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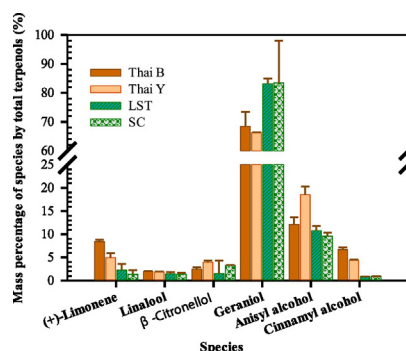
Emission characteristics of allergenic terpenols in PM_{2.5} released from incense burning and the effect of light on the emissions

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HIGHLIGHTS

- This study compared terpenols in two Thai and two Taiwanese brands of incense.
- The powder of the Thai brands contained more allergenic terpenols.
- The smoke from The Taiwanese brands contained far more terpenols.
- Direct contact with the powder of the Thai brands carries greater health risks.
- Inhalation of the smoke from the Taiwanese brands carries greater health risks.

GRAPHICAL ABSTRACT



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ABSTRACT

This study investigated allergenic terpenol compounds in incense powder and smoke. The powder of two Thai brands contained higher concentrations of terpenols up to 6.15 times higher than those of two Taiwanese brands. Consequently, Thai incense makers face a higher potential risk of contact dermatitis than Taiwanese incense makers do. *D*-Limonene was the primary terpenol compound in the powder of Thai B (64.0%) and Thai Y (31.5%), sold in Thailand. By contrast, anisyl alcohol was the primary terpenol compound in the powder of LST (40.3%) and SC (37.7%), sold in Taiwan. After the four brands of incense were ignited, their mean PM_{2.5} emission factor was $18.02 \pm 6.20 \text{ mg g}^{-1}$ incense. The PM_{2.5} mass emission factors of the Taiwanese brands were far higher than those of the Thai brands, and so were the PM_{2.5} terpenol emission factors, showing that the smokes of the Taiwanese incense were potentially more allergenic than those of the Thai incense. Geraniol, the most allergenic terpenol compound, was 2.8%–10.7% of total terpenol compounds in the powder of the four brands, yet was the main contributor to PM_{2.5}, constituting 66.3%–83.5% of terpenol compounds in the smokes of the four brands. Furthermore, geraniol exhibited an IP ratio, defined as the incense/powder (IP) ratio of terpenol-related species, >1 in all four brands, and >5 in the Taiwanese brands, suggesting a greater health risk with the smoke from the Taiwanese incense. The IP ratios of other terpenol species were all <1, indicating

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decomposition through combustion. Additionally, the light/darkroom ratios of the terpenol species were >1, meaning that the generation of PM_{2.5} terpenol compounds was potentially enhanced by indoor lighting.

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1. Introduction

Because most people spend 80–90% of their time indoors, indoor air pollution has been recognised as a notable public health concern. Incense smokes are a common indoor air pollutant in Asian regions such as Taiwan, Thailand, Hong Kong, Vietnam and China, where firecrackers, fireworks, and incense are used in religious rites, festivities, and other worship activities. However, people are often unaware of the health hazards of these smokes (Bari et al., 2011; Wang et al., 2012; Yang et al., 2013). Incense in particular, which is used both indoors and outdoors, elicits little worry owing to the fragrance it emits (Kuo et al., 2015). In fact, incense is always burned in a smoldering, incomplete, slow, and lengthy fashion and produces incense smoke that contains a wide variety of particulate and gaseous substances that are often measured in submicron or smaller sizes (Tsai et al., 2010; See and Balasubramanian, 2011; Kuo et al., 2016; Goel et al., 2017). The particle size distribution is influenced by combustion and environmental factors such as ventilation and detention time in the location where incense is ignited (Kleeman et al., 1999; Englert, 2004). These particles often contain hazardous substances (Cohen et al., 2013) and allergenic stimulants that can affect respiratory health when inhaled (Dalibalta et al., 2015; Kuo et al., 2015, 2016).

Past studies have found that worshipers who offer veneration in temples on the first or fifteenth day of a lunar month face a cancer risk 1000 times greater than non-templegoers because of their exposure to incense smokes rich in polycyclic aromatic hydrocarbons (Chuang et al., 2013). Additionally, cancer risk and duration of worshiper's temple visits were found to be strongly and positively correlated: Research has shown that spending 20 min in front of a censer doubles the risk compared with a 10-minute stay. Temple workers and people who use incense at home every day have also been found to face cancer risks exceeding the 10^{−6} threshold (Kao and Lung, 2000). Moreover, pollutants in incense smokes are known to cause inflammation of the lung cells (Dominici et al., 2006; Chuang et al., 2011). Research has confirmed that incense smokes contain fragrant allergenic substances (Rastogi et al., 1996; Kuo et al., 2015) that increase the risk of contact dermatitis in the event of exposure to high concentrations for a prolonged duration (Marzulli and Maibach, 1980; Thompson et al., 1983; Kim et al., 2015). Comparing the smokes of Taiwanese and Thai incense brands, Kuo et al. (2016) found that carboxylates, primarily acetate, constituted 2.10%–4.93% of the PM_{2.5} mass, and that the Thai brands created higher concentrations of primary PM_{2.5} acetate than the Taiwanese brands, hence posing a greater threat to the respiratory and dermal systems.

Semivolatile organic substances, including alcohol, aldehyde, ketone, terpene, ester, and amine, are classified as indoor air pollutants because they can cause the adverse health effects in nonindustrial indoor settings (Bernstein et al., 2008). According to the U.S. National Institute for Occupational Safety and Health, the presence of volatile organic compounds can induce work-related asthma (WRA), one of the most common respiratory diseases (Weisel, 2002; Dales and Raizenne, 2004; Weschler, 2006). Of these compounds, the terpenols α -limonene, linalool, anisyl alcohol, geraniol, β -citronellol and cinnamyl alcohol have been recognised as marker allergens (Kim et al., 2015). Freund's complete adjuvant tests (FCAT) have proved that α -limonene is a powerful sensitiser because prolonged exposure of α -limonene to the air causes it to oxidise and form (R)-(–)-carvone and a mixture of *cis*- and *trans*- isomers, all of which are allergenic (Karlberg et al., 1992).

Skin patch tests have further revealed that oxidised α -limonene generates strongly allergenic oxygenated monocyclic terpene, which can increase the risk of contact dermatitis through prolonged exposure (Karlberg and Goossens, 1997; Matura et al., 2002). In addition, contact between α -limonene and ozone can create compounds that stimulate the lungs and cause bronchoconstriction (Clausen et al., 2001; Weschler, 2006). Animal tests have indicated that the metabolites of geraniol and linalool can affect the activity of microsomal enzymes in the lungs and liver (Chadha and Madyastha, 1984; Lapczynski et al., 2008a, 2008b). Because of the risks described, terpenol compounds such as α -limonene, linalool, anisyl alcohol, geraniol, β -citronellol and cinnamyl alcohol have been included by the European Union in a list of 26 allergenic substances in cosmetics (Sánchez et al., 2010). The reactions between terpenol compounds and strong oxidisers such as ozone, NO_x, hydroxides, and SO_x tend to form submicronic secondary organic aerosols (SOAs) that oxidise in the human body and induce inflammation and ischaemic heart disease; such SOAs, whose relatively large surface area enables them to carry large quantities of toxicants through catalysis, can also easily find their way deep into the lungs through inhalation (Donaldson et al., 2002; Kreyling et al., 2004) and cause problems such as asthma and chronic obstructive pulmonary disease (COPD) (Pope and Kanner, 1993).

