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Determination of terpene levels after the use of essential oil diffusers in vehicles and studio apartments using passive sampling

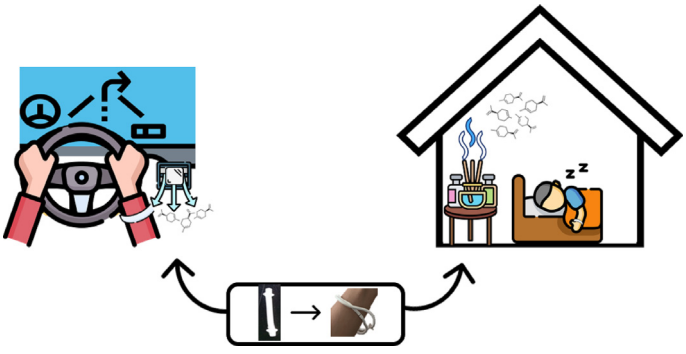
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HIGHLIGHTS

- The exposure to terpenes by using a fragranced product was assessed.
- ePTFE passive sampler was modified and made wearable.
- The use of essential oil diffusers significantly elevated terpene level in vehicles.
- The passive sampler was proven to be an alternative for personal exposure monitoring.

GRAPHICAL ABSTRACT



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ABSTRACT

The exposure levels of selected terpenes (limonene, α- and β-pinenes, and γ-terpinene) emitted by essential oil diffusers in vehicles and studio apartments were assessed using a passive sampling method. A previously developed passive sampler composed of an expanded polytetrafluoroethylene membrane and adsorbent (ePTFE PS) was enlarged and made wearable. Before field deployment, the sampling performance of the modified ePTFE PS for selected terpenes was compared with that of active sampling in a lab-scale 5 m^3 test chamber under constant exposure conditions, supporting that passive sampling provides reasonable estimates of the time-weighted exposure concentration. Fifty volunteers were recruited and asked to wear the ePTFE PS while using an essential oil diffuser inside their own vehicle while commuting and in their studio apartment while sleeping. Terpene levels without an essential oil diffuser were very low in vehicles and 47, 3.6, 1.6, and 0.62 μg m^{-3} for average concentrations of limonene, α- and β-pinenes, and γ-terpinene in studio apartments, respectively, close to those reported in previous studies. The indoor concentrations of all selected terpenes in vehicles and studio apartments were elevated by the use of essential oil diffusers, especially in vehicles. The average concentration of limonene in vehicles after the use of essential oil diffusers was 11 μg m^{-3}, which was greater than that before use by a factor of 30. Therefore, cautious usage of essential oil diffusers indoors where the volume is limited, such as a vehicle, is needed to reduce exposure to terpenes.

Abbreviations: ePTFE, expanded polytetrafluoroethylene; GC-MS, gas chromatography-mass spectrometry; MDL, method detection limit; PS, passive sampler; VOCs, volatile organic compounds.

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1. Introduction

Terpenes, such as limonene, α- and β-pinenes, and γ-terpinene, are widely used in fragrance products worldwide (Chiang et al., 2010; Chiu et al., 2009; Francis and Stusdal, 2014; Nematollahi et al., 2021; Steinemann, 2019a). They possess various health benefits for humans, including anti-insect, antimicrobial, and anti-herbivore properties (Himejima et al., 1992; Popova et al., 2009), as well as being used as medicinal reagents for various ailments (Bahramsoltani et al., 2015; Duschatzky et al., 2005; Jirtle et al., 1993; Jung et al., 2006; Nogueira and Lopes, 2011). However, they may also act as indoor sources of volatile organic compounds (VOCs) (Nematollahi et al., 2019; Steinemann, 2015; Steinemann, 2019b). An essential oil diffuser, one of the most frequently used fragrance products, is a complex mixture of chemicals, including terpenes (Angulo et al., 2020; Huang et al., 2012b; Nematollahi et al., 2018). Although using an essential oil diffuser has potential beneficial effects on human health, including psychological effects on anxiety and antimicrobial effects (De Sousa et al., 2015; Laird and Phillips, 2012; Su et al., 2007; Whiley et al., 2018; Zhang and Yao, 2019), negative side effects are also suspected. Terpenes can act as allergens, potentially resulting in skin irritation, asthma, and chest tightness (Huang et al., 2012a; Sarkic and Stappen, 2018; Su et al., 2007; Wolkoff and Nielsen, 2017). For example, a 100 % increase in the level of indoor limonene was associated with a 17–18 % relative increase in both wheezing and asthma and a 1.55 % increase in exhaled nitric oxide (Dales and Cakmak, 2019). These adverse effects were significantly greater in children (approximately two-fold). Thresholds for eye irritation of limonene ranged from 1700 to 3400 mg m^{-3} (Mølhave et al., 2000). Thus, limonene has been classified as potentially hazardous according to the hazardous chemical information system of Australia (Australia Safe Work, 2020). In addition, secondary products are produced from the reaction of terpenes with oxidants such as ozone and hydroxyl radical (Carslaw, 2013; Huang et al., 2012b; Nørgaard et al., 2014; Sarwar and Corsi, 2007), forming unwanted byproducts that cause adverse effects on human health (Steinemann, 2016). As humans spend approximately 90 % of their lives indoors, indoor air quality is of prime importance for human health (Klepeis et al., 2001; Robinson and Nelson, 1995). Therefore, an effective method for detecting terpene levels in indoor environments is required (Guo et al., 2004; McAlary et al., 2015; Mølhave, 1991; Weisel et al., 2008; Zhou et al., 2011).

