Journal of Wood Science (2018) 64:823–835
https://doi.org/10.1007/s10086-018-1773-1

ORIGINAL ARTICLE

Profiling of volatile compounds from five interior decoration timbers in Taiwan using TD/GC–MS/FID

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Received: 13 April 2018 / Accepted: 22 October 2018 / Published online: 15 November 2018

Abstract

Volatile organic compounds (VOCs) released from *Chamaecyparis formosensis, Cryptomeria japonica, Cunninghamia lanceolata, Chamaecyparis obtusa* var. *formosana*, and *Taiwania cryptomerioides* five major building and interior decoration timbers and their essential oil components were analyzed using GC–MS and TD/GC–MS/FID. Results showed that *C. obtusa* var. *formosana* had the highest yield of essential oil (3.42%), followed by *C. formosensis* (3.14%), while *C. japonica* had the lowest yield (0.95%). Moreover, oxygenated sesquiterpene was the highest relative content in all five essential oils and their main constituents were *trans*-myrtanol (18.04%), 1-*epi*-cubenol (15.99%), cedrol (62.26%), *α*-cadinol (26.42%), and *α*-cadinol (27.98%), respectively. In terms of emission quantity of top VOC, the results showed the decreasing order of *C. formosensis* (myrtenal, 74.21 mg/m^2) > *T. cryptomerioides* (thujopsene, 12.00 mg/m^2) > *C. lanceolata* (*α*-cedrene, 10.27 mg/m^2) > *C. obtusa* var. *formosana* (*α*-pinene, 8.05 mg/m^2) > *C. japonica* (*α*-cedrene, 4.25 mg/m^2). *C. formosensis* had a greater amount of VOCs emitted and hence gave off more fragrance than *C. obtusa* var. *formosana* initially. However, after indoor exposure of 24 weeks, the VOC emission quantity of *C. obtusa* var. *formosana* exceeded that of *C. formosensis. α*-Cedrene and thujopsene were the top two major VOCs of both *C. lanceolata* and *T. cryptomerioides*. However, they both showed a trend of decrease in emission with prolonged exposure. All five plantation timbers showed good antifungal, antimicrobial, antibacterial, and antitermitic properties, making them ideal materials for interior decoration. Not only do they have strong bioactivities, they can also provide a fragrant and healthy living environment.

Keywords Volatile organic compounds (VOCs) · Thermal desorption/gas chromatography–mass spectrometry/flame ionization detector (TD/GC–MS/FID) · Plantation timbers · Essential oils

Introduction

In general, wooden building and decoration materials are main providers to emission sources of volatile organic compounds (VOCs). A lot of VOCs are concerned due to their potential health impact on human and living environment. Therefore, in addition to containing a large amount of glue in the wood composites, more and more people like to use solid wood as building and interior decoration materials. Besides, in Taiwan, over half of the total area is covered with natural forests and timber plantations. Efficient and effective tending of plantations, which involves silvicultural practices such as planting, spacing, thinning and pruning, contributes to sustainable management, and diverse applications of forest resources. Utilization of thinned wood not only increases self-sufficiency of domestic timber but also enhance economic productivity of plantations. Thinned wood of Taiwan is known for its fine texture, natural color, distinctive aroma, comfortable feel and temperature, low thermal conductivity and good insulation [1]. Moreover, it offers good acoustics for the ears and helps reduce irritation caused by indoor lighting to the eyes through absorbing harmful ultraviolet rays [2]. On the other hand, apart from oxygen produced during photosynthesis, tree growth also yields secondary metabolites such as terpenoids, phenolics, alkaloids, and polyketides. Among them, monoterpene and sesquiterpene constituents are volatile and aromatic. Besides the distinctive fragrance, volatile compounds present in certain wood

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species also possess chemical substances that have been found to be insect-repelling or insect-killing [3, 4], antitermitic [5], antibacterial, and antifungal [6]. Moreover, volatile compounds found in essential oils extracted from wood were also found to exhibit antifungal [7] and antibacterial activities [8], which can enhance decay resistance. The odor released also drives away insect, offering protection against biological hazards [9].

In recent years, solid-phase microextraction (SPME) coupled with gas chromatography–mass spectrometry (GC–MS) has been employed to analyze fragrance compositions of precious coniferous woods [4], to monitor emission of VOCs from the leaves of *Barringtonia racemosa* [10] and to profile volatile compounds of *Phyllostachys pubescens* shoots [11]. Moreover, thermal desorption/gas chromatography–mass spectrometry/flame ionization detector (TD/GC–MS/FID) has also been used for quantitative analyses of floral volatiles emitted from *B. racemosa* [10] and flavoring constituents of *Cinnamomum osmophloeum* leaves [12]. To understand better the characteristics and properties of plantation timbers, this study analyzed five coniferous woods, namely *Chamaecyparis formosensis, Cryptomeria japonica, Cunninghamia lanceolata, Chamaecyparis obtusa* var. *formosana*, and *Taiwania cryptomerioides*, with special focus on their essential oils and volatile compounds. Results thus obtained can shed light on benefits of using these woods for construction, interior decoration and furniture, which in turn would promote the applications of thinned wood and the economic value of plantation timber.

Materials and methods

Experimental materials

Heartwood samples were obtained from five different coniferous trees grown in the Experimental Forest of National Taiwan University. The trees were *Chamaecyparis formosensis* Matsum., *Cryptomeria japonica* (L.f.) D. Don, *Cunninghamia lanceolata* (Lamb.) Hook, *Chamaecyparis obtusa* var. *formosana* (Hayata) Rehder, and *Taiwania cryptomerioides* Hayata aged 45, 30, 30, 55, and 25 years, respectively. The samples were cut into size 5.0 cm × 5.0 cm × 2.0 cm and then stored in a sealed bag at 4 °C without exposure to light prior to analysis.

Chemical compounds used for identification and quantification included α-pinene (98%), myrtenal (98%), and verbenone (94%) purchased from Acros Organics (Dinhaw Enterprise Co., Ltd, New Taipei City, Taiwan); α-cubebene (97%) and α-cedrene (99%) purchased from Sigma–Aldrich (Uni-onward Corp., New Taipei City, Taiwan); and thujopsene (97%) purchased from Fluka (Uni-onward Corp.).

Extraction of essential oils

Essential oils were obtained from the different wood species using steam distillation. Air-dried heartwood sample (200 g) was mixed with distilled water (1000 mL) in a round-bottom extraction flask and subjected to water distillation in a Clevenger-type apparatus for 6 h, followed by determination of oil content (mL/kg) according to sample dry weight. Oil extraction from samples of each wood species was performed in triplicate.

