. S7.

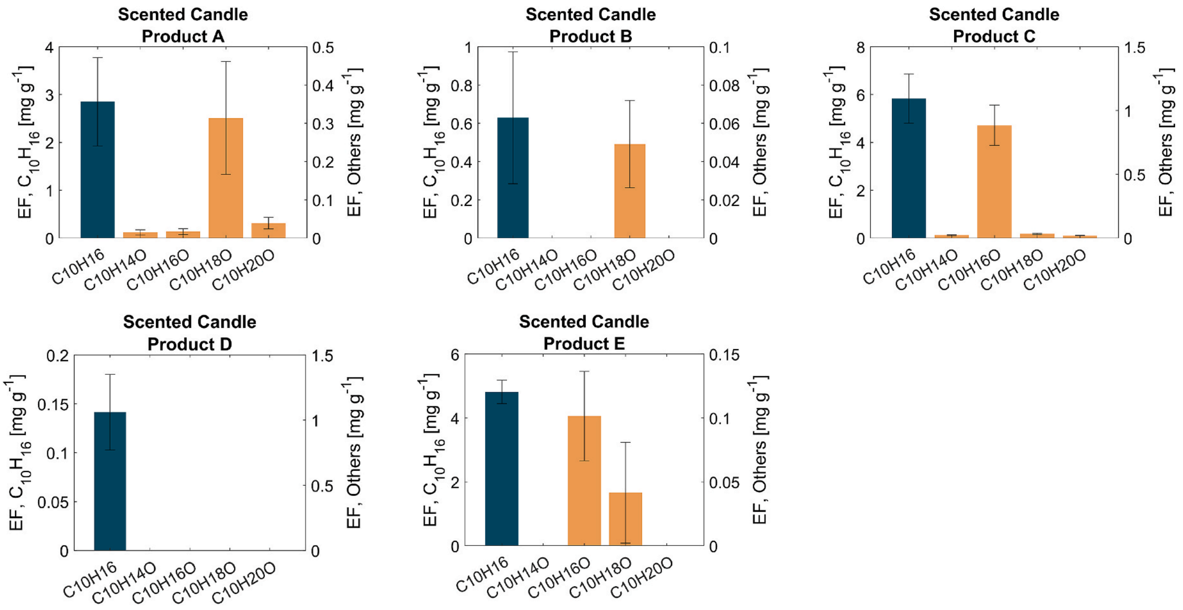

Fig. 5. Mean speciated emission factors (EFs) of $C_{10}H_{16}$ (left y-axis), $C_{10}H_{14}O$ (right y-axis), $C_{10}H_{16}O$ (right y-axis), $C_{10}H_{18}O$ (right y-axis), and $C_{10}H_{20}O$ (right y-axis) in mg per g wax consumed for scented candle products A to E (error bars represent the standard deviation of triplicate experiments for each product).