Generally, active sampling with high accuracy and competence for short-term monitoring is used to assess exposure to VOCs (Amaral et al., 2010; Bohlin et al., 2007; Gallego et al., 2017; Horii et al., 2017). However, due to fluctuating concentrations and use of bulky equipment, active sampling is unfavorable for relatively long-term monitoring (Li et al., 2020; Seethapathy et al., 2008; Vallecillos et al., 2020; Vallecillos et al., 2021; Vallecillos et al., 2019). On the other hand, passive sampling is simple, convenient, and inexpensive compared with active sampling (Bohlin et al., 2007). In the case of relatively long-term monitoring, passive sampling may be an appropriate alternative to active sampling because it determines the time-weighted average exposure under fluctuating concentrations (Chen and Pawliszyn, 2003; Martos and Pawliszyn, 1999; Namieśnik et al., 2005; Wennrich et al., 2002). However, passive sampling also has several limitations. Environmental factors including air turbulence, temperature, and humidity may influence passive sampling (Lan et al., 2020). Compared with active sampling, the general accuracy of passive sampling could be lower by a factor of 3 (Bohlin et al., 2007; Gioia et al., 2007).

In our previous study, we developed a passive sampler composed of an expanded polytetrafluoroethylene membrane with an adsorbent (ePTFE PS) to detect VOCs in indoor air (Kim et al., 2021). It showed high accuracy compared to real-time monitoring of VOCs using proton-transfer-reaction mass spectrometry. Its sampling rate is rarely affected by air turbulence around the sampler, and the overall mass transfer is determined by the resistance in the porous membrane tube of the sampler. Considering the mass of the inserted absorbent, the ePTFE PS has higher sampling rates than other passive samplers reported in the literature (Kim et al., 2021). Because the tubes and absorbents used were relatively cheaper than other materials, the ePTFE PS is also cost-effective. However, compared to active sampling, the sampling rates of ePTFE PS for VOCs were lower (1.2–1.7 L h^{-1}). The recommended active sampling rates for the determination of toxic organic compounds in ambient air range from 0.6 to 12 L h^{-1} (Woolfenden and McClenny, 1999), while the typical sampling rates of low-volume air samplers for VOCs range from 0.1 to 10 L min^{-1} (Araki et al., 2009; Melymuk et al., 2011; Morales-McDevitt et al., 2021), requiring an increase in the passive sampling rate for routine VOCs monitoring.

The main objectives of this study were to improve the ePTFE PS for monitoring VOCs at lower concentrations within a reasonably short exposure duration and to monitor the exposure level of terpenes from an essential oil diffuser, a common fragrance product. The ePTFE PS was enlarged 10-fold and made wearable for personal exposure assessment of VOCs at lower exposure concentrations. The sampling performance of the modified ePTFE PS was tested by comparing it with a low-volume air sampler in a 5.0 m^3 test chamber at constant terpene concentrations. To assess the exposure to terpenes in vehicles and studio apartments using an essential oil diffuser, 50 participants were recruited, and their personal exposure levels were monitored before and after the use of the essential oil diffusers.

2. Materials and methods

2.1. Chemicals and instrumental analyses

Limonene, α- and β-pinene, and γ-terpinene were purchased from Sigma-Aldrich (St. Louis, MO, USA). The internal standards toluene-d_8 and chlorobenzene-d_5 were purchased from Sigma-Aldrich. To extract terpenes from the absorbent, acetone (Sigma-Aldrich) spiked with the internal standards was used. Gas chromatography–mass spectrometry (GC–MS) equipped with the Agilent 6890A gas chromatograph and Agilent 5973 N mass selective detector with the Agilent 7683 auto-injector and Agilent DB-5 column (30 m × 0.25 mm × 0.25 μm) was used for analyses. Helium was used as the carrier gas at a flow rate of 1.0 mL min^{-1}. The oven temperature was set at 40 °C for 2 min, increased to 100 °C at 10 °C min^{-1}, and then to 270 °C at 10 °C min^{-1}. The injection volume was 2 μL, with a split ratio of 5:1.

2.2. Passive sampler

The passive sampler developed in our previous study (Kim et al., 2021) was improved to enhance the sampling rate. The sampler was made of 500 mm expanded PTFE tubes (Huizhou Besteflon Industries Co. Ltd.) (ID 2 mm; OD 4 mm) inserted with 0.9 g Optipore-L493 (Sigma Aldrich) as the adsorbent with a high sorption capacity for VOCs (Bonifacio et al., 2017). The tubes were cleaned via Soxhlet extraction for 12 h with a mixture of 50 mL *n*-hexane and 50 mL acetone. The ePTFE PS was wearable like a wristband, as shown in Fig. S1 (Supplementary Material). After exposure, the ePTFE PS was submerged in a 40-mL vial containing 30 mL of acetone for 1 d to extract terpenes. Terpene concentration in acetone was analyzed using GC–MS.

The kinetics of the ePTFE PS (diffusion coefficient and sampling rate) was measured in a 125-L acrylic chamber. Limonene, α- and β-pinenes, and γ-terpinene were injected into the chamber at volumes of 0.5, 1, and 1.5 μL, respectively. Three ePTFE PS were placed in the test chamber, and extraction was performed for 5, 10, and 15 min after deployment. Mass accumulation rates were measured using linear regression between the extracted mass of terpenes and the sampling time. The sampling rates were then obtained using linear regression of the mass accumulation rate versus the concentration of terpenes in the chamber. The diffusion coefficients of the terpenes in the ePTFE PS tube were determined using Eq. (1).

$$M_{\mathrm{ePTFE}} = \frac{2\pi C_g t D_{\mathrm{ePTFE}} L}{\ln\left(r_1/r_0\right)} \quad (1)$$

where M_{ePTFE} is the mass of terpenes in the ePTFE tubes [μg], t is the

exposure time [s], L is the length of the sampler [m], r_1 and r_0 are the outer and inner radii of the ePTFE tube [m], respectively, D_{ePTFE} is the diffusion coefficient in the ePTFE tube [$m^2\ s^{-1}$], and C_g is the terpene gas phase concentration in air [$\mu g\ m^{-3}$]. Derivation of Eq. (1) is shown in the Supplementary Material (Kim et al., 2021).