Extraction of VOCs

Figure 1 shows the experimental setup for collection of VOCs at ambient temperature. First, heartwood sample (5.0 cm × 5.0 cm × 2.0 cm) was enclosed in a 3 L glass chamber for 10 min. Air from an air compressor was filtered by the zero air generator (75-83NA, Parker-Balston, Haverhill, MA, USA) and then introduced into the glass chamber at a flow rate of 24 L/h controlled by a mass flow controller (Type 8711, Bürkert Co., Baden-Württemberg, Germany). VOCs were then collected by a low-flow sampler (Gilian Model LFS-113DC, USA) equipped with sampling tubes (Tenax TA mesh 60/80 mesh, Markes International Ltd., Wales, UK) at a flow rate of 100 mL/min for 10 min. To avoid sampling the contaminants from tubing, all tubes were heated at 320 °C for 20 min. The emission rate of VOCs (mg/h·m^2) from the wood sample were calculated using Eq. 1.

$$ER = (C \times Q)/A, \tag{1}$$

where ER is the emission rate (mg/h·m^2), C is the concentration of VOCs (mg/L air), Q is the flow rate of air (24 L/h), and A is the surface area of wood sample (0.0065 m^2).

Thermal desorption

VOCs adsorbed in the sampling tube were thermally desorbed using a thermal desorber (Unity 2, Markes International Ltd., UK). The sorbent tubes were connected to the injection port of the calibration solution loading rig (CSLR, Markes International Ltd., USA) through which the liquid standard was injected. Helium of 99.99% purity was used as the carrier gas with flow rate adjusted by the valve to 100 mL/min. The tubes were purged for 2 min to remove the carrier solvent, methanol. TD proceeded in two stages, first at 240 °C for 10 min and then at 250 °C for another 10 min. The VOCs retained in the tube were then qualitatively and quantitatively analyzed.

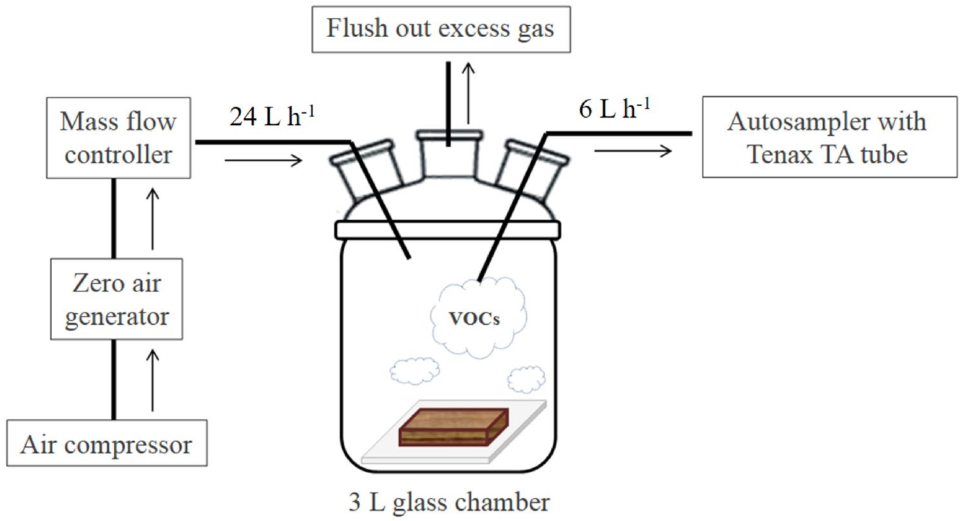

Fig. 1 Experimental setup for VOCs collection from different plantation timbers at ambient temperature

Qualitative analysis of VOCs with GC–MS

Compositions of VOCs emitted from essential oils and wood samples were identified using both GC (Trace GC Ultra, Thermo Scientific, Waltham, MA, USA) and MS (PoLaris Q MSD detector, Thermo Scientific, Waltham, MA, USA). The GC was equipped with a DB-5ms capillary column (J&W Scientific, Folsom, CA, USA) with a length of 30 m, diameter of 0.25 mm and film thickness of 0.25 µm. The other parameters used were injector temperature, 250 °C; carrier gas, helium at a flow rate of 1 mL/min; ion source temperature, 230 °C; and split ratio, 10:1. The initial oven temperature was held at 50 °C for 2 min, then heated at 3 °C/min to 200 °C and held for 1 min, finally at 10 °C/min to 250 °C and held for 2 min. Identification of components in VOCs was confirmed by comparison and co-injection with standards using the National Institute of Standards and Technology (NIST) V2.0, Wiley 7.0 and algorithmic indices (*AI*) [13]. The *AI* was calculated by retention time of sample's and *n*-alkanes' peaks using Eq. 2.

$$AI = 100 \times \left[n + \left(RT_{(x)} - RT_{(n)}\right) / \left(RT_{(n+1)} - RT_{(n)}\right)\right], \quad (2)$$

where $RT_{(n)}$ and $RT_{(n+1)}$ are the adjusted retention time for n and $n+1$ carbon atoms, respectively in n-alkanes, $RT_{(x)}$ is the adjusted retention time for an unknown compound X, $RT_{(x)}$ should fall between $RT_{(n)}$ and $RT_{(n+1)}$.

Quantitative analysis of VOCs with GC–FID

Thermally desorbed VOCs were quantitatively analyzed using GC with a flame ionization detector (7890A, Agilent Technologies, Santa Clara, CA, USA). GC was performed on a DB-5 column (J & W Scientific, Folsom, CA, USA). Two thermal desorption temperatures of 240 and 250 °C were used, and desorption time was 10 min. The oven temperature was held at 40 °C for 4 min, then programmed to increase from 40 to 250 °C at a rate of 4 °C/min and held for 5 min. Other parameters included injector temperature, 260 °C; carrier gas, helium at a flow rate of 1 mL/min; split ratio, 1:30. Each extraction takes about 46.67 min. Quantification was performed by percentage peak area calculations using the GC–FID.

Results and discussion

Yield of essential oils extracted from plantation timbers

Table 1 lists the yield of essential oils from the five plantation timbers studied. As can be seen, *C. obtusa* var. *formosana* had the highest yield of essential oil (3.42%) followed by *C. formosensis* (3.14%), while *C. japonica* had the lowest yield (0.95%). These findings echo those of Chen et al. [14] that *C. obtusa* var. *formosana* yielded more essential oil than *C. formosensis* (18.0 mL/kg vs 16.2 mL/kg, respectively). In the study of Ku et al. [8], essential oils obtained from

Table 1 Yield of essential oils of five different plantation timbers in Taiwan

Species	Yield (%, w/w)
C. formosensis	3.14 ± 0.77
C. japonica	0.95 ± 0.02
C. lanceolata	1.47 ± 0.17
C. obtusa var. *formosana*	3.42 ± 0.10
T. cryptomerioides	1.83 ± 0.06

Data are expressed as mean ± standard error ($n = 3$)

different plant parts of *C. japonica* were in order of leaves (1.42%) > wood (0.38%) > twig (0.05%) > bark (0.03%). Although essential oil from *C. japonica* wood shows the strongest antibacterial activity, its yield is relatively low. Cheng et al. [15] also observed similar low yield in different species of *C. japonica* as in red and black heartwood-type having yield of 0.12–0.30% and 0.34–0.69%, respectively.