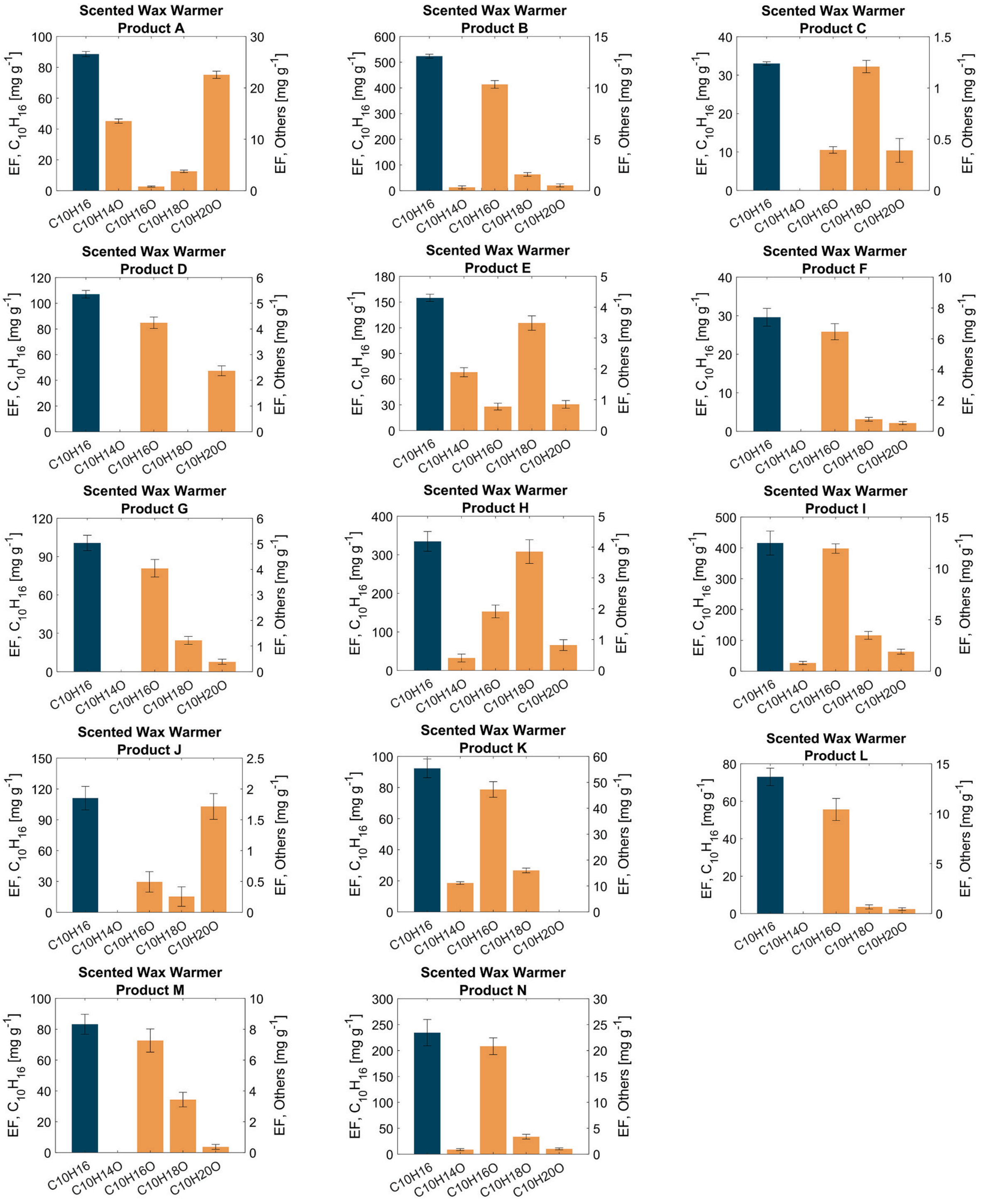

Fig. 6. Mean speciated emission factors (EFs) of $C_{10}H_{16}$ (left y-axis), $C_{10}H_{14}O$ (right y-axis), $C_{10}H_{16}O$ (right y-axis), $C_{10}H_{18}O$ (right y-axis), and $C_{10}H_{20}O$ (right y-axis) in mg per g wax consumed for scented wax warmer products A to N (error bars represent the standard deviation of individual experiments for each product).

For each product, both EFs and ERs for monoterpenes are significantly higher than monoterpenoids. For scented candles, product C has the highest monoterpene EF and ER (5.833 mg g^{-1} and 2.625 mg min^{-1}, respectively) and product D has the lowest (0.141 mg g^{-1} and 0.048 mg min^{-1}, respectively). The EFs of monoterpenoids emitted from scented candle products are mostly < 0.1 mg g^{-1} and ERs are mostly < 0.07 mg min^{-1}, except for product C which has an EF and ER for $C_{10}H_{16}O$ at 0.844 mg g^{-1} and 0.398 mg min^{-1}. For scented wax warmers, product B has the highest monoterpene EF and ER (523.368 mg g^{-1} and 5.234 mg min^{-1}, respectively) and product F has the lowest EF (29.602 mg g^{-1}). Based on empirical data, the EFs of terpenes emitted from scented wax products are positively correlated with ERs. For monoterpenoids, scented wax warmer product A contributes the highest EFs for $C_{10}H_{14}O$ and $C_{10}H_{20}O$ at 13.533 mg g^{-1} and 22.544 mg g^{-1}, respectively. Scented wax warmer product K contributes the highest EFs for $C_{10}H_{16}O$ and $C_{10}H_{18}O$ at 47.248 mg g^{-1} and 16.013 mg g^{-1}, respectively. Due to the measurement limitations of the PTR-TOF-MS mentioned in Sections 2.2 and 3.2, the EFs for monoterpenes could be over-estimated and the EFs for monoterpenoids could be under-estimated. To avoid the impact of the underestimation of monoterpenoids, the total EF (sum of all five