For comparison, the diffusion coefficients and sampling rates of short ePTFE PS for limonene and α-pinene were determined from their absorption kinetics measured in a 125 L acrylic chamber. Limonene and α-pinene were injected into the chamber at volumes of 1.3, 2.5, and 3.8 μL. Three ePTFE PS were placed in the chamber, and extraction was performed after 10, 20, and 30 min.

2.3. Determination of time-weighted average concentration in a test chamber

Before the consumer panel study, the performance of the modified ePTFE PS was tested in a 5.0 m^3 test chamber (5.0 m^3, 1.70 m × 1.55 m × 1.90 m) made with stainless steel. The temperature and humidity of the chamber were maintained at 25 ± 0.5 °C and 50 ± 3 %, respectively. The air change rate was 0.5 h^{-1}. Limonene and α- and β-pinenes were spiked via vaporization from a capillary bottle. Three ePTFE PS were deployed in the chamber, and samples were collected after 24, 48, and 72 h. Active sampling using a low-volume air sampler (MP-Σ30KN Mini Pump, Taewon Sibata, Republic of Korea) was also conducted to measure terpene concentration in the chamber. The pump sampling rate was 0.1 L min^{-1} for 30 min and 200 mg Tenax-TA® (KnR, Republic of Korea) was used as adsorbent. Active sampling was performed at 0, 24, 48, and 72 h. Considering the amount and adsorption capacity of Optipore-L493 and Tenax-TA®, active sampling time was much shorter than passive sampling time to avoid overloading.

2.4. Consumer panel study

To detect the levels of terpenes indoors while using essential oil diffusers in vehicles and studio apartments, a panel of 50 volunteers was recruited. They were asked to wear an ePTFE PS while they slept or drove. To choose the model essential oil diffuser, ten different commercial essential oil diffusers (five each for vehicle and residence) were purchased, and the target terpene content was analyzed (Table S1). Commercial essential oil diffusers with the highest concentrations of target terpenes were selected. Two microliters of the diffuser was added to 1 mL of acetone, and the concentrations of the target terpenes in acetone were measured using GC–MS. This panel study was approved by the Institutional Review Board (IRB) of Korea University (approval number: KUIRB-2022-0264). The panel study was conducted in November 2022.

To assess exposure in vehicles, four ePTFE PS, four 40-mL vials containing 30 mL of acetone, and one commercial essential oil diffuser were distributed to each of the 25 volunteers. All vehicles recirculated air inside. On the first day, all volunteers drove twice a day, in the morning and afternoon, while wearing an ePTFE PS on hand while driving. After driving, the ePTFE PS was submerged in the vials for extraction. After the second drive, an essential oil diffuser was installed in each vehicle. The following day, all volunteers drove twice a day, in the morning and afternoon, wearing an ePTFE PS on the wrist while driving. After driving, the ePTFE PS was submerged in the vials for extraction.

To assess exposure in studio apartments, two ePTFE PS, two 40-mL vials containing 30 mL of acetone, and one commercial essential oil diffuser were distributed to each of the 25 volunteers. On the first day, all the volunteers wore an ePTFE PS on their wrists while sleeping. After they woke up, the ePTFE PS was submerged into vials for extraction, and then essential oil diffusers were placed in their residence. After each day, all volunteers wore an ePTFE PS on their wrists while sleeping that night. When they woke up, the ePTFE PS was submerged in vials for extraction.

Assuming that the indoor temperature was not significantly different from 25 °C, obtained sampling rates of ePTFE PS for terpenes were used to calculate time-weighted average concentration in vehicles and studio apartments. Effects of temperature fluctuation on passive sampling rates of ePTFE PS require further investigations although the temperature range in indoor environments should be much narrower than those of outdoor environments.

2.5. Indoor air quality model

The steady-state concentrations of terpenes in vehicles and studio apartments were also estimated using a mass balance equation, considering the generation and elimination rates of terpenes (VanLoon and Duffy, 2017). In previous studies, the outdoor concentration of terpenes was negligible compared with the indoor concentration (Hodgson et al., 2002; Rehwagen et al., 2003). For example, Rehwagen et al. (2003) reported that the average outdoor concentrations of limonene and α- and β-pinenes were 0.57, 1.40, and 0.16 μg m^{-3}, respectively, whereas those in dwellings were 36.69, 24.77, and 3.01 μg m^{-3}, respectively. Hodgson and Beal (2002) also showed that outdoor concentrations for limonene and α- and β-pinenes were < 0.5, 1.4, and < 0.5 μg m^{-3}, respectively, whereas indoor concentrations were 40.3, 232, and 73.9 μg m^{-3}, respectively. Thus, assuming that the outdoor concentration of terpenes is negligible, eq. (2) can be derived as follows:

$$C_g = \frac{R_{emission}}{Vk_e} + C_{in} \quad (2)$$

where k_e is the overall elimination rate constant (h^{-1}), C_{in} is the indoor concentration of terpenes before using the essential oil diffuser (μg m^{-3}), $R_{emission}$ is the rate of emission of terpenes from the essential oil diffuser (μg h^{-1}), and V is the volume of a vehicle or a studio apartment (m^3). We assumed that air change was the dominant elimination process for terpenes in vehicles and studio apartments. The volumes of vehicles and studio apartments were obtained via a survey of the volunteers. In vehicles, the measured k_e values were 1.6, 2.4, and 2.8 h^{-1} while driving at 20 mph with recirculation (Ott et al., 2008). In studio apartments, the geometric mean of the measured k_e values was 0.77 h^{-1} with a geometric standard deviation of 1.435 (Johnson et al., 2004). For model essential oil diffusers in vehicle and studio apartments, $R_{emission}$ values were measured from triplicate tests of the mass loss of terpenes in the purchased essential oil diffusers. Three model essential oil diffusers for vehicle and studio apartments were deployed in a fume hood for 2 h immediately after the cap was removed, and the mass loss of terpenes in essential oil diffusers was measured. The model essential oil diffusers for the studio apartments were deployed by removing the cap and submerging three capillary sticks inside the diffusers. Model essential oil diffusers for the vehicle were deployed by removing the cap.