Composition of essential oils extracted from plantation timbers

Figure 2 shows the GC spectra of essential oils from the five plantation timbers studied and Table 2 lists their constituents and relative contents. As shown, *C. formosensis* contains 41 compounds; *C. japonica*, 49; *C. lanceolata*, 40; *C. obtusa* var. *formosana*, 44; and *T. cryptomerioides*, 46, accounting for relative contents of 86.24, 89.21, 94.03, 81.12, and 95.60%, respectively.

Moreover, the compounds identified and shown in Table 2 are mainly terpenoids, which can be further classified into six different types. Figure 3 illustrates the relative contents of different types of terpenoids present in the essential oil of the five plantation timbers studied. As can be seen, oxygenated sesquiterpene is the main compound of the highest relative content in all five essential oils. It accounts for 75.08% of the essential oil content of *T. cryptomerioides*, 66.88% of *C. lanceolata*, and 50.45% of *C. obtusa* var. *formosana*. In *C. japonica* essential oil, sesquiterpene (34.63%) is the component of the second highest relative content after oxygenated sesquiterpene (46.69%). In *C. formosensis*, the relative content of oxygenated monoterpene (33.30%) is lower than but close to that of oxygenated sesquiterpene (38.68%). In comparison, the essential oil of the five plantation timbers studied had relatively insignificant contents of monoterpene, diterpene and oxygenated diterpene.

The top three major constituents of essential oils from the five different plantation timbers studied and summarizes their bioactivities against various bacteria, fungi, and insects reported in previous studies. As shown, *C. formosensis* comprises mainly *trans*-myrtanol (18.04%), *α*-cadinol + *α*-eudesmol (17.26%), and myrtenal (14.20%). Its essential oil has been shown to exhibit strong antimicrobial [16], insecticidal [17], and antifungal activities [2, 18]. 1-*epi*-Cubenol (15.99%), *δ*-cadinene (11.96%), and *T*-cadinol + *T*-muurolol (9.26%) are the main constituents in essential oil of *C. japonica* which have been found to possess antifungal [15, 19, 20], antimicrobial [21], insecticidal [3, 9], and antibacterial [8] characteristics. The major constituent in essential oil of *C. lanceolata* is cedrol (62.26%), followed by *α*-terpineol (10.00%) and *α*-cedrene (5.13%). It has shown both antimicrobial [22] and antimite [23] activities. *C. obtusa* var. *formosana* comprises mainly *α*-cadinol (26.42%), *T*-cadinol + *T*-muurolol + *δ*-cadinol (21.49%), and *α*-pinene (4.59%). Its essential oil exhibits antitermitic [5] as well as antibacterial and antifungal activities [6]. *α*-Cadinol (27.98%) is also the main component of *T. cryptomerioides*, followed by *T*-cadinol + *T*-muurolol (23.45%) and cedrol (16.29%). Essential oil of *T. cryptomerioides* possesses antifungal [7], antibacterial [6], and antimite [24] characteristics. In sum, all five plantation timbers show good antifungal, antimicrobial antibacterial, and antitermitic properties, making them ideal materials for interior decoration. Not only do they have strong natural resistance against decay, they can also provide a fragrant and healthy living environment.

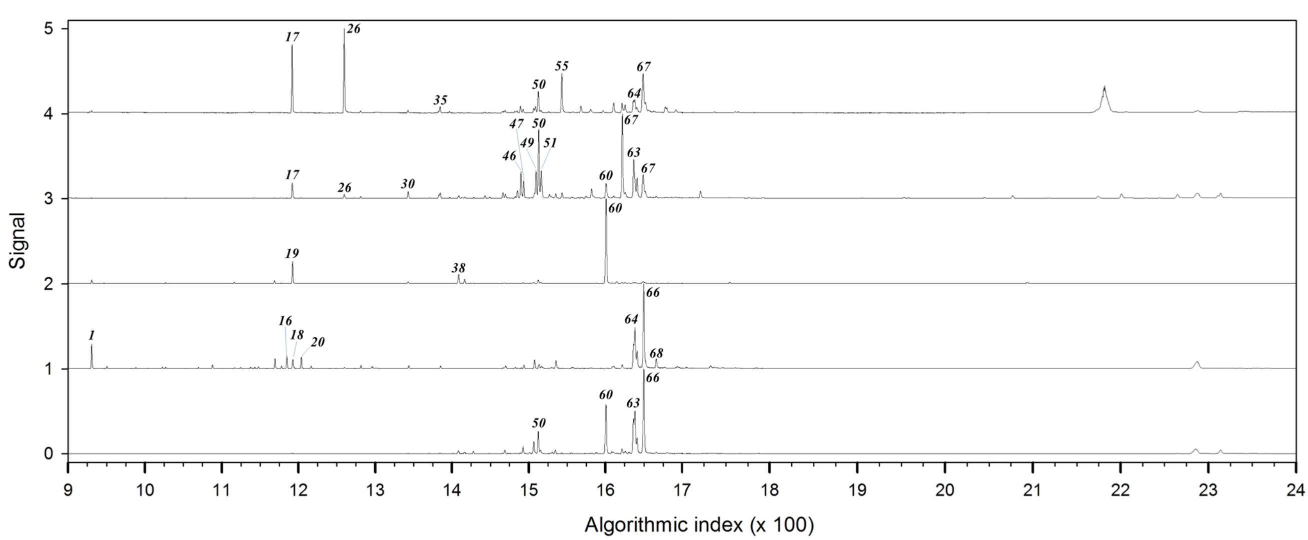

Fig. 2 Gas chromatography (GC) spectra of essential oils from five different plantation timbers in Taiwan (from top to bottom: *C. formosensis, C. japonica, C. lanceolata, C. obtusa* var. *formosana*, and *T. cryptomerioides*)

Table 2 Constituents and relative contents (%, w/w) of essential oils from five different plantation timbers in Taiwan