Table 3
Emission rates (ERs) of identified terpene species for each scented wax product.

Scented Candle Product	Emission Rate (ER, mg min^{-1})				
	$C_{10}H_{16}$	$C_{10}H_{14}O$	$C_{10}H_{16}O$	$C_{10}H_{18}O$	$C_{10}H_{20}O$
A	0.646 ± 0.206	0.003 ± 0.001	0.004 ± 0.001	0.070 ± 0.028	0.009 ± 0.003
B	0.126 ± 0.069	–	–	0.010 ± 0.005	–
C	2.625 ± 0.462	0.010 ± 0.002	0.398 ± 0.071	0.015 ± 0.002	0.008 ± 0.001
D	0.048 ± 0.013	–	–	–	–
E	0.801 ± 0.010	–	0.016 ± 0.006	0.007 ± 0.006	–
Scented Wax Warmer Product	**Emission Rate (ER, mg min^{-1})**				
	$C_{10}H_{16}$	$C_{10}H_{14}O$	$C_{10}H_{16}O$	$C_{10}H_{18}O$	$C_{10}H_{20}O$
A	0.886	0.135	0.008	0.038	0.225
B	5.234	0.003	0.103	0.016	0.005
C	0.132	–	0.002	0.005	0.002
D	1.070	–	0.042	–	0.024
E	1.549	0.019	0.008	0.035	0.009
F	0.296	–	0.065	0.008	0.005
G	1.007	–	0.040	0.012	0.004
H	3.347	0.004	0.019	0.039	0.008
I	4.157	0.008	0.119	0.035	0.019
J	1.111	–	0.005	0.003	0.017
K	0.924	0.111	0.472	0.160	–
L	0.730	–	0.104	0.007	0.004
M	0.833	–	0.073	0.034	0.004
N	2.345	0.009	0.208	0.034	0.010

terpene EFs) for each product is provided in Fig. S8.

As higher EFs indicated a higher level of VOC emission per mass of wax consumed, the results illustrate that scented wax warmer products tend to have higher terpene emission potentials compared to scented candle products. This is important as scented wax warmers are widely sold in retail stores in the U.S. and are a popular alternative to scented candles as they produce aromas without combustion, nor require the use of ultrasonic diffusers. As the molecular structures of terpenes consist of one or more C=C bonds, this chemical property causes terpenes to have a high reactivity with O_3, NO_x, and OH to form secondary products [47,117]. The high flame temperature while burning candles can accelerate the oxidation of terpenes to form oxygen-containing products, acetones, aldehydes, SOA, and other oxidation products that are not evaluated in this study [42,118,119]. Thus, the lower terpene EFs for scented candle products could be driven by the high oxidation reactivity of terpenes within the candle flames.

The emission of terpenes during the use of scented wax products is dependent on various properties, including the type of wax used and the amount and types of essential oils used. It has been reported that the quality of wax can strongly influence the emission of chemical air pollutants [23]. As soft wax can melt faster and emit higher concentrations of VOCs compared to hard wax within a certain period, it is possible that wax warmers are produced with softer wax for the purpose of an easy melt, which contributes to higher terpene EFs and ERs. Previous studies have reported that essential oils usually contain various terpene species as their constituents [116]. According to Table S3, the saturation concentration (C*) varies among terpenes. Greater C* values indicate higher compound volatility. This suggests that products containing higher volatility terpene species such as α-pinene, β-pinene, and eucalyptol can contribute to higher terpene emissions. As the essential oils used in scented wax products are not provided on the product labels, we are not able to know the exact terpene species used in these products. There is currently a lack of research evaluating the amount and types of essential oils used in fragranced consumer products and their associated terpene emission profiles. However, the labeled fragrances can still provide some references for consumers to predict potential terpene emissions. According to Table 1, the fragrances of wax warmer product E are lime and eucalyptus, and Fig. 6 has revealed that the EF of $C_{10}H_{18}O$ (possibly eucalyptol) is the highest among the four monoterpenoids.