2.6. Quality assurance and quality control

To determine the method detection limit (MDL) of ePTFE PS, seven ePTFE PS were spiked with 12, 4.4, 3.4, and 11 μg m^{-3} of limonene, α- and β-pinene, and γ-terpinene, respectively, for 0.5 h. The MDL of the ePTFE PS for terpenes was calculated from the standard deviations of the spiked terpenes multiplied by 3.14 (Glaser et al., 1981). When sampling for 0.58 h, the average time for wearing ePTFE PS while driving, the MDL for concentrations of limonene, α- and β-pinenes, and γ-terpinene in air were 4.76, 1.93, 2.09, and 4.24 μg m^{-3}, respectively. When sampling for 8.9 h, the average time for wearing ePTFE PS while sleeping, corresponding values of limonene, α- and β-pinenes, and γ-terpinene were 0.31, 0.13, 0.14, and 0.27 μg m^{-3}, respectively. None of the selected terpenes was detected in the 21 blank ePTFE PS samples. The extracts were calibrated using the internal standards toluene-d_8 and chlorobenzene-d_5.

3. Results and discussion

3.1. Time-weighted average concentration of indoor terpenes

The mass accumulation of terpenes in the short (45 mm) and long (500 mm) ePTFE PS placed in the 125 L chamber at a constant terpene

concentration are shown in Fig. S2 and S3 (Supplementary Material). The accumulated mass of terpenes increased linearly with time. The slopes in Fig. S2 and S3 represent the mass accumulation rates (μg h^{-1}). The mass accumulation rates of the short and long ePTFE PS for limonene and α-pinene increased linearly with the concentration in air (Fig. 1). The coefficients of determination (R^2) were >0.98. The sampling rates of the ePTFE PS were determined from the slopes of the figures. As the length of the sampler and mass of the absorbent increased, the sampling rates of the sampler increased by approximately 10-fold. For limonene, sampling rates of the short and long ePTFE PS were 0.86 and 8.3 L h^{-1}, respectively. For α-pinene, the sampling rates of the short and long ePTFE PS were 0.83 and 11 L h^{-1}, respectively. As shown in Fig. S4 (Supplementary Material), sampling rates of long ePTFE PS for β-pinene and γ-terpinene were 9.7 and 7.9 L h^{-1}, respectively. The sampling rates of commercial passive samplers for VOCs, such as the SKC Ultra passive sampler, Radiello diffusive sampler, and cylinder sampler with Tenax, ranged from 1 to 1.8 L h^{-1} (Coutant et al., 1986; Król et al., 2012; Lewis and Mulik, 1985; Lewis et al., 1985; McAlary et al., 2015; Pennequin-Cardinal et al., 2005). An ePTFE PS with a higher sampling rate can be an efficient alternative to existing passive samplers.

From Eq. (1), the diffusion coefficients (m^2 s^{-1}) of limonene and α-pinene for both the short and long ePTFE PS were obtained. As shown in Table 1, the diffusion coefficients obtained at different concentrations were similar. For example, diffusion coefficients of long ePTFE PS for limonene and α-pinene ranged 2.9–5.9 and 5.0–7.8 (× 10^{-7} m^2 s^{-1}), respectively, in different air concentrations. Meanwhile, diffusion coefficients of short ePTFE PS for limonene and α-pinene ranged 4.5–5.9 and 4.9–5.8 (× 10^{-7} m^2 s^{-1}), respectively. As expected, the diffusion coefficients of limonene and α-pinene for both short and long ePTFE PS were almost the same because both ePTFE PS were made of the same material. The average effective diffusion coefficients of the long ePTFE PS for limonene and α-pinene were 4.3 and 6.3 (× 10^{-7} m^2 s^{-1}), respectively. Corresponding values of short ePTFE PS for limonene and α-pinene were 5.0 and 5.1 (× 10^{-7} m^2 s^{-1}), respectively. Those values are in the same order of magnitude for other VOCs (e.g., 8.5 × 10^{-7} and 8.2 × 10^{-7} m^2 s^{-1} for 1,3,5-trimethylbenzene and 1,3-dichlorobenzene) tested previously (Kim et al., 2021). As shown in Table S2, the effective diffusion coefficients of terpenes in the long ePTFE PS were at least one order of magnitude lower than those in air, suggesting that the resistance of mass transfer in the tube determines the overall mass transfer rate.

Table 1
Diffusion coefficients of long and short passive samplers for limonene and α-pinene.

	Long sampler		Short sampler	
	Concentration of air (μg L^{-1})	Diffusion coefficient × 10^{-7} (m^2 s^{-1})	Concentration of air (μg L^{-1})	Diffusion coefficient × 10^{-7} (m^2 s^{-1})
Limonene	10	5.9 (± 0.5)	2.5	4.5 (± 0.4)
	6.7	4.2 (± 0.6)	1.7	4.7 (± 0.8)
	3.4	2.9 (± 0.3)	0.84	5.9 (± 0.8)
α-pinene	10	7.8 (± 0.7)	2.6	4.9 (± 0.2)
	6.9	6.0 (± 0.7)	1.7	4.6 (± 0.8)
	3.4	5.0 (± 0.8)	0.86	5.8 (± 1.0)

Values in parentheses are standard deviations (n = 3).