Incense smokes are among the primary sources of indoor airborne particles in Taiwan and Thailand, where the use of incense associated with religious rites in temples and households is fairly common (Weschler, 2006; Waring and Siegel, 2013). These smokes have been identified as a notable public health concern because prolonged exposure to them has been proven to cause chronic contact dermatitis, migraines, and chemical allergies of the respiratory system (Navasumrit et al., 2008). However, various questions remain unanswered, such as whether these smokes contain terpenol compounds that stimulate the skin, eyes, and nose (Karlberg and Goossens, 1997; Matura et al., 2002), whether the terpenol particles vary with incense type and brand, and whether the terpenol compounds are contained in the incense powder or the products of combustion. Furthermore, whether smoke composition changes with indoor lighting conditions is also unclear. Therefore, the present study explored the changes in photochemical properties of the allergenic PM_{2.5} terpenol compounds in incense smokes by simulating indoor conditions with/without lighting.

2. Experimental

2.1. Selection of incense sticks and pre-treatment of incense samples

Four brands of traditional incense, Shang Chen incense (SC) and Lao Shang Tou incense (LST), sold in Taiwan, and Thai Black incense (Thai B) and Thai Yellow incense (Thai Y), sold in Thailand, were chosen to investigate the emission properties of allergenic terpenol compounds and changes in the compounds' photochemical properties in light/dark indoor conditions.

Table 1 lists the physical properties of the incense sticks. To minimise their physical and chemical differences, 18 sticks were carefully picked from each brand, all similar in total length, holder length, incense diameter, holder diameter, and weight. Nonetheless, differences in length and weight did exist between the brands. The average lengths of the Taiwanese and Thai brands were 39.0 ± 0.11 cm and 33.0 ± 0.06 cm, respectively; the Taiwanese brands were markedly longer than the Thai ones both in terms of total length and combustible length.

Table 1

Physical characteristics of four types of incense sticks.

Incense	Length (cm)		Diameter (mm)	
	Whole stick	Combustible part	Combustible part	Holding stick
Thailand incense				
Thai B	33.6 ± 0.03	20.3 ± 0.10	2.65 ± 0.48	1.34 ± 0.05
Thai Y	32.4 ± 0.08	22.5 ± 0.12	2.62 ± 0.47	1.00 ± 0.15
Taiwan incense				
LST	39.0 ± 0.13	28.0 ± 0.11	1.70 ± 0.21	1.00 ± 0.15
SC	39.0 ± 0.09	28.0 ± 0.09	1.05 ± 0.05	1.00 ± 0.15

N = 18 for each type of incense.

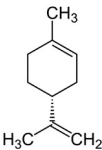
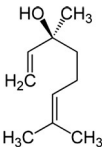
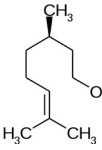
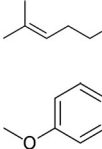
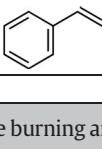
By contrast, the Thai brands were thicker in diameter (2.64 ± 0.48 cm) than the Taiwanese ones (1.38 ± 0.13 cm). The holders of the Taiwanese brands were made of Makino bamboo (*Phyllostachys makinoi* Hayata), while the holders of the Thai brands were made of an unspecified type of bamboo. The incense sticks were all stored in a desiccator at 25 ± 1 °C and $45 \pm 5\%$ humidity for 24 h before being weighed (with the results termed the starting weight). The difference between an incense sticks starting weight and its holders' weight after combustion denoted the combusted weight. The 37-mm quartz fibre filters (Pall Tissuquartz™ filters, 2500 QAT-UP, 37 mm) used were prepared before use by baking at 900 °C for 4 h to eliminate any possible interference of carbonaceous aerosols, after which they were stored in a desiccator at 25 ± 1 °C and $45 \pm 5\%$ humidity for 24 h. The filters were subsequently weighed (at least three times) using a microbalance with sensitivity of 10 µg (Mettler Toledo AT261) under a relative humidity of $45 \pm 5\%$ to determine their precise weights. The weighing process was conducted in accordance with the Shewart rules to ensure its reliability (Kuo et al., 2015).

2.2. Verification of the feasibility of the method for extraction of terpenol compounds from incense powder and quartz fibre filters

All reagents were of analytical grade, obtained from Merck (Darmstadt, Germany), and were used without further purification unless otherwise indicated. All solutions were prepared using deionized water (Resistivity > 18.2 MΩ·cm at 25 °C, Direct16, Millipore). Precisely 0.5 g of incense powder was scraped from the sample incense sticks of each brand and placed separately in glass bottles (4 mL) that were subsequently spiked with 10 µL of a standard mixture composed of D-limonene (278 ppm), linalool (290 ppm), β-citronellol (286 ppm), geraniol (293 ppm), anisyl alcohol (371 ppm), and cinnamyl alcohol (348 ppm). Table 2 shows the structures of these terpenol compounds. To extract terpenol compounds in the powder samples, 3 mL of an extraction solution (99.9% methanol and 0.1% ethyl acetate) was added to the bottle, which was then immediately wrapped in foil to prevent photochemical reactions. The solution then underwent 1 h of ultrasonication (Branson 5210) before being passed through a Pall hydrophilic nylon membrane filter of 25 mm in diameter with a pore size of 0.2 µm to extract the solution, which was subsequently injected into a gas chromatography-flame ionisation detector (GC-FID, Agilent 6890 GC) to analyse the composition of its terpenol compounds in accordance with the procedures outlined by Kuo et al. (2015). The average extraction ratios of the aromatic compounds were $107.1 \pm 1.18\%$ (D-limonene), $101.2 \pm 0.75\%$ (linalool), $94.84 \pm 1.86\%$ (β-citronellol), $88.38 \pm 1.08\%$ (geraniol), $91.59 \pm 3.45\%$ (anisyl alcohol), and $95.87 \pm 0.46\%$ (cinnamyl alcohol).

For the simulated extraction of PM_{2.5} terpenol compounds, 10 µL of the standard mixture composed of D-limonene (278 ppm), linalool (290 ppm), β-citronellol (286 ppm), geraniol (293 ppm), anisyl alcohol (371 ppm), and cinnamyl alcohol (348 ppm) was added to a 47-mm

Table 2Chemical information on terpenols in PM_{2.5} emitted from incense burning.

Terpenols	Chemical structure formula
D-Limonene, C ₁₀ H ₁₆ (1-Methyl-4-(1-methylethenyl)-cyclohexene)	
Linalool, C ₁₀ H ₁₈ O ((±)-3,7-Dimethyl-3-hydroxy-1,6-octadiene)	
β-Citronellol, C ₁₀ H ₂₀ O (3,7-Dimethyloct-6-en-1-ol)	
Geraniol, C ₁₀ H ₁₈ O ((trans)-3,7-Dimethyl-2,6-octadien-1-ol)	
Anisyl alcohol, C ₈ H ₁₀ O ₂ ((4-Methoxyphenyl)methanol)	
Cinnamyl alcohol, C ₉ H ₁₀ O ((E)-3-phenylprop-2-en-1-ol)	

quartz fibre filter, and extraction then proceeded as for the powder samples: The filter was rolled up and placed in a 4 mL bottle, 3 mL of an extraction solution (99.9% methanol and 0.1% ethyl acetate) was added, followed by ultrasonication (Branson 5210) for 1 h. Next, the solution was filtered through a Pall hydrophilic nylon membrane filter of 25 mm in diameter with a pore size of 0.2 μm and analysed using GC-FID. The average extraction ratios of the aromatic compounds in the smoke particles were $98.06 \pm 0.50\%$ (α -limonene), $90.77 \pm 0.26\%$ (linalool), $86.47 \pm 5.57\%$ (β -citronellol), $85.56 \pm 0.86\%$ (geraniol), $90.96 \pm 0.14\%$ (anisyl alcohol), and $88.40 \pm 0.98\%$ (cinnamyl alcohol).