Although terpenes are widely used in fragranced consumer products [120], their contribution to total VOC emissions varies among different products. For scented wax products, terpene species were found to be the most abundant VOCs emitted to indoor air (Fig. 2). During the use of other fragranced consumer products, such as hair care products, significant emissions of non-terpenes have been reported, including siloxane species (e.g., D4 – D6) [121–123]. Different fragranced consumer products have varying concentrations of terpenes and can exist in different forms (e.g., solid wax or liquid spray). Such factors can influence mass transfer processes, resulting in variable terpene ERs and EFs. Table 4 has summarized monoterpene ERs and EFs for different fragranced products, including scented wax products, personal care products, and air fresheners, and for a Christmas tree. Scented wax warmers have similar or greater monoterpene ERs and EFs compared to other household and consumer products that are commonly used in the home. Furthermore, a previous study has reported a monoterpene ER of 0.2 mg min^{-1} while burning a scented candle, which is within the monoterpene ER range of this study [22].

3.4. Human inhalation exposure to terpenes released from scented candles and wax warmers

In addition to the evaluation of terpene emissions via calculation of EFs and ERs, the potential impact of using scented wax products on human inhalation exposures to VOCs under different outdoor AERs is also evaluated through the determination of inhalation intakes (Fig. 7) and inhalation intake fractions (iFs) (Fig. 8). The inhalation intake is the mass of VOC inhaled by an occupant (Eq. (10)), and the iF is the ratio of the VOC mass inhaled by an individual to the VOC mass emitted into an indoor environment during a specific period (Eq. (11)). As people tend to engage in only light activities (e.g. reading) while using scented wax products, inhalation intakes and iFs are calculated based on an inhalation rate of 1.25 m^3 h^{-1} [69]. Values of inhalation intakes and iFs are

Table 4
Comparison of monoterpene emission rates (ERs) and emission factors (EFs) from different fragranced consumer products and a Christmas tree. The value in brackets represents the mean value of all tested products in a particular study.

Fragranced Consumer Product	$C_{10}H_{16}$ ER (mg min^{-1})	$C_{10}H_{16}$ EF (mg g^{-1})	Ref.
Scented candles (this study)	0.13–2.63 (0.85)	0.14–5.83 (2.85)	–
Scented wax warmers (this study)	0.13–5.23 (1.69)	29–523 (179)	–
Botanical disinfectants	–	0.1–10.9 (3.24)	[69]
Hair care products	0.07–2.4 (0.39)	0.16–13.48 (2.06)	[123]
Air fresheners	0.025–0.13	–	[124]
Scented sprays	0.15	–	[7]
Christmas tree	0.2	–	[125]