Table S3 shows the concentrations of terpenes in a 5 m^3 chamber after 0, 24, 48, and 72 h, measured using active sampling. As shown in Fig. S5 (Supplementary Material), the time-dependent mass accumulations of the passive samplers were highly linear, with R^2 values of 0.999 for all terpenes. As shown in Fig. 2, the concentrations of limonene and α- and β-pinenes measured using both active and passive sampling in a 5 m^3 chamber were compared. For limonene, time-weighted average concentrations at 24, 48, and 72 h measured using passive sampling were 1.56, 1.50, and 1.96 times higher than the average concentrations measured using active sampling, respectively. For β-pinene, time-weighted average concentrations measured at 24, 48, and 72 h using passive sampling were 1.44, 1.93, and 1.20 times lower than the average concentrations measured by active sampling, respectively. For α-pinene, the differences between passive sampling and active sampling were very small, by factors of 1.12, 1.07, and 1.00 at 24, 48, and 72 h, respectively. Assuming that the

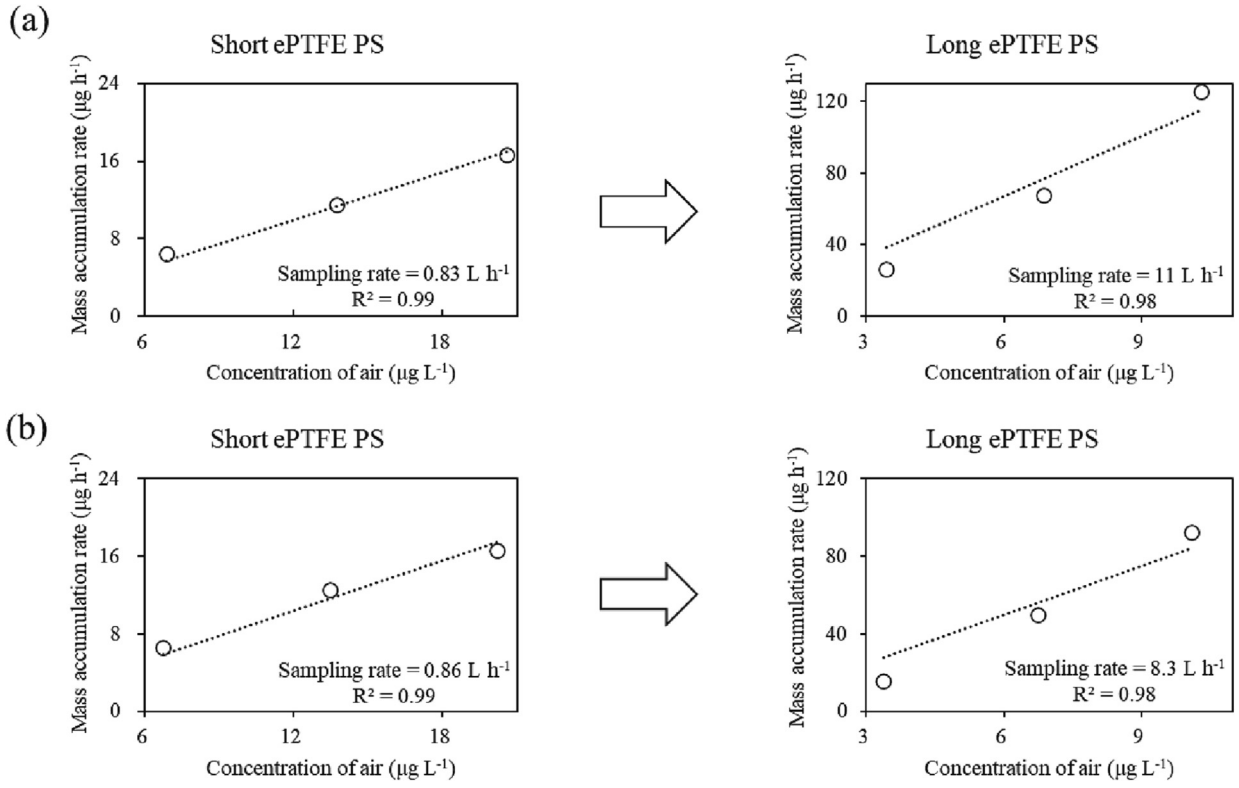


Fig. 1. Mass accumulation rates (μg h^{-1}) of (a) α-pinene and (b) limonene (b) in short and long ePTFE PS determined at varying atmospheric concentrations.