The simulation tests showed that the extraction ratios of the compounds in the incense powder and smoke particles were 85%–108%, and the extraction ratios of geraniol were lower than all others in both cases. The powder sample extraction ratios were all higher than those of the smoke samples; although the filter extraction ratios were $<100\%$, they were all $>85\%$, indicating that the results were still satisfactory as per the quality control standards of the analysis. The method detection limits (MDLs) of the compounds were found to be 0.422 ppm (α -limonene), 0.435 ppm (linalool), 0.429 ppm (β -citronellol), 0.557 ppm (anisyl alcohol), and 0.522 ppm (cinnamyl alcohol), indicating that the extraction method and detection efficiency employed in the present study satisfied its analytic requirements.

2.3. Collection and analysis of $\text{PM}_{2.5}$ smoke particles

The prepared and weighed quartz fibre filters (Pall Tissuquartz™ filters, 2500 QAT-UP, 37 mm) were placed in Personal Environmental Monitors ($\text{PEM}_{2.5}$, MSP Model 200 $\text{PEM}-10-2.5$) to collect $\text{PM}_{2.5}$ from a smoke stream controlled at 10.0 ± 0.1 Lpm by a rotameter in a control chamber (Fig. 1). The apparatus features a 1.0 m (L) $\times$ 0.5 m (W) $\times$ 0.65 m (H) operation chamber of a volume of 0.325 m^3 , made of transparent polymethylmethacrylate (PMMA) with anti-static coating to reduce wall deposition. Four carefully angled small fans were

installed in the control chamber to ensure even distribution of the smoke without blowing directly at its source (the incense sticks). The function of the control chamber was to prevent the invasion of external aerosols and ensure that all of the particles collected came from the incense smokes. The pure air needed to balance the inside and outside air pressures was fed into the chamber through a gas flow meter at 10.0 ± 0.2 Lpm. This air was supplied by a compressor, which sent fresh air through a silica gel desiccant to remove moisture, through activated carbon to remove airborne organics, and then through a high-efficiency particulate-arresting filter (HEPA) to filter it again before directing it to the gas flow meter. The actual amount of air exchanged in a well-mixed ventilation scenario by four fans was kept at 185% during the 1-hour sampling period. The air exchange rates of 1.85 h^{-1} are enough to collect the PM emitted from incense burning completely. The temperature and relative humidity in the chamber are monitored at the condition of $25 \pm 2^\circ\text{C}$ and $45 \pm 5\%$ relative humidity. Six full-spectrum fluorescent bulbs (6500K-T5 fluorescent, 14 W, wavelength range between 380 and 780 nm, Philips) were installed in the control chamber to allow the investigation of differences caused by lighting (Kuo et al., 2015, 2016). Smoke particles were collected in three lighting settings: *collection upon burning*, *dark*, and *light*. *Collection upon burning* refers to the condition in which the incense stick was ignited under the full illumination of the six bulbs, followed by the $\text{PEM}_{2.5}$ being immediately turned on to collect $\text{PM}_{2.5}$ until 60 min after the incense stick burned out. The *dark* condition involved blocking out all light sources before the incense stick was ignited and leaving the joss stick to burn out in the dark before the $\text{PEM}_{2.5}$ was turned on to collect $\text{PM}_{2.5}$ for 60 min. The *light* condition was the same as the *dark* setting, except that the incense stick burned in the light of the six bulbs (Kuo et al., 2015, 2016).

The quartz fibre filters were then placed, together with the $\text{PM}_{2.5}$ collected on them, in a desiccator at $25 \pm 1^\circ\text{C}$ and $45 \pm 5\%$ humidity for 24 h, after which they were weighed using a microbalance with sensitivity of $10 \mu\text{g}$ at $25 \pm 1^\circ\text{C}$ and $45 \pm 5\%$ humidity. Thereafter, each

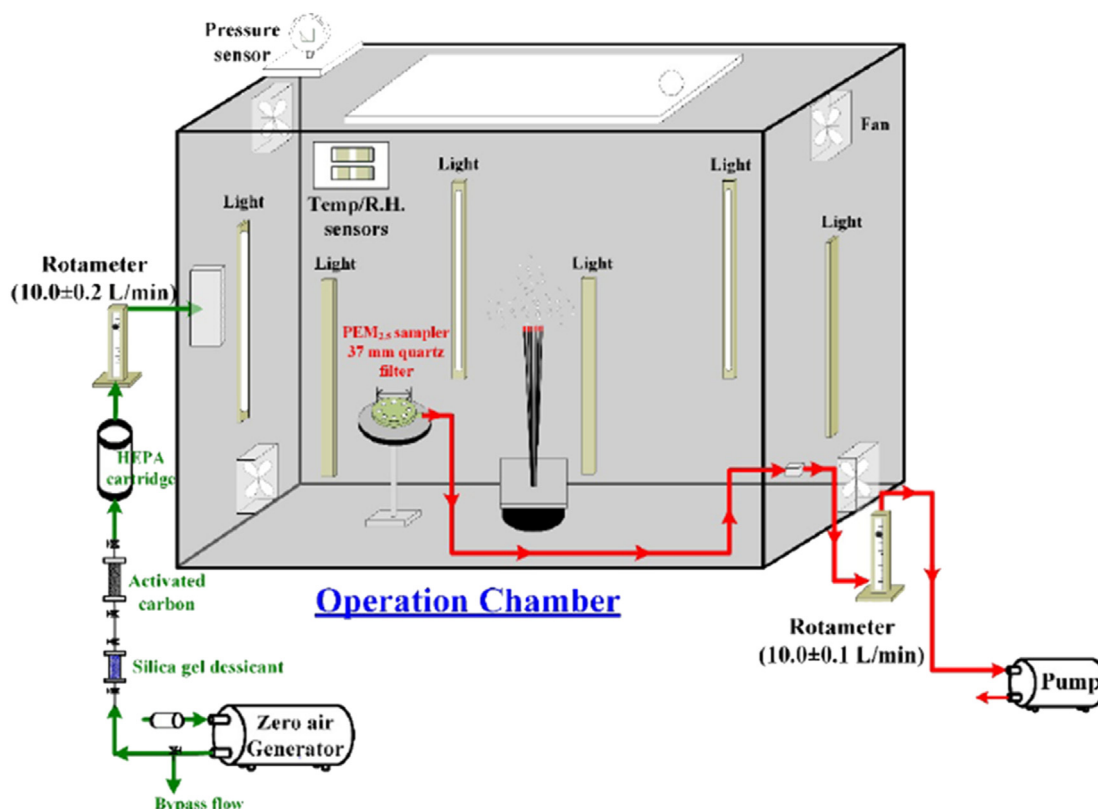


Fig. 1. A schematic diagram of the sampling apparatus and configuration.