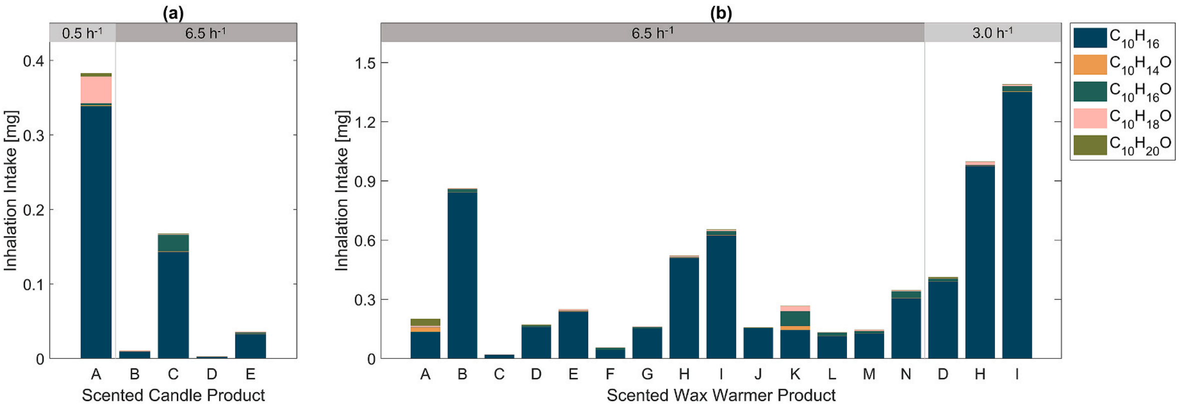

Fig. 7. Inhalation intake for each scented wax product for $C_{10}H_{16}$ (moderate blue), $C_{10}H_{14}O$ (orange), $C_{10}H_{16}O$ (teal), $C_{10}H_{18}O$ (pink), and $C_{10}H_{20}O$ (olive) in mg for (a) scented candle product A (AER ~ 0.5 h^{-1}) and products B to E (AER ~ 6.5 h^{-1}); (b) scented wax warmer products A to N (AER ~ 6.5 h^{-1}) with products D, H, and I repeated under a lower AER (AER ~ 3.0 h^{-1}). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

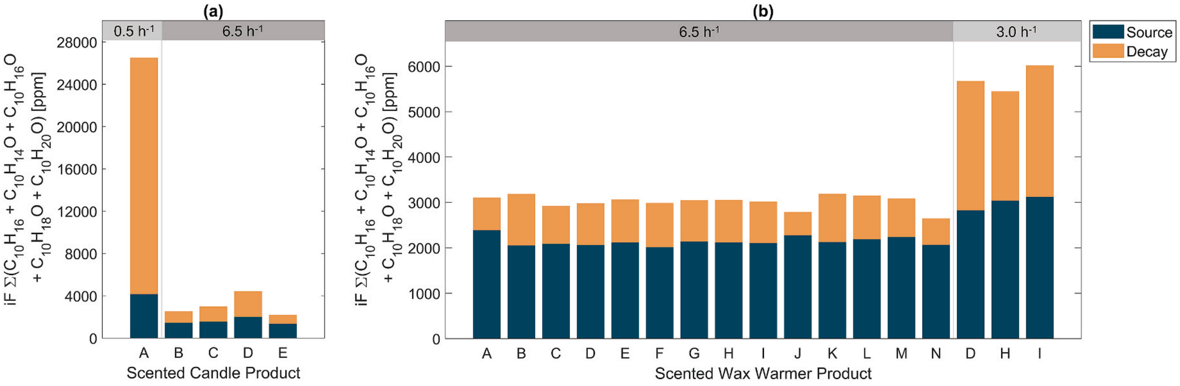

Fig. 8. Inhalation intake fractions (iFs) for terpenes ($\sum$ ($C_{10}H_{16}$ + $C_{10}H_{14}O$ + $C_{10}H_{16}O$ + $C_{10}H_{18}O$ + $C_{10}H_{20}O$)) for (a) scented candle product A (AER ~ 0.5 h^{-1}, source period = 20 min, decay period = 120 min), and products B to E (AER ~ 6.5 h^{-1}, source period = 20 min, decay period = 60 min); and (b) scented wax warmer products A to N (AER ~ 6.5 h^{-1}, source period = 50 min, decay period = 60 min) with product D, H, and I repeated under a lower AER (AER ~ 3.0 h^{-1}, source period = 50 min, decay period = 60 min). The source period is presented in moderate blue and the decay period is presented in orange. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

also summarized in Table S6 and Table S7, respectively. In this study, the reported iF of each product is the overall iF for all terpene species (sum of $C_{10}H_{16}$, $C_{10}H_{14}O$, $C_{10}H_{16}O$, $C_{10}H_{18}O$, and $C_{10}H_{20}O$). Thus, the under- and over-estimation of terpene emissions due to fragmentation in the PTR-TOF-MS drift tube will not affect the comparison of terpene exposures between products.