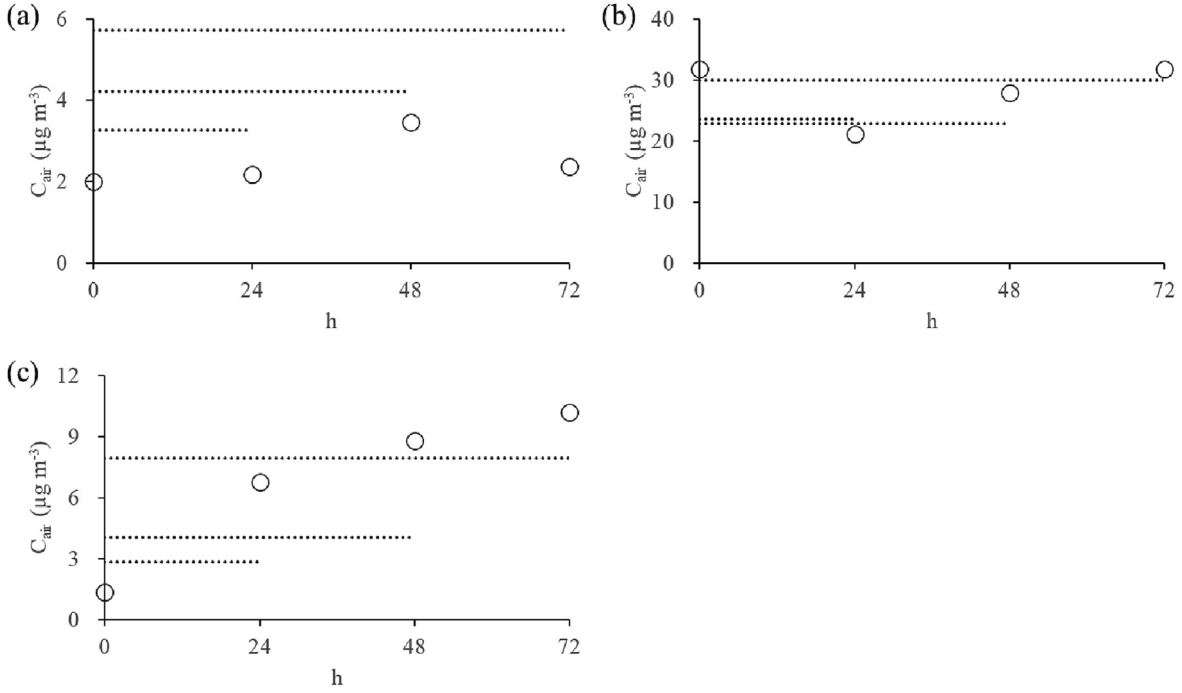


Fig. 2. Comparison of monitored concentrations of (a) limonene and (b) α- and (c) β-pinenes using active and passive sampling methods in the 5 m^3 test chamber. Dotted lines show the time-weighted average concentrations of limonene and α- and β-pinenes in air (μg m^{-3}) measured using passive sampling with exposure durations. Circles represent the concentrations of limonene and α- and β-pinenes in air (μg m^{-3}) measured using active sampling for 30 min.

concentration measured using active sampling is the 'true' concentration in the chamber, ePTFE PS shows an accuracy within a factor of 2, satisfying the general accuracy criteria of passive sampling (Bohlin et al., 2007; Gioia et al., 2007). These differences may be because passive sampling measures time-weighted average concentrations over the entire experimental duration, whereas active sampling measures a "snapshot" concentration for 30 min. In our previous study, a short ePTFE PS showed high accuracy compared with real-time analysis (Kim et al., 2021). Although the developed ePTFE PS was improved, we assumed that the long ePTFE PS also had a high accuracy. Overall, the results in the 5 m^3 test chamber indicate that the ePTFE PS can be applied for assessing the time-weighted exposure concentration of terpenes under real exposure conditions.

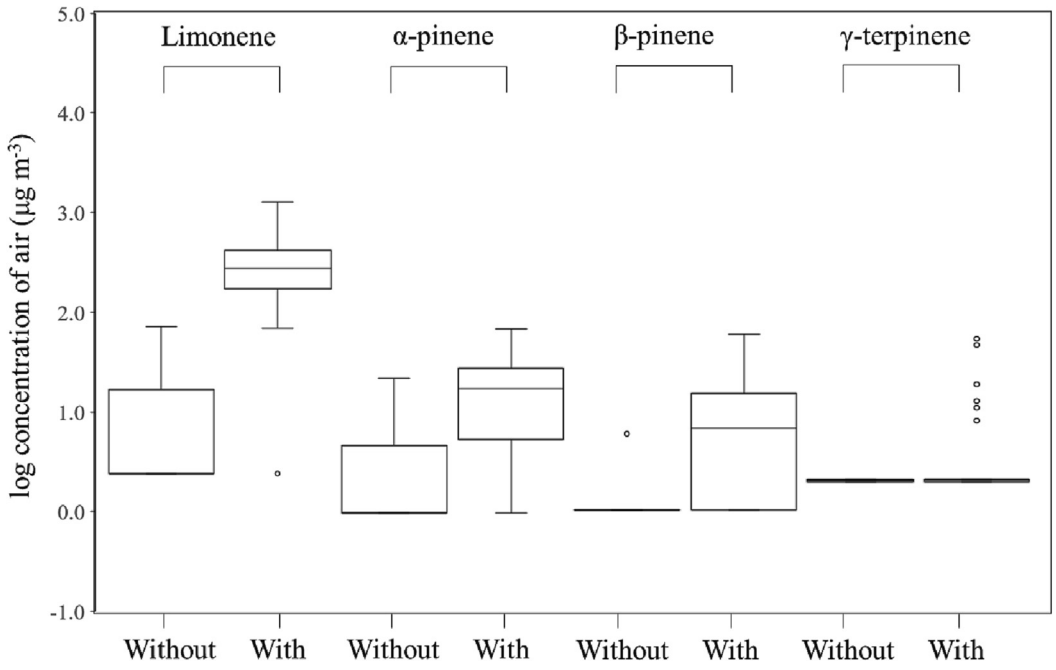


Fig. 3. Box-and-whisker plot showing the log concentrations (μg m^{-3}) of (a) limonene, (b) α-pinene, (c) β-pinene, and (d) γ-terpinene in vehicles with and without the use of the model essential oil diffusers. All experimental results from 25 participants at morning and afternoon were merged. The median is shown as the vertical line splitting the box in two means. The upper and lower boxes indicate the upper and lower quartiles. The whiskers are the two lines outside the box; the minimum value is at the start of the box, while the maximum is at end of the box. Points outside the box are potential outliers.