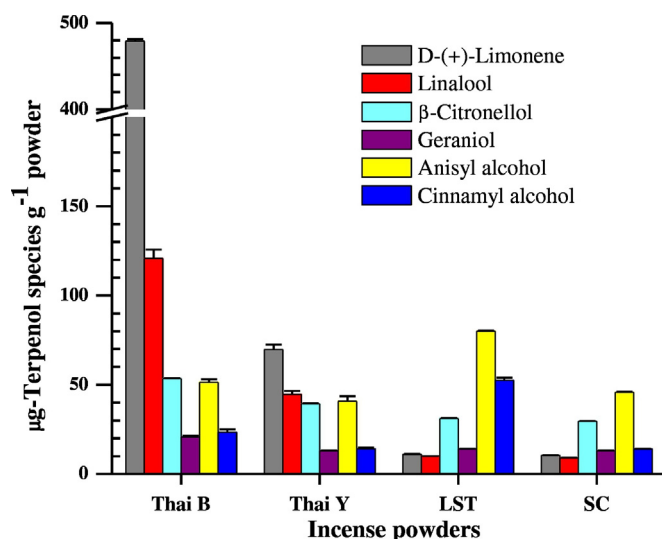


Fig. 2. Fragrant terpenol-related compounds in incense powders.

quartz fibre filter was cut in half. One half was placed in a glass bottle with 3 mL of an extraction solution (99.9% methanol and 0.1% ethyl acetate) for a 1-hour ultrasonication, and the extract then passed through a Pall hydrophilic nylon membrane filter of 25 mm in diameter with a pore size of 0.2 μm. The resulting extraction solution was subsequently injected into a GC-FID (Agilent 6890 GC) for analysis of the composition of its terpenol compounds.

3. Results and discussion

3.1. Terpenol compounds in incense powder

The concentrations of allergenic terpenol compounds found in the scraped incense powder were between 121.75 ± 0.06 and $748.72 \pm 1.66 \mu\text{g g}^{-1}$ powder (Fig. 2). The highest content was found in Thai B ($748.72 \pm 1.66 \mu\text{g g}^{-1}$ -powder), followed by Thai Y ($221.11 \pm 1.29 \mu\text{g g}^{-1}$ -powder), LST ($198.43 \pm 0.56 \mu\text{g g}^{-1}$ -powder), and SC ($121.75 \pm 0.06 \mu\text{g g}^{-1}$ -powder). The Thai brands contained more terpenol compounds than the Taiwanese brands did, implying that the Thai brands, particularly Thai B, potentially carry greater allergenic risks of contact dermatitis for incense makers.

In terms of individual compounds, the concentrations of D-limonene varied considerably among the samples, averaging 10.35 ± 0.02 – $479.16 \pm 2.36 \mu\text{g g}^{-1}$ powder (Thai B $\gg$ Thai Y $>$ LST $\approx$ SC). Because the Thai brands contained far more D-limonene than the Taiwanese brands ($p < 0.01$), the Thai brands are potentially more allergenic for incense makers (Karlberg et al., 1992; Karlberg and Goossens, 1997; Matura et al., 2002). The concentrations of linalool also varied

considerably among the samples, averaging 8.97 ± 0.08 – $120.73 \pm 4.95 \mu\text{g g}^{-1}$ powder, the Thai brands containing notably more linalool than the Taiwanese brands (Thai B $\gg$ Thai Y $>$ LST $>$ SC). The concentrations of β-citronellol averaged 29.54 ± 0.09 – $53.42 \pm 0.15 \mu\text{g g}^{-1}$ powder (Thai B $>$ Thai Y $>$ LST $>$ SC), while the concentrations of geraniol varied slightly among the brands, averaging 12.92 ± 0.22 – $20.73 \pm 0.82 \mu\text{g g}^{-1}$ powder (Thai B $>$ LST $>$ SC $\approx$ Thai Y). The concentrations of anisyl alcohol averaged 40.65 ± 2.90 – $79.97 \pm 0.33 \mu\text{g g}^{-1}$ powder (LST $>$ Thai B $>$ SC $>$ Thai Y) and those of cinnamyl alcohol averaged 14.00 ± 0.02 – $52.36 \pm 1.52 \mu\text{g g}^{-1}$ powder (LST $>$ Thai B $>$ Thai Y $\approx$ SC). Thai B contained high amounts of all terpenols and the highest amounts of four terpenol compounds while LST contained the most anisyl alcohol and cinnamyl alcohol. SC contained slightly more geraniol and anisyl alcohol than Thai Y, but its concentrations of the other four terpenol compounds were the lowest among the four brands. For incense makers, the results indicate that Thai B has higher allergenic potential than the other three brands.

3.2. Emission factors of terpenol compounds in smoke PM_{2.5} particles

Table 3 lists the smoke PM_{2.5} mass and emission factors of terpenol compounds obtained by collecting the PM_{2.5} aerosols. According to these results, the PM_{2.5} emission factors of the four incense brands ranged from 11.08 ± 0.56 to $24.52 \pm 1.95 \text{ mg g}^{-1}$ incense (averaging $18.02 \pm 6.02 \text{ mg g}^{-1}$ -incense), with SC $>$ LST $\gg$ Thai B $>$ Thai Y, confirming that the PM_{2.5} emission factors of the Taiwanese brands were far higher than those of the Thai brands. The average values of the PM_{2.5} emission factors of the terpenol compounds ranged from 35.98 ± 0.32 to $89.09 \pm 5.16 \mu\text{g g}^{-1}$ incense, with LST $>$ SC $>$ Thai Y $>$ Thai B. This ranking contradicted that of the powder samples, in which the Thai brands contained higher concentrations of terpenol compounds. Because of the markedly higher concentrations of PM_{2.5} terpenol compounds in the smokes of the Taiwanese incense ($p < 0.01$), the Taiwanese incense is more fragrant but also potentially poses a higher allergenic risk in an indoor setting.

Regarding individual terpenol compounds, the PM_{2.5} emission factors of D-limonene ranged from 1.21 ± 1.06 to $3.59 \pm 0.17 \mu\text{g g}^{-1}$ -incense, with Thai B $>$ LST $>$ Thai Y $>$ SC. The PM_{2.5} emission factors of linalool ranged from 0.67 ± 0.04 to $1.31 \pm 0.24 \mu\text{g g}^{-1}$ incense, with LST $>$ SC $>$ Thai B $>$ Thai Y. The PM_{2.5} emission factors of β-citronellol ranged from 1.04 ± 0.16 to $2.87 \pm 0.57 \mu\text{g g}^{-1}$ -incense (SC $\gg$ Thai Y $\approx$ LST $>$ Thai B) and those of geraniol were 23.87 ± 0.04 – $74.13 \pm 1.30 \mu\text{g g}^{-1}$ incense (LST $>$ SC $\gg$ Thai B $>$ Thai Y). The markedly higher concentration of geraniol (which primarily contributes the rosy aromas of the incense) emitted from Taiwanese incense gives it a stronger fragrance than Thai incense, but exposure to this toxic compound can induce respiratory allergies. The Taiwanese brands emitted more anisyl alcohol than the Thai brands (PM_{2.5} emission factors of anisyl alcohol ranged from 5.19 ± 0.66 to $9.61 \pm 0.69 \mu\text{g g}^{-1}$ incense, with LST $>$ SC $>$ Thai Y $>$ Thai B), while the Thai brands emitted more cinnamyl alcohol (PM_{2.5} emission factors of cinnamyl alcohol ranged

Table 3
Emission factors of fragrant terpenol-related compounds in smoke PM_{2.5}.