No.	Compound	*C. formosensis*	*C. japonica*	*C. lanceolata*	*C. obtusa* var. *formosana*	*T. cryptomerioides*
1	*α*-Pinene	0.32 ± 0.14		1.42 ± 0.22	**4.59 ± 1.13**	
2	Camphene			0.09 ± 0.02	0.09 ± 0.05	
3	Thuja-2,4(10)-diene	0.02 ± 0.02	0.01 ± 0.01		0.45 ± 0.10	
4	Limonene	0.03 ± 0.02	0.04 ± 0.01	0.31 ± 0.08	0.29 ± 0.02	
5	Terpinolene			0.08 ± 0.05	0.07 ± 0.00	
6	Fenchone			0.05 ± 0.01		
7	*α*-*p*-Dimethylstyrene	0.11 ± 0.03	0.02 ± 0	0.08 ± 0.08	0.91 ± 0.03	
8	*endo*-Fenchol		0.03 ± 0	0.65 ± 0.15	0.26 ± 0.00	
9	*α*-Campholenal	0.03 ± 0.03	0.01 ± 0.01		0.32 ± 0.02	
10	*cis*-Verbenol	0.13 ± 0.02	0.06 ± 0.01		0.43 ± 0.01	
11	*trans*-Verbenol	0.19 ± 0.02	0.10 ± 0		0.41 ± 0.08	
12	Pinocarvone				0.12 ± 0.00	
13	Isoborneol			0.06 ± 0.05		
14	Borneol	0.14 ± 0.03	0.08 ± 0.01	1.41 ± 0.28	2.94 ± 0.11	0.02 ± 0.01
15	Terpinen-4-ol	0.10 ± 0.02	0.08 ± 0.01	0.14 ± 0.03	0.66 ± 0.01	
16	2,4-Dimethylstyrene	0.04 ± 0.01	0.05 ± 0.00	0.15 ± 0.10	3.54 ± 0.07	
17	Myrtenal	**14.20 ± 3.00**	2.87 ± 0.11			
18	Myrtenal + *α*-Terpineol				3.17 ± 0.13	0.18 ± 0.03
19	*α*-Terpineol			**10.00 ± 2.13**		
20	Verbenone	0.15 ± 0.02	0.09 ± 0.04		3.19 ± 0.03	
21	*endo*-Fenchyl acetate			0.04 ± 0.01		
22	*trans*-Carveol		0.03 ± 0.01		0.68 ± 0.03	
23	*cis*-Carveol				0.06 ± 0.01	
24	Carvone				0.09 ± 0.01	
25	Geraniol				0.15 ± 0.03	
26	*trans*-Myrtanol	**18.04 ± 2.70**	0.87 ± 0.07		0.40 ± 0.05	0.03 ± 0.00
27	Isoborneol acetate	0.33 ± 0.07	0.29 ± 0.02	0.13 ± 0.03	0.92 ± 0.01	0.01 ± 0.01
28	Methyl myrtenate			0.08 ± 0.02		
29	Carvacrol				0.76 ± 0.36	
30	*α*-Cubebene	0.55 ± 0.06	1.27 ± 0.06			0.14 ± 0.01
31	*α*-Terpinyl acetate			0.82 ± 0.16	0.87 ± 0.02	
32	2-Allyl-4-methylphenol				0.07 ± 0.02	
33	*α*-Copaene	0.07 ± 0.01	0.11 ± 0.02	0.05 ± 0.02	0.06 ± 0.00	0.09 ± 0.01
34	*β*-Cubebene + *β*-Elemene	1.56 ± 0.09	1.65 ± 0.02			0.24 ± 0.02
35	*β*-Elemene			0.13 ± 0.01	0.80 ± 0.05	
36	Sibirene	0.27 ± 0.03	0.15 ± 0.01		0.06 ± 0.01	
37	Longifolene			0.02 ± 0.02		0.09 ± 0.01
38	*α*-Cedrene		0.67 ± 0.01	**5.13 ± 0.81**	0.06 ± 0.01	1.12 ± 0.14
39	*trans*-*β*-Caryophyllene	0.03 ± 0.02	0.24 ± 0.01			0.11 ± 0.01
40	*β*-Cedrene		0.29 ± 0.01	2.33 ± 0.21		0.53 ± 0.08
41	Thujopsene		0.13 ± 0.01	0.26 ± 0.04		0.77 ± 0.08
42	*cis*-Muurola-3,5-diene	0.18 ± 0.01	0.48 ± 0.02			0.06 ± 0.00
43	*α*-Humulene	0.06 ± 0.01	0.21 ± 0.01	0.07 ± 0.01		0.10 ± 0.01
44	*β*-Selinene	0.35 ± 0.02		0.11 ± 0.01	0.42 ± 0.04	0.09 ± 0.01
45	*trans*-Muurola-4(14),5-diene	0.41 ± 0.02	1.70 ± 0.12			0.11 ± 0
46	*γ*-Amorphene	1.58 ± 0.11	4.75 ± 0.48			0.24 ± 0.02
47	*α*-Muurolene	0.73 ± 0.03	2.93 ± 0.08	0.48 ± 0.05	1.12 ± 0.10	2.18 ± 0.14
48	*γ*-Cadinene	0.93 ± 0.01		0.70 ± 0.08	2.59 ± 0.15	3.76 ± 0.19
49	Cubebol	1.97 ± 0.48	5.69 ± 0.56			

Table 2 (continued)

No.	Compound	*C. formosensis*	*C. japonica*	*C. lanceolata*	*C. obtusa* var. *formosana*	*T. cryptomerioides*
50	δ-Cadinene	5.31 ± 0.34	**11.96 ± 0.46**	1.86 ± 0.16	1.36 ± 0.16	7.60 ± 0.50
51	*trans*-Calamenene		5.76 ± 0.11	0.70 ± 0.09	1.05 ± 0.08	
52	*trans*-Cadina-1,4-diene	0.20 ± 0.02	1.19 ± 0.11			
53	α-Cadinene	0.11 ± 0.00				0.56 ± 0.05
54	α-Calacorene	0.13 ± 0.01	0.84 ± 0.01	0.12 ± 0.02	2.77 ± 0.25	1.17 ± 0.08
55	β-Elemol	9.20 ± 0.37	1.03 ± 0.05	0.05 ± 0.00	0.16 ± 0.04	0.19 ± 0.01
56	β-Calacorene		0.28 ± 0.01		0.64 ± 0.06	0.30 ± 0.01
57	*E*-Nerolidol	0.24 ± 0.02		0.01 ± 0.01		
58	Globulol			0.31 ± 0.02		0.48 ± 0.03
59	9,11-Epoxy-guaia-3,10(14)-diene	0.54 ± 0.02				
60	Cedrol		3.96 ± 0.48	**62.26 ± 5.17**		**16.29 ± 0.30**
61	1-*epi*-Cubenol	2.32 ± 0.10	**15.99 ± 0.69**	0.44 ± 0.03	1.38 ± 0.17	1.41 ± 0.06
62	γ-Eudesmol	2.28 ± 0.13		0.47 ± 0.05		0.71 ± 0.04
63	*T*-Cadinol + *T*-Muurolol		**9.26 ± 0.16**			**23.45 ± 0.43**
64	*T*-Cadinol + *T*-Muurolol + δ-Cadinol	6.84 ± 0.45		1.13 ± 0.10	**21.49 ± 2.13**	
65	δ-Cadinol		3.79 ± 0.19			4.00 ± 0.17
66	α-Cadinol				**26.42 ± 1.94**	**27.98 ± 0.51**
67	α-Cadinol + α-Eudesmol	**17.26 ± 4.04**	6.97 ± 0.19	1.88 ± 0.16		
68	Cadalene		0.37 ± 0.02	0.22 ± 0.03	3.06 ± 0.12	0.46 ± 0.01
69	α-Bisabolol			0.34 ± 0.03		0.46 ± 0.03
70	10-*nor*-Calamenen-10-one				0.37 ± 0.07	
71	Cryptomerione		1.36 ± 0.05			
72	γ-Costol	0.31 ± 0.25			0.36 ± 0.06	
73	β-Costol	0.55 ± 0.54				0.04 ± 0.01
74	β-Costol + α-Costol				0.27 ± 0.06	
75	α-Costol	0.56 ± 0.58				
76	14-Hydroxy-α-muurolene					0.07 ± 0.06
77	Abietatriene		0.14 ± 0.01			
78	Abietadiene		0.51 ± 0.02			
79	Sandaracopimarinal		0.37 ± 0.05			
80	Phyllocladan-16α-ol		0.88 ± 0.19			
81	Sandaracopimarinol		0.73 ± 0.23			
82	6,7-Hydro-ferruginol		0.27 ± 0.47			
83	Ferruginol		1.30 ± 0.58			1.07 ± 0.76
	Total	86.24	89.21	94.03	81.12	95.60