The inhalation intakes of all five terpenes for each scented candle and scented wax warmer product are illustrated in Fig. 7a and 7b, respectively. The total terpene mass intake for scented candle products tested under AER ~ 6.5 h^{-1} (products B to E) range between 0.002 and 0.17 mg, while the total intake for scented wax warmer products under AER ~ 6.5 h^{-1} (products A to N) range between 0.02 and 0.86 mg. The predominant species being inhaled by an occupant are monoterpenes, which for 16 out of 19 products contribute more than 85% to the total intake. The total inhalation intakes are also significantly higher when using scented wax products with a lower AER. The total intakes of scented candle products B to E under AER ~ 6.5 h^{-1} are all $<$ 0.17 mg. For scented candle product A under AER ~ 0.5 h^{-1}, the total intake is $>$ 0.38 mg. As scented candle product A is the only product that was tested under AER ~ 0.5 h^{-1}, we were not able to provide a comparison between high and low AERs with the same scented candle product. However, this comparison is available for scented wax warmer products. For scented wax warmers, products D, H, and I were examined under two ventilation conditions – AER ~ 6.5 h^{-1} and AER ~ 3.0 h^{-1}. The total inhalation intake for products D, H, and I at AER ~ 6.5 h^{-1} are 0.17, 0.52, and 0.65 mg, respectively. After we adjust the ventilation condition of zEDGE to AER ~ 3.0 h^{-1}, the total inhalation intake of these products became 0.41, 1.00, and 1.39 mg, respectively. The lower outdoor AER indicates less ideal ventilation conditions, which consequently, increase human exposure to terpenes.

Fig. 8 illustrates the terpene iFs for each scented candle and wax warmer product under their outdoor AER condition during the source and decay periods of the experiments. The iFs for scented candle products B to D (AER ~ 6.5 h^{-1}) range between 2100 and 4500 ppm (source + decay), which indicates that 0.21% – 0.45% of terpenes emitted into the indoor atmosphere can be inhaled during the use of scented candles in a well-ventilated residential building. The iFs for scented wax warmer products A to N (AER ~ 6.5 h^{-1}) range between 2600 and 3200 ppm (source + decay), which indicates that 0.26% – 0.32% of terpenes emitted into indoor air can be inhaled during the use of scented wax warmers in a well-ventilated indoor environment. It is interesting to note that the lower AER will increase the iFs during the concentration decay period. Fig. 8a indicates that the iFs during the decay period contribute to greater than 84% of the total iFs for scented candle product A at AER ~ 0.5 h^{-1}, while for scented candle products B to D at AER ~ 6.5 h^{-1} the decay period only contributed to less than 55% of the total iFs. Fig. 8b indicates that the iFs for the decay periods while using scented wax warmer products D, H, and I are only contributing to around 30% to the total iFs under an AER ~ 6.5 h^{-1}. At a lower AER of 3.0 h^{-1}, this proportion increased to almost 50%. This means that the large increase in the terpene inhalation intakes at low AERs is due to the increase in the inhalation intake during the concentration decay period after the product was removed from zEDGE.

The increase in the total terpene iFs has highlighted the importance of using scented wax products under well-ventilated conditions to minimize human exposure to terpenes, especially during the decay period. Other exposure metrics, such as the product intake fraction (PiF) or individual product intake fraction (PiF_i), are not applicable in this study as the mass of terpene used in the scented wax products is not declared by the manufactures [57,126]. In addition to inhalation intake, dermal uptake of terpenes via penetration into human skin may also cause health risks. It has been demonstrated that direct dermal uptake of monoterpenoids (e.g., linalool, eugenol, and α-terpineol) can occur at rates that are comparable to or larger than inhalation intake rates [127]. Thus, future research is needed to evaluate the dermal uptake of different terpenes during the use of scented wax products in residential buildings. It is important to note that the calculated inhalation intakes and iFs of terpenes for scented candle and wax warmer products can only provide human exposure information for selected VOCs released from these products. Other factors, such as exposure to direct emissions of other gas- and particle-phase pollutants and exposure to secondary products (e.g., SOA) should also be considered in future works.