Species	Emission factor ($\mu\text{g g}^{-1}$ of incense)			
	Thai B	Thai Y	LST	SC
PM _{2.5} mass (mg g^{-1} of incense)	13.93 ± 0.06	11.08 ± 0.56	22.55 ± 3.09	24.52 ± 1.95
D-Limonene	3.59 ± 0.17	1.78 ± 0.72	2.00 ± 0.95	1.17 ± 1.06
Linalool	0.84 ± 0.04	0.67 ± 0.04	1.31 ± 0.24	1.22 ± 0.06
β-Citronellol	1.04 ± 0.16	1.42 ± 0.13	1.32 ± 0.08	2.87 ± 0.57
Geraniol	29.29 ± 2.19	23.87 ± 0.04	74.13 ± 1.30	72.97 ± 2.26
Anisyl alcohol	5.19 ± 0.66	6.67 ± 0.62	9.61 ± 0.69	8.35 ± 2.18
Cinnamyl alcohol	2.86 ± 0.20	1.57 ± 0.04	0.73 ± 0.01	0.80 ± 0.01
Σ PM _{2.5} terpenols	42.81 ± 3.42	35.98 ± 1.59	89.09 ± 11.04	87.38 ± 6.13

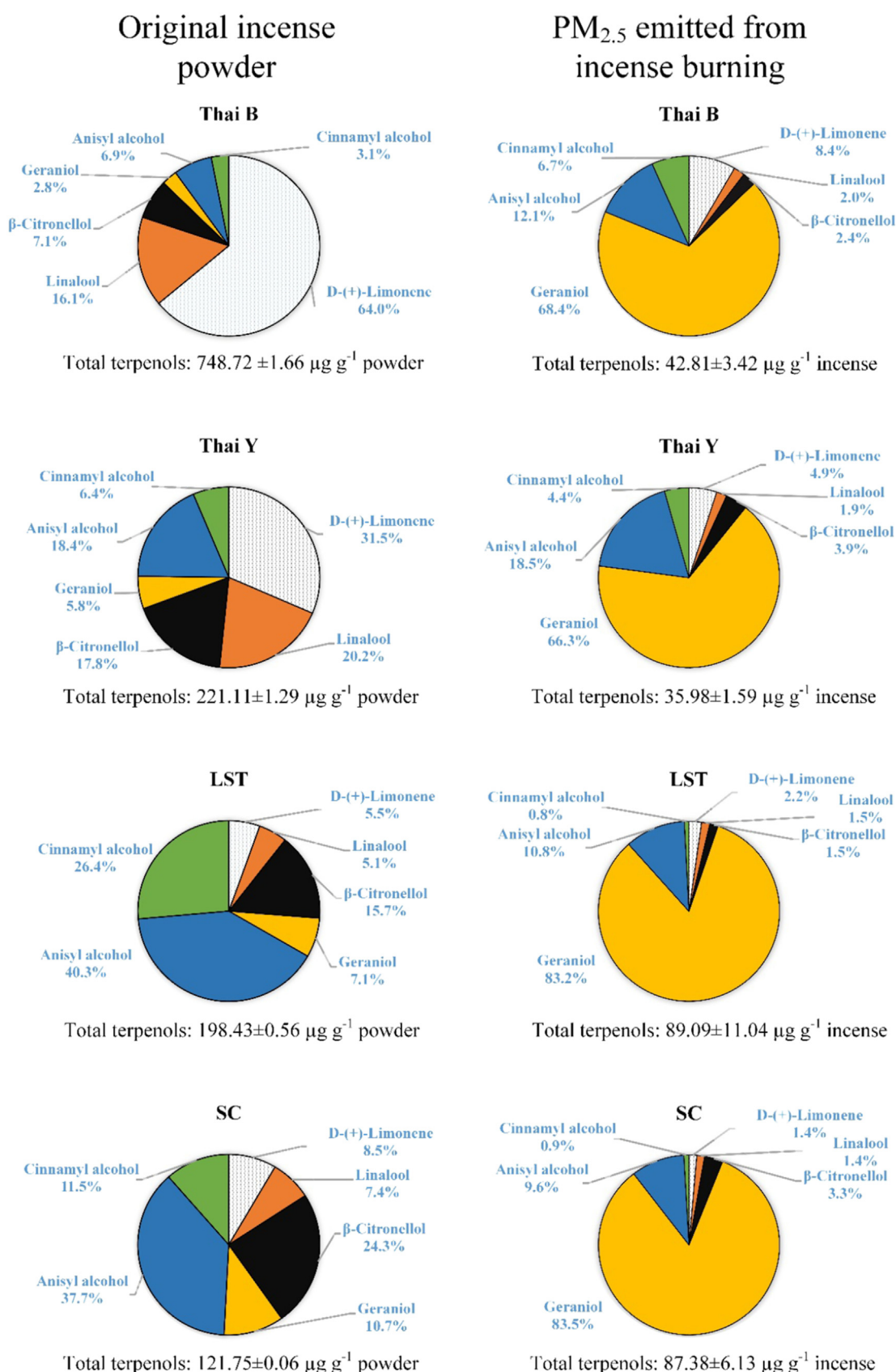


Fig. 3. Contribution of individual species to total amount of terpenols in incense powders and in PM_{2.5} emitted from incense burning.

from 0.73 ± 0.01 to $2.86 \pm 0.20 \mu\text{g g}^{-1}$ incense, with Thai B $\gg$ Thai Y $>$ SC $\approx$ LST).

3.3. Disparities in the proportions of terpenol compounds in incense powder and smoke PM_{2.5} particles

Fig. 3 lists the proportions of individual terpenol compounds to total terpenol compounds in both incense powder and incense smokes by brand. The terpenol compound found in the highest proportions in the powder of the Thai brands was *D*-limonene (as high as 64.0% in Thai B), followed by linalool. The proportion of anisyl alcohol in Thai Y was slightly higher than that of β -citronellol, whereas in Thai B this relationship was reversed. The proportions of cinnamyl alcohol and geraniol were lowest in the Thai brands, particularly geraniol which constituted only 2.8% and 5.8% of the total terpenol compounds in Thai B and Thai Y, respectively. By contrast, in the powder of the Taiwanese brands, anisyl alcohol, cinnamyl alcohol, and β -citronellol were found to be the most abundant terpenol compounds, particularly anisyl alcohol, which made up 40.3% and 37.7% of the total terpenol compounds in LST and SC, respectively. In comparison to the abundance of *D*-limonene and linalool in the Thai brands, *D*-limonene constituted only 5.5% (LST) and 8.5% (SC) of total terpenol compounds in the Taiwanese brands. The highly allergenic geraniol only accounted for 7.0% (LST) and 10.7% (SC), respectively of the Taiwanese brands, although this was slightly higher than in the Thai brands. These results illustrate that there are distinct regional differences in the terpenol composition of incense powder.

Examining the terpenol compounds in the PM_{2.5} of the incense smokes revealed that the highly allergenic geraniol was the most abundant species in both Thai brands (68.4% in Thai B and 66.3% in Thai Y), while geraniol was even more abundant in the PM_{2.5} of the Taiwanese brands (83.2% in LST and 83.5% in SC). All four brands were therefore determined to emit large quantities of hazardous PM_{2.5} after being ignited, and the allergenic terpenol compounds in their smokes pose a considerable threat to human health. Among these terpenol compounds, the exceptionally abundant geraniol was identified as the greatest health hazard, particularly the emissions from the two Taiwanese brands. Of the remaining terpenols, anisyl alcohol was the second most abundant terpenol compound in PM_{2.5} after geraniol, and its proportions in the PM_{2.5} of the Thai brands were slightly higher than its proportions in their powder. In the Taiwanese brands, by contrast, the proportions of anisyl alcohol in the PM_{2.5} were notably lower than that in the powder. Likewise, the proportions of cinnamyl alcohol in the PM_{2.5} of the Taiwanese brands were also markedly lower than in the powder. *D*-limonene, which exhibited the highest proportions in the powder of the Thai brands, only made up 4.9% (Thai Y) to 8.4% (Thai B) of the PM_{2.5} after the incense was burned. In the PM_{2.5} of the Taiwanese brands, the proportions of *D*-limonene were even lower, only making up 1.4% and 2.2% of the total terpenol compounds in SC and LST, respectively. The PM_{2.5} proportions of linalool and β -citronellol were also found to be lower than their proportions in incense powder.