Top three major constituents in bold

Emission of VOCs from plantation timber at ambient temperature

Figure 4 shows the GC spectra of VOCs emitted from five different plantation timbers at ambient temperature. As can be seen, *C. obtusa* var. *formosana* comprises a greater variety of VOCs than the other four timbers. According to the relative contents of VOCs emitted shown in Fig. 5, sesquiterpene is the dominant VOC released by all five plantation timbers. It accounts for 39.32% of volatiles released from *C. formosensis*, followed very closely by oxygenated monoterpene (38.87%). Emission of these two compounds from *C. formosensis* are markedly higher than those from the other four timbers. Furthermore, as seen in Fig. 5, the relative content of monoterpene emitted from *C. obtusa* var. *formosana* (6.43%) far exceeded that of the other four timbers. Monoterpene compounds are of low molecular weight and have low boiling points, thus making them more voltaile. Hence, compared with that of the four timbers, the fragrance of *C. obtusa* var. *formosana* is more distinctive and more easily sensed.

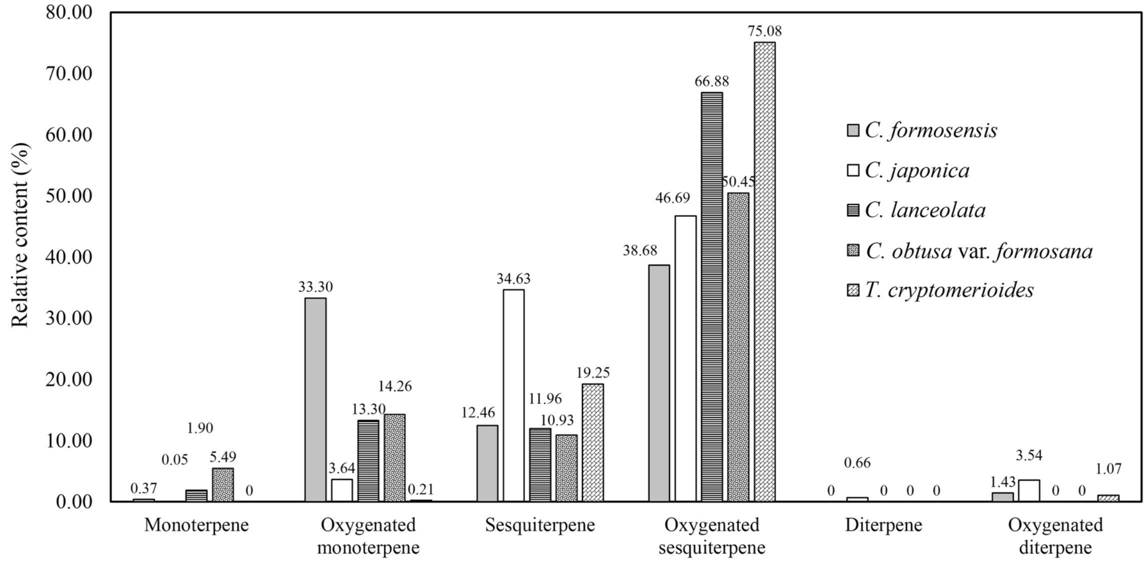

Fig. 3 Relative contents (%, w/w) of terpenoids in essential oil of five different plantation timbers

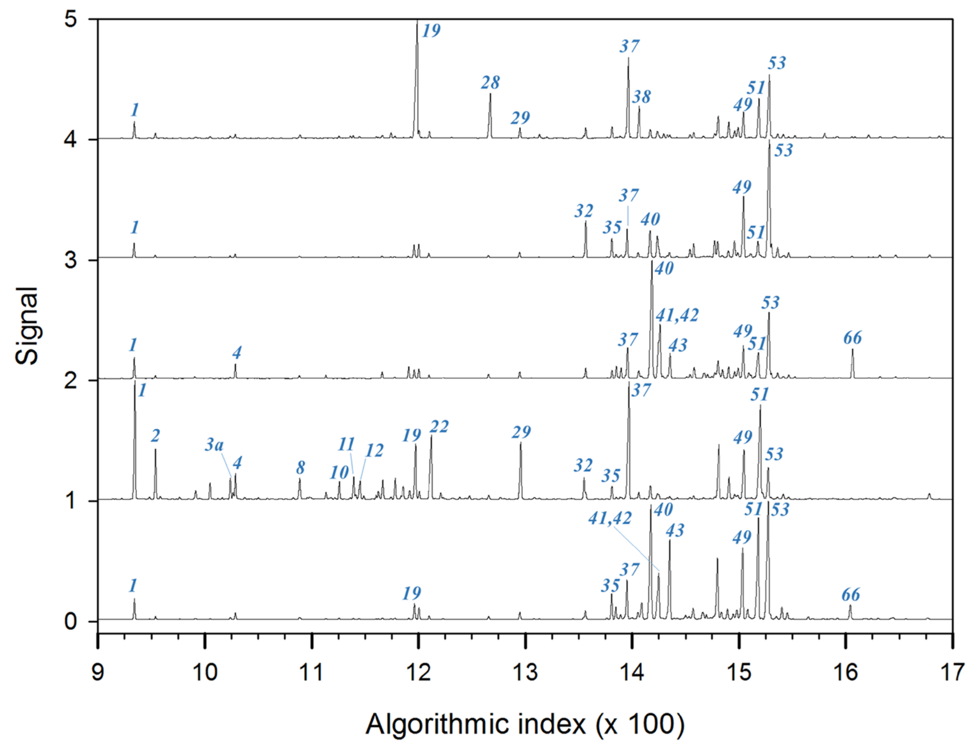

Fig. 4 GC spectra of VOCs emitted from five different plantation timbers at ambient temperature (from top to bottom: *C. formosensis, C. japonica, C. lanceolata, C. obtusa* var. *formosana*, and *T. cryptomerioides*)