3.2. Consumer panel study

Fig. 3 shows the measured concentrations of terpenes in vehicles with and without the model essential oil diffuser. The measured values are listed in Table S4 and S5 (Supplementary Material). After using the model essential oil diffuser, the measured concentrations of all target terpenes in the vehicles increased on average. The effects of the model essential oil diffuser were statistically significant at $p < 0.05$ for limonene and α- and β-pinene in vehicles. In the morning, the *p*-values of limonene and α- and β-pinenes were 8.5×10^{-6}, 8.6×10^{-5}, and 3.5×10^{-3}, respectively. In the afternoon, the *p*-values of limonene and α- and β-pinenes were 2.9×10^{-8}, 1.3×10^{-5}, and 5.2×10^{-5}, respectively. Before using the model essential oil diffuser, the measured concentrations of all the target terpenes in the vehicles were extremely low. In previous studies, terpenes, such as limonene and α-pinene, were not found in vehicles where fragrance products were not used (Kim et al., 2016; Lv et al., 2020). After using the model essential oil diffuser, the measured concentration of limonene increased from 11 to 340 μg m^{-3} on average. Jo et al. (2008) showed a similar trend, in which concentrations of limonene increased from 5.4 to 19 μg m^{-3} and 5.9 to 28 μg m^{-3} in gasoline and diesel vehicles, respectively. For γ-terpinene, *p*-values of driving in the morning and afternoon were 0.26 and 0.058, respectively. The average concentrations of limonene in vehicles measured with and without the use of the essential oil diffuser were the highest, at least 17 times greater than those of other target terpenes. Because increased exposure to limonene was associated with an increase in both wheezing and asthma, and these effects were more significant in children (Dales and Cakmak, 2019), health effects with increased limonene concentration in vehicles need further investigation. In this study, the exposure concentration of limonene was elevated by approximately 3000 % after using the model essential oil diffuser in the vehicle. Dales and Cakmak (2019) used a reference exposure concentration of limonene at 55 μg m^{-3} (Wallace, 1991) in their epidemiological study, whereas the short-term exposure concentration in vehicles could be 620 % greater than this reference value. Therefore, the potential health effects on vulnerable populations, such as children, should be further evaluated, although the exposure duration is short. In addition, potential secondary products from the reaction of terpenes with oxidants, such as ozone and hydroxyl radicals (Carslaw, 2013; Huang et al., 2012b; Nørgaard et al., 2014; Sarwar and Corsi, 2007) may cause adverse health effects (Steinemann, 2016).

Fig. 4 shows the measured concentrations of terpenes in studio apartments with and without the use of an essential oil diffuser. All measured values are listed in Table S6. With the use of the model essential oil diffuser, the measured concentrations of all target terpenes in the studio apartments increased on average, although the data were scattered. The average concentrations of limonene, α- and β-pinenes, and γ-terpinene without using the product were 47, 3.6, 1.6, and 0.62 μg m^{-3}, respectively, whereas they increased by factors of 1.3, 1.2, 1.9, and 1.9, respectively, after the use of the essential oil diffuser. The measurements in this study showed a range similar to that of previous studies. Among the 18 indoor VOCs measured in a previous TEAM study, the average concentration of limonene was the highest at 55 μg m^{-3} (Wallace, 1991). Kostiainen (1995) showed that concentrations of limonene and α-pinene ranged 1.34–51.56 and 1.05–36.41 μg m^{-3}, respectively. Su et al. (2007) showed that concentrations of limonene were between 2.37 and 69.32 μg m^{-3} in offices and homes after using passive diffusers. Among the 25 studio apartments, the concentrations of limonene and α- and β-pinene in 14 studio apartments increased after using an essential oil diffuser. However, the concentrations of γ-terpinene in 14 studio apartments were below the detection limit. Although the essential oil diffuser affected the concentrations of terpenes in the studio apartments, the effects were not statistically significant for limonene, α-pinene, and γ-terpinene. For limonene, α-pinene, and γ-terpinene, *p*-values of the paired samples *t*-test were 0.35, 0.18, and 0.23 at the 5 % level of significance, respectively. During the experiments, only the distributed-model essential oil diffuser was deployed. However, before the panel study, we found that 15 volunteers had used other essential oil diffusers. After using the model essential oil diffuser, average concentrations of α-pinene, β-pinene, and γ-terpinene in their studio apartments increased by factors of 1.03, 1.83, and 1.79, respectively. The average concentrations of limonene in their studio apartments were similar, and the effects of the model essential oil diffuser were not statistically significant. The *p*-values of the paired sample *t*-test for limonene, α- and β-pinene, and γ-terpinene were 0.96, 0.87, 0.14, and 0.40 at the 5 % level of significance, respectively. The effects of the model essential oil diffuser were more significant for volunteers who had not used other essential oil diffusers prior to the test. After using the model essential oil diffuser, the average concentrations of limonene, α- and β-pinene, and γ-terpinene in the studio apartments increased by factors of 1.55, 1.54, 2.06, and 4.15, respectively. Although the effects of the model essential oil diffuser were not