Table 4

Ratios of various terpenol-related compound emission factors in smoke PM_{2.5} to the content per gram of incense powder.

Species	Incense/powder (IP) ratio of terpenol-related species ($\mu\text{g g}^{-1}$ of incense/ $\mu\text{g g}^{-1}$ of powder)			
	Thai B	Thai Y	LST	SC
<i>D</i> -Limonene	0.007	0.026	0.182	0.117
Linalool	0.004	0.015	0.130	0.135
β -Citronellol	0.019	0.036	0.042	0.097
Geraniol	1.413	1.848	5.291	5.599
Anisyl alcohol	0.101	0.164	0.120	0.182
Cinnamyl alcohol	0.123	0.111	0.014	0.057
Σ PM _{2.5} terpenols	0.057	0.163	0.449	0.718

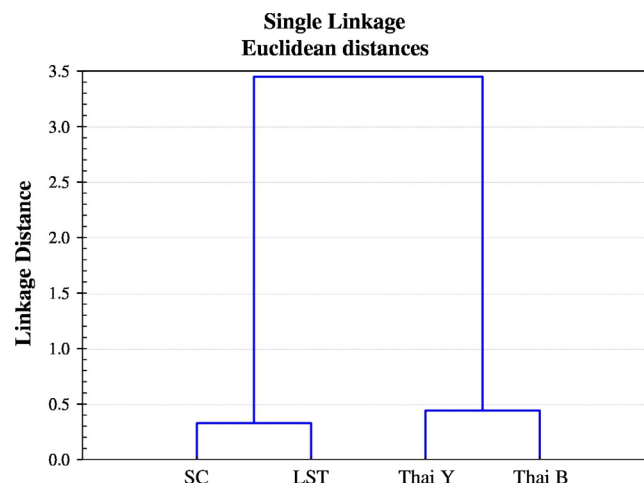


Fig. 4. Tree clustering of IP ratios with Euclidean distances for the four incense types.

The present study defines the ratio of the emission factor of a specific terpenol compound in smoke PM_{2.5} to its content per gram of powder as the IP ratio, and has compiled the IP ratios of the terpenol compounds in Table 4. According to the results, the IP ratios of geraniol in the four brands were all >1 , in the descending order of SC $>$ LST $\gg$ Thai Y $>$ Thai B. This indicates that the combustion of incense generates a large amount of geraniol. In the two Taiwanese brands, the IP ratios of geraniol both exceeded 5, presenting a notably greater health risk than the Thai brands, but also a far richer rosy aroma. The IP ratios of other terpenol compounds were determined to be markedly smaller than 1, verifying that the compounds were decomposed after combustion. *D*-Limonene, linalool, and β -citronellol in particular produced smaller IP ratios in the Thai brands than in the Taiwanese brands. In particular, linalool was mostly decomposed in Thai B, shown by its extremely small IP ratio of 0.004. The IP ratios of anisyl alcohol ranged from 0.101 to 0.182 (SC $>$ Thai Y $>$ LST $>$ Thai B), suggesting that a relatively high proportion of anisyl alcohol was decomposed through the combustion of Thai B. Cinnamyl alcohol produced smaller IP ratios in the Taiwanese brands than in the Thai brands.

A cluster analysis of the IP ratios was conducted to explore the terpenol compounds in the powder of different brands and the generation and decomposition of these compounds in the smokes. The Euclidean distances of the IP ratios are presented in Fig. 4, which shows that LST and SC exhibited the shortest linkage distance, indicating that the two brands shared common characteristics in the generation and decomposition of terpenol compounds. Likewise, the relatively short linkage distance between Thai B and Thai Y also indicated that the two brands shared common characteristics in the generation and decomposition of terpenol compounds. The extremely large linkage distance between the Taiwanese and Thai brands was indicative of a considerable disparity in the characteristics of the generation and decomposition of terpenol compounds.

3.4. Mass concentrations of terpenol compounds in incense smokes

The mass concentrations of terpenol compounds in smoke PM_{2.5} aerosols were as listed in Table 5. The PM_{2.5} mass emission factors of the four brands averaged $18.02 \pm 6.02 \text{ mg g}^{-1}$ incense, with the Taiwanese brands exhibiting much higher PM_{2.5} mass emission factors than the Thai brands (SC $>$ LST $\gg$ Thai B $>$ Thai Y). Geraniol produced the highest PM_{2.5} mass concentrations of the four brands, ranging from 2072.3 ± 223.3 to $3358.6 \pm 103.7 \mu\text{g g}^{-1}$ PM_{2.5} mass (mean = $2628.5 \pm 722.6 \mu\text{g g}^{-1}$ PM_{2.5} mass), and constituting 76.31% of the total PM_{2.5} terpenol compounds. The smokes of LST were found to contain the most geraniol per gram PM_{2.5} mass, followed by SC, Thai B, and Thai Y. Geraniol is a potential respiratory hazard to people who are

Table 5
Contribution of individual terpenol compounds to PM_{2.5} mass emitted from incense burning.

Species		Thai B	Thai Y	LST	SC	Average	SD	Percent (%)
PM _{2.5} mass	(mg g ⁻¹ incense)	13.93 ± 0.06	11.08 ± 0.56	22.55 ± 3.09	24.52 ± 1.95	18.02	6.20	
D-Limonene	(μg g ⁻¹ PM _{2.5} mass)	257.97 ± 11.25	151.54 ± 45.30	86.45 ± 30.18	47.71 ± 39.4	135.92	88.94	3.95
Linalool	(μg g ⁻¹ PM _{2.5} mass)	60.23 ± 2.31	57.86 ± 3.17	57.89 ± 2.61	49.50 ± 1.62	56.37	4.76	1.64
β-Citronellol	(μg g ⁻¹ PM _{2.5} mass)	74.92 ± 11.45	122.83 ± 2.22	58.93 ± 4.43	116.40 ± 13.93	93.27	29.76	2.70
Geraniol	(μg g ⁻¹ PM _{2.5} mass)	2102.1 ± 147.8	2072.3 ± 223.3	3358.6 ± 103.7	2980.9 ± 144.6	2628.5	722.6	76.31
Anisyl alcohol	(μg g ⁻¹ PM _{2.5} mass)	372.31 ± 45.65	576.08 ± 9.31	427.99 ± 28.13	337.93 ± 62.07	428.58	102.19	12.44
Cinnamyl alcohol	(μg g ⁻¹ PM _{2.5} mass)	205.43 ± 13.31	136.33 ± 11.35	32.87 ± 4.04	32.69 ± 2.46	101.83	78.60	2.96
Σ PM _{2.5} terpenols	(μg g ⁻¹ PM _{2.5} mass)	3072.9 ± 231.8	3117.0 ± 204.1	4022.7 ± 1040.4	3565.1 ± 33.3	3444.4	581.6	