Table 3 lists the emission ratio of VOCs from five different plantation timbers at ambient temperature. As can be seen, *C. formosensis* has the highest emission ratio (87.59) while *C. lanceolata* has the lowest emission ratio (18.92). The three main VOCs released by *C. formosensis*, namely myrtenal, β-elemene, and δ-cadinene, more than 61.29% of the total emission amount, while β-elemene, cubebol, α-pinene, and verbenone made up more than 54.21% of the VOCs released from *C. obtusa* var. *formosana*. Chen et al. [14] indicated that essential oils of both *C. formosensis* and *C. obtusa* var. *formosana* have been found to affect the human autonomic nervous system. The physiological effects after 5-min inhalation of *C. formosensis* essential oil included decreased systolic blood pressure and heart rate, increased diastolic blood pressure, decreased sympathetic nervous activity, and increase in parasympathetic activity.

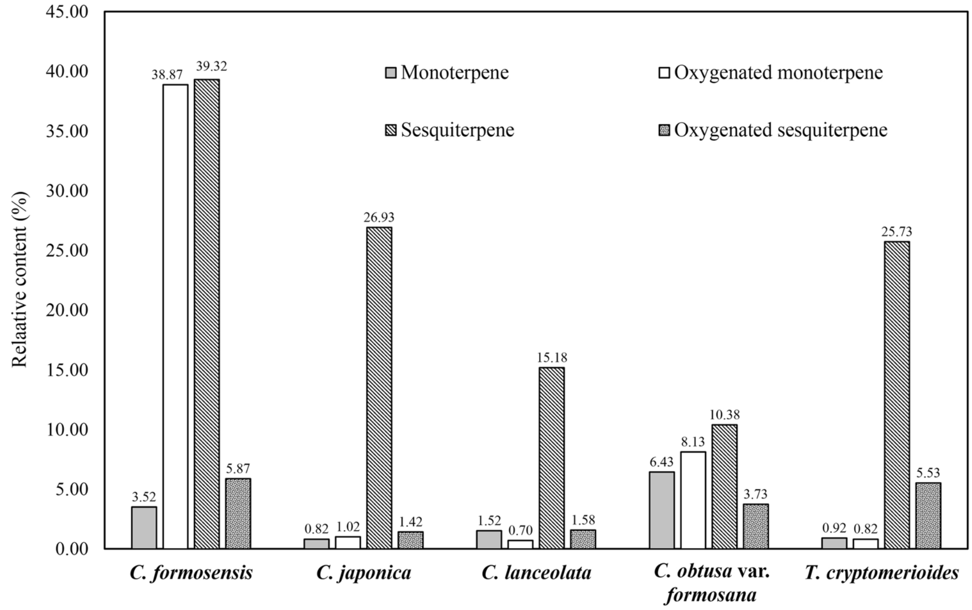

Fig. 5 VOCs emitted from five different plantation timbers at ambient temperature

Table 3 Emission ratio of VOCs from five different plantation timbers at ambient temperature

No.	RT	Compound	*C. formosensis*	*C. japonica*	*C. lanceolata*	*C. obtusa* var. *formosana*	*T. cryptomerioides*
1	10.70	*α*-Pinene	2.31 ± 1.82	0.50 ± 0.10	1.02 ± 0.32	**3.52 ± 2.52**	0.58 ± 0.12
2	11.58	Camphene	0.53 ± 0.20	0.14 ± 0.02	0.10 ± 0.03	1.54 ± 0.93	0.11 ± 0.02
4	14.72	Limonene	0.52 ± 0.34	0.15 ± 0.14	0.36 ± 0.22	1.07 ± 0.51	0.21 ± 0.12
19	22.26	Myrtenal	**28.40 ± 18.02**	0.58 ± 0.33	0.30 ± 0.23	2.64 ± 1.71	0.35 ± 0.21
22	22.84	Verbenone	0.54 ± 0.12	0.12 ± 0.11	0.10 ± 0.12	**3.51 ± 1.84**	0.10 ± 0.11
28	25.09	*trans*-Myrtanol	8.73 ± 5.11	0.14 ± 0.12	0.11 ± 0.12	0.24 ± 0.12	0.12 ± 0.00
29	26.28	Isobornyl acetate	1.24 ± 0.70	0.15 ± 0.10	0.20 ± 0.13	1.82 ± 1.54	0.15 ± 0.10
32	29.08	*α*-Cubebene	1.64 ± 1.11	2.61 ± 1.21	0.35 ± 0.13	0.24 ± 0.22	0.33 ± 0.13
35	30.17	*α*-Copaene	1.60 ± 1.16	1.22 ± 0.53	0.27 ± 0.10	0.44 ± 0.32	1.13 ± 0.31
37	30.84	*β*-Elemene	**15.23 ± 11.52**	1.46 ± 0.80	1.21 ± 0.40	**4.84 ± 3.83**	1.52 ± 0.62
38	31.27	Sibirene	5.11 ± 4.41	0.24 ± 0.20	0.25 ± 0.11	0.32 ± 0.32	0.26 ± 0.10
40	31.73	*α*-Cedrene	1.02 ± 0.40	**2.72 ± 2.11**	**6.81 ± 0.62**	0.83 ± 0.72	**7.00 ± 1.74**
43	32.47	Thujopsene	0.53 ± 0.40	0.43 ± 0.30	1.12 ± 0.00	0.24 ± 0.21	4.21 ± 1.21
45	33.36	*α*-Humulene	0.84 ± 0.50	1.10 ± 0.53	0.63 ± 0.10	0.22 ± 0.11	0.65 ± 0.13
49	35.22	*α*-Muurolene	3.44 ± 1.91	**4.61 ± 2.31**	1.21 ± 0.31	1.72 ± 1.32	3.45 ± 0.84
52	35.73	Cubebol	5.71 ± 3.52	1.02 ± 0.60	**1.42 ± 0.32**	**3.64 ± 3.02**	**5.54 ± 1.31**
53	36.09	*δ*-Cadinene	**10.05 ± 6.43**	**12.63 ± 6.83**	**3.24 ± 0.72**	1.64 ± 1.03	**7.21 ± 1.62**
59	36.82	*β*-Elemol	0.16 ± 0.11	0.32 ± 0.20	0.22 ± 0.00	0.14 ± 0.03	
Total			87.59	30.14	18.92	28.61	32.92

Top three emission ratios in bold

These findings suggest that essential oils of both *C. formosensis* and *C. obtusa* var. *formosana* could be suitable agents for developing regulators of sympathetic nervous system dysfunctions. Furthermore, the increase in parasympathetic activity was attributed to high *δ*-cadinene and *α*-muurolene contents in *C. formosensis* essential oil [14]. This study found that *δ*-cadinene and α-muurolene made up more than 50% of the VOCs emitted from *C. japonica*, and *δ*-cadinene was the major VOCs released from *T. cryptomerioides*. Hence, it can be inferred that *C. japonica* and *T. cryptomerioides* can also stimulate pleasant emotions.