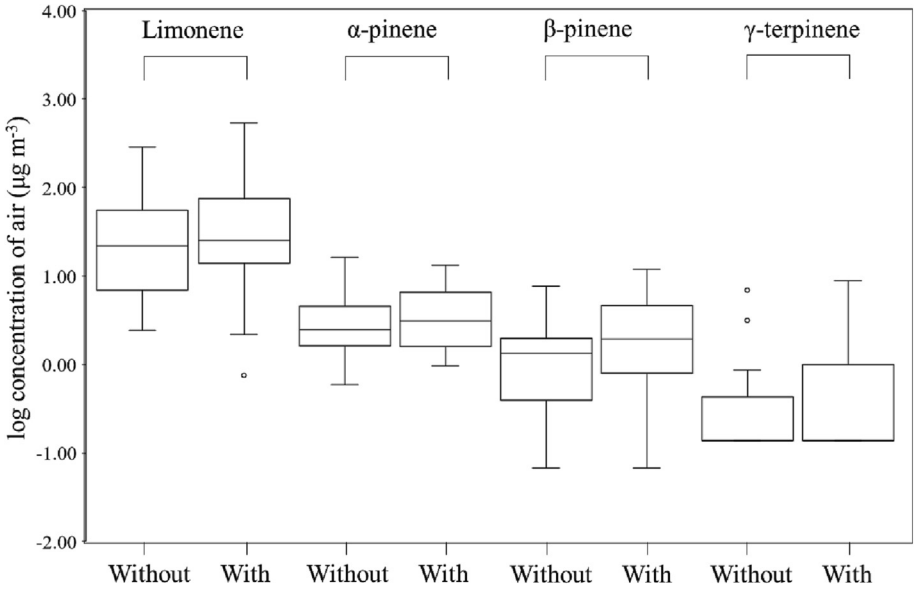


Fig. 4. Box-and-whisker plot showing the log concentrations (μg m^{-3}) of (a) limonene, (b) α-pinene, (c) β-pinene, and (d) γ-terpinene in studio apartments of 25 participants with and without the use of the model essential oil diffuser. The median is shown as the vertical line splitting the box in two means. The upper and lower boxes are the upper and lower quartiles. The whiskers are the two lines outside the box; the minimum value is at the start of the box, while the maximum value is at the end of the box. Points outside the box are potential outliers.

statistically significant for limonene, β-pinene, and γ-terpinene, whose *p*-values of paired samples *t*-test were 0.22, 0.20, and 0.37 at the 5 % level of significance, respectively, the effect was statistically significant for α-pinene, whose *p*-value was 0.040. In addition to fragrance products, personal care products, cleaning supplies, laundry products, sunscreens, and fragrance-free products contain terpenes (Nematollahi et al., 2019). Exposure to terpenes from other products could be a reason for the elevated levels of terpenes in studio apartments without using a model essential oil diffuser. For β-pinene, the *p*-value was 0.043 at the 5 % level of significance. The relatively lower frequency of β-pinene in other products compared to other terpenes could be a reason (Nematollahi et al., 2019). Among the target terpenes, limonene had the highest concentration. The average concentrations of limonene in studio apartments measured with and without an essential oil diffuser were at least 13 times higher than those of other target terpenes. The higher use frequency of limonene in various consumer products than that of other terpenes could be a reason (Nematollahi et al., 2019). Additionally, as shown in Table S1, the concentration of limonene in the model essential oil diffuser was at least seven times higher than that of other terpenes.

3.3. Comparison of measured values with an indoor air model

The concentrations of terpenes were roughly estimated because we only measured the volume of space, $R_{emission}$, and exposure time. For the studio apartments, the measured $R_{emission}$ values of limonene, α- and β-pinene, and γ-terpinene were 230, 35, 36, and 62 μg h^{-1}, respectively. The corresponding values for the vehicles were 2600, 220, 320, and 461 μg h^{-1}, respectively. The volumes of the studio apartments and vehicles are listed in Table S7 (Supplementary Material). With eq. (2), concentrations of terpenes in vehicles and studio apartments were estimated as shown in Table S8, S9, and S10 (Supplementary Material). The measured and estimated β-pinene concentrations in the vehicles were similar. Estimated concentrations of limonene and α-pinene were higher by factors of 3.6 and 1.9, respectively. Volunteers wore an ePTFE PS as a wristband and were located close to the diffuser while driving. When using consumer products, chemicals may not be evenly distributed (Jung et al., 2021). The uniform distribution assumed in a well-mixed compartment model may underestimate the exposure concentration if the near-field exposure zone is close to the source. However, the estimated concentration of γ-terpinene was higher than that measured by factor of 3.3 and 2.2 in vehicles and studio apartments, respectively. The faster photodegradation of γ-terpinene compared to limonene could explain this deviation, because degradation was not considered in the estimation (Bitterling et al., 2022). The measured and estimated concentrations of limonene and α- and β-pinene in studio apartments were similar. Considering the spatially nonhomogeneous distribution, decomposition, and sorption onto indoor surfaces, personalized passive sampling can overcome the limitations in the prediction of exposure using indoor air models, and the ePTFE PS can be used for higher-tier exposure estimation of VOCs, including terpenes.

4. Conclusions

The results of the panel study showed that an essential oil diffuser in a vehicle significantly increased the exposure concentration of selected terpenes, especially limonene. The average exposure concentration of limonene was 340 μg m^{-3}, approximately 3000 % higher than the background concentration. With increasing evidence of the adverse health effects of limonene, further investigation of repeated short-term exposure to limonene while riding on a vehicle with a fragrance product such as an essential oil diffuser is needed. In studio apartments, the exposure levels of the selected terpenes were similar to those in previous studies, and the effects of an essential oil diffuser were not as significant as those in vehicles where the volume was limited. The elongated ePTFE PS was also proven to be a cheap alternative for personal exposure monitoring of terpenes. Although the sampling rate of the ePTFE PS was greater than commercial passive samplers for VOCs, its detection limit may be higher than that of traditional passive samplers because it requires solvent extraction whereas those commercial samplers can be coupled with thermal desorption analysis. Further modification of ePTFE PS to being compatible with thermal desorption would not only lower its detection limit, but also enhance its greenness avoiding solvent extraction step. Furthermore, extensive studies using the ePTFE PS would provide a reliable exposure dataset for evaluating the health effects of VOCs at lower and fluctuating concentrations.

CRediT authorship contribution statement

Pil-Gon Kim: Conceptualization, Methodology, Investigation, Writing – original draft. **Arum Lee:** Methodology, Investigation. **Jaeho Shin:** Investigation. **Eugene Song:** Investigation. **Yerim Koo:** Investigation. **Dana Fahad M.S. Mohamed:** Investigation. **Soobin Choi:** Investigation. **Yongseok Hong:** Conceptualization, Writing – review & editing. **Seokho Song:** Investigation. **Incheol Noh:** Investigation. **Jung-Hwan Kwon:** Conceptualization, Supervision, Funding acquisition, Writing – review & editing.

Data availability

Data will be made available on request.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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