exposed to it for a long duration, such as worshipers and workers in temples. Because the amounts of geraniol in the PM_{2.5} of the Taiwanese brands were markedly higher than those in the Thai brands, the former pose a greater threat than the latter, although they also provide a richer aroma. Anisyl alcohol was the second most abundant terpenol in the PM_{2.5} after geraniol. Its PM_{2.5} mass concentrations in the four brands ranged from 337.93 ± 62.07 to 576.08 ± 9.31 μg g⁻¹ PM_{2.5} mass, with Thai Y > LST > Thai B > SC, and a mean of 428.58 ± 102.19 μg g⁻¹ PM_{2.5} mass, accounting for 12.44% of the total PM_{2.5} terpenol compounds. The amounts of D-limonene were 47.71 ± 3.94–257.97 ± 11.25 μg g⁻¹ PM_{2.5} mass, with Thai B > Thai Y > LST > SC, and a mean of 135.92 ± 14.87 μg g⁻¹ PM_{2.5} mass, making up 3.95% of total PM_{2.5} terpenol compounds. For cinnamyl alcohol, the amounts were 32.69 ± 2.46–205.43 ± 13.31 μg g⁻¹ PM_{2.5} mass, with Thai B > Thai Y > LST ≈ SC and a mean of 101.83 ± 78.60 μg g⁻¹ PM_{2.5} mass, accounting for 2.96% of the total PM_{2.5} terpenol compounds. The two Taiwanese brands contained similar amounts of cinnamyl alcohol, with SC having the lowest at 32.69 ± 2.46 μg g⁻¹ PM_{2.5} mass, making

it the least abundant terpenol compound in the two Taiwanese brands. The Thai brands produced far more cinnamyl alcohol per gram mass than the Taiwanese brands; Thai B 6.3 times as much and Thai Y 4.2 times as much. The amounts of β-citronellol ranged from 58.93 ± 4.43 to 122.83 ± 2.22 μg g⁻¹ PM_{2.5} mass, with Thai Y > SC > Thai B > LST and a mean of 93.27 ± 29.76 μg g⁻¹ PM_{2.5} mass, making up 2.70% of the total PM_{2.5} terpenol compounds. Finally, the amounts of linalool ranged from 49.50 ± 1.62 to 60.23 ± 2.31 μg g⁻¹ PM_{2.5} mass (mean = 56.37 ± 4.76 μg g⁻¹ PM_{2.5} mass), accounting for 1.64% of the total PM_{2.5} terpenol compounds. Linalool was the least abundant terpenol compound in the Thai brands, and it was found in the order Thai Y > LST ≈ Thai B > SC.

3.5. Total average emissions and mass fractions of terpenol compounds in the smoke PM_{2.5}

Fig. 5 illustrates the proportions of PM_{2.5} organic carbon (OC) in the four brands under the condition of collection upon burning. Of the OC species, the proportion of clove phenols, previously identified by Kuo et al. (2015), was 2.74 ± 0.13–3.33 ± 0.38% of the total PM_{2.5} OC species in the four brands, of which Thai B had the highest proportion, followed by SC > Thai Y > LST. The terpenol compounds made up 0.31 ± 0.09–0.40 ± 0.16% of the total PM_{2.5} OC species. As noted, the Taiwanese brands, particularly LST, contained higher proportions of terpenol compounds than the Thai brands. Furthermore, comparing the proportions of PM_{2.5} terpenol compounds revealed that D-limonene made up 1.39 ± 0.89–8.39 ± 0.40% of PM_{2.5} terpenol compounds, and the Thai brands contained significantly higher proportions of D-limonene than the Taiwanese brands, with Thai B > Thai Y > LST > SC. Linalool accounted for 1.39 ± 0.27–1.96 ± 0.08% of PM_{2.5} terpenol compounds, and the difference in its proportions was negligible. β-Citronellol made up 1.48 ± 2.89–3.95 ± 0.36% of PM_{2.5} terpenol compounds, with Thai Y containing the highest proportion, followed by SC > Thai B > LST. Geraniol, which accounted for 66.33 ± 0.11–83.48 ± 14.59% of PM_{2.5} terpenol compounds, was identified as the primary terpenol compound in the four brands, and the Taiwanese brands contained significantly higher proportions than the Thai brands (SC ≈ LST > Thai B > Thai Y). Anisyl alcohol made up 9.55 ± 0.77–18.53 ± 1.73% of PM_{2.5} terpenol compounds and was thus identified as the second most abundant PM_{2.5} terpenol compound in the four brands. The Thai brands contained higher proportions of this terpenol (Thai Y > Thai B > LST > SC). Cinnamyl alcohol, which accounted for 0.82 ± 0.06–6.69 ± 0.46% of PM_{2.5} terpenol compounds, was also more abundant in the Thai brands (Thai B > Thai Y > SC > LST). The remaining unidentified OC amounted to 96.36 ± 0.19% and 96.88 ± 0.60% of the total, showing that a large number of organic substances were not investigated in the present study.

3.6. Photochemical properties of terpenol compounds in the incense smokes

To investigate the photochemical properties of the PM_{2.5} terpenol compounds, the L/D ratio, defined as the ratio of a particular PM_{2.5} terpenol compound's concentrations in the light and dark indoor settings, of each terpenol compound was calculated (Fig. 6). The L/D ratios

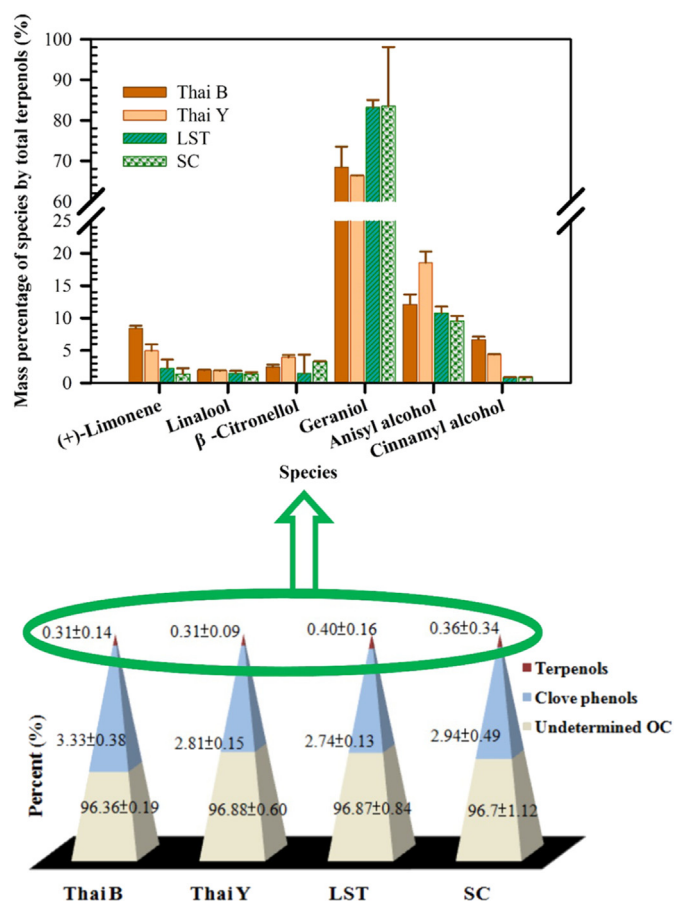


Fig. 5. Average contribution of total terpenols and individual terpenol species to organic carbon (OC) in PM_{2.5} emitted from incense burning.