4. Conclusions

This study has shed light on the potential indoor environmental and human exposure risks associated with terpene emissions during the use of scented wax products in residential buildings. Online indoor VOC measurements with a proton transfer reaction mass spectrometer revealed that scented candles and wax warmers/melts released a variety of monoterpenes and monoterpenoids to the indoor atmosphere during their use. Speciated emission factors and emission rates suggest that the use of scented wax warmer products can emit greater amounts of reactive terpenes per mass of wax consumed compared to scented candle products. However, this does not suggest that burning scented candles has less of an impact on indoor air quality, as candles co-emitted NO and NO_2, which are not released from wax warmers. As scented wax warmers also have higher emission factors and emission rates compared to other fragranced consumer products, people may be exposed to higher terpene concentrations while using scented wax warmers in their homes. It was also determined that low building ventilation rates can significantly increase human exposure to VOCs, especially during the concentration decay period. Our results demonstrate that at low ventilation rates, occupants can inhale more terpene mass after the scented wax product is removed from an indoor space compared to the active emission period.

The findings of this study highlight the need for further research on indoor air quality in residential buildings during the use of scented wax warmers. The emission of a variety of monoterpenes and monoterpenoids that are highly reactive with ozone suggests that scented wax warmers may initiate indoor SOA formation events due to terpene ozonolysis. Thus, such products have the potential to induce multiphase inhalation exposures to secondary gas- and particle-phase species. In addition, given the increasing contribution of volatile chemical products to the VOC burden of the urban atmosphere, it is possible that scented wax products can contribute to outdoor concentrations of terpenes via indoor-to-outdoor VOC transport in densely populated cities where such products are widely used in the home environment.

CRediT authorship contribution statement

Jianghui Liu: Writing – review & editing, Writing – original draft, Visualization, Validation, Methodology, Investigation, Formal analysis, Data curation. **Jinglin Jiang:** Writing – review & editing, Methodology, Investigation, Formal analysis, Data curation. **Xiaosu Ding:** Writing – review & editing, Methodology, Investigation, Formal analysis, Data curation. **Satya S. Patra:** Writing – review & editing, Methodology, Investigation, Formal analysis, Data curation. **Jordan N. Cross:** Writing – review & editing, Methodology, Investigation. **Chunxu Huang:** Writing – review & editing, Methodology, Investigation, Formal analysis, Data curation. **Vinay Kumar:** Writing – review & editing, Methodology, Investigation, Formal analysis, Data curation. **Paige Price:** Writing – review & editing, Methodology, Investigation, Formal analysis, Data curation. **Emily K. Reidy:** Writing – review & editing, Methodology, Investigation, Formal analysis, Data curation. **Antonios Tasoglou:** Writing – review & editing, Resources, Methodology, Investigation, Formal analysis, Data curation. **Heinz Huber:** Writing – review & editing, Resources, Methodology, Investigation, Formal analysis, Data curation. **Philip S. Stevens:** Writing – review & editing, Supervision, Resources, Project administration, Methodology, Investigation, Funding acquisition, Formal analysis, Data curation, Conceptualization. **Brandon E. Boor:** Writing – review & editing, Writing – original draft, Visualization, Validation, Supervision, Resources, Project administration, Methodology, Investigation, Funding acquisition, Formal analysis, Data curation, Conceptualization. **Nusrat Jung:** Writing – review & editing, Writing – original draft, Visualization, Validation, Supervision, Software, Resources, Project administration, Methodology, Investigation, Funding acquisition, Formal analysis, Data curation, Conceptualization.

Declaration of competing interest

The authors declare that they have no conflict of interest.

Data availability

Data will be made available on request.

Acknowledgements

Financial support was provided by Purdue University (start-up funds to N.J.), the Alfred P. Sloan Foundation (G-2018-11061 to P.S.S. and B. E.B), and the National Science Foundation (CBET-1847493 to B.E.B.). The authors wish to thank Dr. Case Tompkins in the Lyles School of Civil Engineering at Purdue University for his valuable guidance on technical writing.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.buildenv.2024.111314.

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