It is generally believed that the fragrance emitted by the timber corresponds to the dominant compound of its essential oil. According to the results mentioned in Table 3, the top three major compounds in the essential oil of the five plantation timbers studied are listed to compare with the VOCs of the highest emission ratios. As seen in Table 4, the two columns contain more differences than similarities. In other words, the dominant compounds in the essential oil are not necessarily the major VOCs emitted. Such discrepancy may be caused by the different extraction approaches. Extraction of essential oil using steam distillation may have yielded more compounds than extraction of VOCs by thermal desorption. This possible reason merits further exploration.

Changes in amounts of VOCs emitted from five plantation timbers over time

Table 5 lists the quantities of major VOCs emitted from five different plantation timbers at ambient temperature. In terms of amount of top VOC emitted from each species, the order was *C. formosensis* (myrtenal, 74.21 mg/m^2) > *T. cryptomerioides* (thujopsene, 12.00 mg/m^2) > *C. lanceolata* (α-cedrene, 10.27 mg/m^2) > *C. obtusa* var. *formosana* (α-pinene, 8.05 mg/m^2) > *C. japonica* (α-cedrene, 4.25 mg/m^2). As can be seen, at ambient temperature, the five plantation timbers showed significant variations in types of VOCs and the amounts emitted. With the noticeably large amount of VOCs emitted, *C. formosensis* gives off a distinctive fragrance, characteristic of myrtenal.

Table 4 Comparison of essential oils and volatile compounds with characteristic odor from five different plantation timbers at ambient temperature in Taiwan

Species	Compounds in essential oil (Relative contents, %)	Volatile components (Emission ratio)
C. formosensis	*trans*-Myrtanol (18.04) *α*-Eudesmol (17.26) Myrtenal (14.20)	Myrtenal (28.40) *β*-Elemene (15.23) *δ*-Cadinene (10.05)
C. japonica	1-*epi*-Cubenol (15.99) *δ*-Cadinene (11.96) *T*-Cadinol + *T*-Muurolol (9.26)	*δ*-Cadinene (12.63) *α*-Muurolene (4.61) *α*-Cedrene (2.72)
C. lanceolata	Cedrol (62.26) *α*-Terpineol (10.00) *α*-Cedrene (5.13)	*α*-Cedrene (6.81) *δ*-Cadinene (3.24) Cubenol (1.42)
C. obtusa var. *formosana*	*α*-Cadinol (26.42) *T*-Cadinol + *T*-Muurolol + *δ*-Cadinol (21.49) *α*-Pinene (4.59)	*β*-Elemene (4.84) Cubenol (3.64) *α*-Pinene (3.52) Verbenone (3.51)
T. cryptomerioides	*α*-Cadinol (27.98) *T*-Cadinol + *T*-Muurolol (23.45) Cedrol (16.29)	*δ*-Cadinene (7.21) *α*-Cedrene (7.00) Cubenol (5.54)

Table 5 Amounts of major VOCs emitted from five different plantation timbers at ambient temperature in Taiwan (mg/m^2)

No.	Compound	*C. formosensis*	*C. japonica*	*C. lanceolata*	*C. obtusa* var. *formosana*	*T. cryptomerioides*
1	*α*-Pinene	6.40 ± 3.75	0.85 ± 0.21	**1.96 ± 0.42**	**8.05 ± 5.61**	1.13 ± 0.27
3a	*p*-Cymene	0.46 ± 0.19	0.14 ± 0.03	0.12 ± 0.03	1.28 ± 0.60	0.13 ± 0.04
4	Limonene	1.53 ± 0.46	0.54 ± 0.30	0.95 ± 0.49	1.95 ± 1.38	0.60 ± 0.14
19	Myrtenal	**74.21 ± 27.13**	1.36 ± 0.76	0.60 ± 0.40	**6.53 ± 4.23**	1.04 ± 0.56
22	Verbenone	1.13 ± 0.10	0.28 ± 0.18	0.19 ± 0.15	**6.91 ± 3.40**	0.21 ± 0.13
28	*trans*-Myrtanol	**22.75 ± 5.00**	0.42 ± 0.11	0.47 ± 0.20	0.72 ± 0.25	0.42 ± 0.10
29	Isobornyl acetate	3.68 ± 1.53	0.57 ± 0.30	0.62 ± 0.18	4.77 ± 3.73	0.58 ± 0.24
35	*α*-Copaene	**6.58 ± 3.95**	**3.72 ± 1.33**	0.95 ± 0.23	1.49 ± 1.11	**3.70 ± 1.00**
40	*α*-Cedrene	1.95 ± 0.18	**4.25 ± 3.29**	**10.27 ± 2.18**	1.52 ± 1.17	**11.43 ± 2.45**
43	Thujopsene	1.74 ± 0.92	1.26 ± 0.95	**2.90 ± 0.41**	0.72 ± 0.64	**12.00 ± 3.04**
45	*α*-Humulene	2.17 ± 0.94	**2.34 ± 1.06**	1.23 ± 0.20	0.41 ± 0.32	1.56 ± 0.29

Top three major VOCs in bold

Tables 6, 7 and 8 display the changes in amounts of VOCs emitted from *C. formosensis, C. obtusa* var. *formosana*, and *C. japonica*, respectively over time, with indoor exposure of different durations ranging from 1 to 24 weeks. As seen in Table 6, myrtenal, *trans*-myrtanol, and *α*-copaene remained the top three major VOCs emitted by *C. formosensis* till exposure of 4 weeks. However, they all showed a trend of decrease in emission with prolonged exposure from the initial amounts of 74.21, 22.75, and 6.58 mg/m^2 to 3.07, 1.78, and 0.37 mg/m^2 at 4 weeks and 0.06, 0.20, and 0 mg/m^2 at 24 weeks, respectively. Note that after 12 weeks of exposure, myrtenal was no longer the major VOC emitted; its emission quantity was surpassed by that of *trans*-myrtanol, *α*-cedrene, and thujopsene.