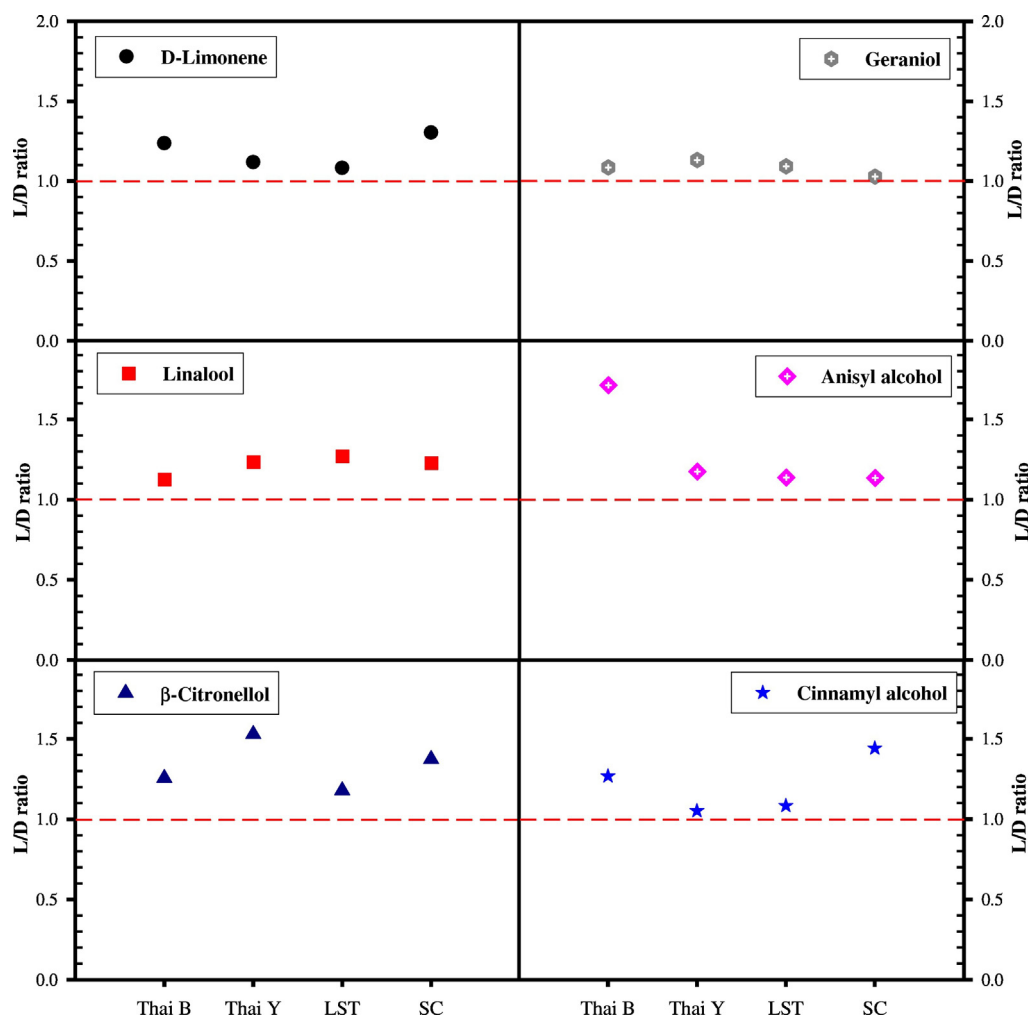


Fig. 6. Photovariation of PM_{2.5} terpenol species emitted from four types of incense burning under the presence and absence of light.

of the terpenol compounds in the four brands were all > 1 , suggesting that the generation of PM_{2.5} terpenol compounds is promoted by both indoor and outdoor lighting. Moreover, because the targeted allergenic substances, namely D-limonene, linalool, β -citronellol, geraniol, anisyl alcohol, and cinnamyl alcohol, are all photosensitive substances that tend to react with ozone and form SOAs in a lighted setting (Donaldson et al., 2002; Kreyling et al., 2004), prolonged exposure to incense smokes in an ill-ventilated and lighted indoor environment could pose an increased allergenic health hazard.

4. Conclusions

This study sought to investigate the emission properties of the allergenic terpenol compounds in four incense brands and compare the photochemical properties of PM_{2.5} terpenol compounds in both light and dark indoor settings. Results revealed that the major terpenol compounds in incense powder varied by brand and place of origin. The powder of the two Thai brands contained more terpenol compounds per unit mass than the two Taiwanese brands, particularly Thai B, with a concentration of $748.72 \pm 1.66 \mu\text{g g}^{-1}$ powder. The most abundant allergenic terpenol compounds in the Thai brands were D-limonene, linalool, and β -citronellol. Of the Taiwanese brands, the powder of SC was found to contain fewer terpenol compounds, with a concentration of only $121.75 \pm 0.06 \mu\text{g g}^{-1}$ powder. The most abundant allergenic terpenol compounds in the powder of the Taiwanese brands were anisyl alcohol, cinnamyl alcohol, and β -citronellol. Thai B contained 6.15 times

more terpenol compounds per unit mass than SC, suggesting that people who experience prolonged exposure to the Thai brands' incense powder, such as incense makers in Thailand, might face a particularly high risk of contact dermatitis. Geraniol, is the most allergenic terpenol but is not one of the major terpenol compounds in the incense powder of the sampled brands.

After ignition, the average PM_{2.5} mass emission factor of the four brands was determined to be $18.02 \pm 6.20 \text{ mg g}^{-1}$ incense. The PM_{2.5} mass emission factors of the two Taiwanese brands were found to be markedly higher than those of the Thai brands; the highest of which, SC, was $24.52 \pm 1.95 \text{ mg g}^{-1}$ incense, whereas the lowest, Thai Y, was $11.08 \pm 0.56 \text{ mg g}^{-1}$ incense. LST had the highest PM_{2.5} emission factor for terpenol compounds, at $89.09 \pm 11.04 \mu\text{g g}^{-1}$ incense, followed by SC $>$ Thai B $>$ Thai Y, suggesting that the smokes of Taiwanese incense are potentially more allergenic than those of Thai incense. The most allergenic terpenol compound, geraniol, was also the greatest contributor to the PM_{2.5} terpenol compounds. In the four brands, geraniol made up 66.3%–83.5% of the PM_{2.5} terpenols, notably at variance with its low concentration of 2.8%–10.7% in incense powder. The IP ratios of geraniol in the four brands were > 1 (SC $>$ LST $\gg$ Thai Y $>$ Thai B), suggesting that large quantities of geraniol are created through the combustion of incense. This may be particularly true for the two Taiwanese brands: as both produced IP ratios > 5 , they pose a much higher health risk than the Thai brands do. The IP ratios of other terpenol compounds were all much smaller than 1, indicating that they were decomposed through combustion. The large distance of Euclidean linkage between the

Taiwanese and Thai brands also indicated that incense from the two locations exhibits markedly different properties in the generation and decomposition of terpenol compounds.

Because the L/D ratios of terpenol compounds were all >1 for the four brands, this study concluded that lighting from both indoor and outdoor sources can increase the generation of PM_{2.5} terpenol compounds in incense smokes. These compounds are also photosensitive target allergenic substances and hence people who are exposed to incense smokes in an indoor environment for a considerable duration, such as workers and worshippers in temples or people who habitually use incense at home, without adequate ventilation, have increased risk of negative health impacts from detained allergenic particles.

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