Similar trend of decrease in emission quantity with increase in exposure time was also observed in *C. obtusa* var. *formosana*. As shown in Table 7, *α*-pinene, verbenone, and myrtenal were the top three major VOCs emitted by *C. obtusa* var. *formosana* only till 1-week exposure. Thereafter, *trans*-myrtanol exceeded myrtenal in terms of emission quantity. Although *α*-pinene and verbenone were persistently among the top three, the amounts emitted by them dropped from the initial 8.05 and 6.91 mg/m^2 to 0.68 and 0.44 mg/m^2 at 4 weeks and 0.12 and 0.13 mg/m^2 at 24 weeks, respectively.

Comparing Tables 6 and 7 reveals that *C. formosensis* has a greater amount of VOCs emitted and hence gives off more fragrance than *C. obtusa* var. *formosana* initially. However, after indoor exposure of 24 weeks, the VOCs emission quantity of *C. obtusa* var. *formosana* exceeded that of *C. formosensis*. In other words, over time, the aroma of *C. obtusa* var. *formosana* will surpass that of *C. formosensis*.

As for *C. japonica*, the same trend of the longer the exposure, the smaller the amount of VOC emitted was observed.

Table 6 Changes in amounts of major VOCs emitted from *C. formosensis* at ambient temperature over time (mg/m^2)

No.	RT	Compound	Exposure time (weeks)					
			0	1	2	4	12	24
1	10.70	*α*-Pinene	6.40 ± 3.75	1.00 ± 0.03	0.54 ± 0.03	0.07 ± 0.05		
3a	14.52	*p*-Cymene	0.46 ± 0.19	0.08 ± 0.01	0.07 ± 0.00	0.04 ± 0.00	0.04 ± 0.00	0.01 ± 0.00
4	14.72	Limonene	1.53 ± 0.46	0.22 ± 0.06	0.14 ± 0.01	0.10 ± 0.01	0.02 ± 0.00	0.01 ± 0.00
19	22.26	Myrtenal	**74.21 ± 27.13**	**7.17 ± 2.64**	**5.27 ± 1.29**	**3.07 ± 0.63**	0.08 ± 0.01	**0.06 ± 0.01**
22	22.84	Verbenone	1.13 ± 0.10	0.15 ± 0.03	0.11 ± 0.01	0.05 ± 0.01	0.02 ± 0.00	
28	25.09	*trans*-Myrtanol	**22.75 ± 5.00**	**2.99 ± 0.22**	**2.03 ± 0.15**	**1.78 ± 0.14**	**0.47 ± 0.05**	**0.20 ± 0.04**
29	26.28	Isobornyl acetate	3.68 ± 1.53	0.41 ± 0.13	0.29 ± 0.07	0.19 ± 0.04	0.04 ± 0.00	0.01 ± 0.00
35	30.17	*α*-Copaene	**6.58 ± 3.95**	**0.85 ± 0.32**	**0.60 ± 0.16**	**0.37 ± 0.10**	0.01 ± 0.00	
40	31.73	*α*-Cedrene	1.95 ± 0.18	0.54 ± 0.04	0.45 ± 0.01	0.29 ± 0.02	**0.19 ± 0.00**	**0.05 ± 0.01**
43	32.47	Thujopsene	1.74 ± 0.92	0.30 ± 0.01	0.24 ± 0.01	0.14 ± 0.05	**0.09 ± 0.00**	0.01 ± 0.00
45	33.36	*α*-Humulene	2.17 ± 0.94	0.33 ± 0.09	0.23 ± 0.04	0.16 ± 0.03		

Top three major VOCs in bold

Table 7 Changes in amounts of major VOCs emitted from *C. obtusa* var. *formosana* at ambient temperature over time (mg/m^2)

No.	RT	Compound	Exposure time (weeks)					
			0	1	2	4	12	24
1	10.70	*α*-Pinene	**8.05 ± 5.61**	**1.28 ± 0.25**	**0.76 ± 0.24**	**0.68 ± 0.19**	**0.10 ± 0.07**	**0.12 ± 0.03**
3a	14.52	*p*-Cymene	1.28 ± 0.60	0.20 ± 0.04	0.14 ± 0.07	0.11 ± 0.01	0.07 ± 0.01	0.02 ± 0.01
4	14.72	Limonene	1.95 ± 1.38	0.36 ± 0.06	0.27 ± 0.09	0.18 ± 0.05	0.09 ± 0.01	0.03 ± 0.02
19	22.26	Myrtenal	**6.53 ± 4.23**	**0.54 ± 0.10**	0.31 ± 0.18	0.17 ± 0.02	0.01 ± 0.02	0.02 ± 0.02
22	22.84	Verbenone	**6.91 ± 3.40**	**0.94 ± 0.42**	**0.70 ± 0.66**	**0.44 ± 0.16**	**0.18 ± 0.09**	**0.13 ± 0.10**
28	25.09	*trans*-Myrtanol	0.72 ± 0.25	0.45 ± 0.01	**0.35 ± 0.01**	**0.34 ± 0.02**	**0.27 ± 0.00**	**0.06 ± 0.00**
29	26.28	Isobornyl acetate	4.77 ± 3.73	0.42 ± 0.09	0.31 ± 0.18	0.21 ± 0.02	0.09 ± 0.01	0.04 ± 0.01
35	30.17	*α*-Copaene	1.49 ± 1.11	0.14 ± 0.02	0.08 ± 0.03	0.04 ± 0.00		
40	31.73	*α*-Cedrene	1.52 ± 1.17	0.35 ± 0.03	0.26 ± 0.02	0.21 ± 0.01	0.20 ± 0.01	0.04 ± 0.01
43	32.47	Thujopsene	0.72 ± 0.64	0.19 ± 0.02	0.11 ± 0.03	0.09 ± 0.00	0.09 ± 0.00	0.01 ± 0.00
45	33.36	*α*-Humulene	0.41 ± 0.32	0.08 ± 0.03				

Top three major VOCs in bold

Table 8 Changes in amounts of major VOCs emitted from *C. japonica* at ambient temperature over time (mg/m^2)

No.	RT	Compound	Exposure time (weeks)					
			0	1	2	4	12	24
1	10.70	*α*-Pinene	0.85 ± 0.21					
3a	14.52	*p*-Cymene	0.14 ± 0.03	0.05 ± 0.01	0.04 ± 0.00	0.05 ± 0.01	0.04 ± 0.00	0.01 ± 0.00
4	14.72	Limonene	0.54 ± 0.30	0.10 ± 0.02	0.04 ± 0.02	0.07 ± 0.01	0.03 ± 0.02	0.01 ± 0.00
19	22.26	Myrtenal	1.36 ± 0.76	0.40 ± 0.06	0.09 ± 0.03	0.11 ± 0.02		
22	22.84	Verbenone	0.28 ± 0.18	0.11 ± 0.01	0.04 ± 0.00	0.06 ± 0.01	0.02 ± 0.00	0.01 ± 0.00
28	25.09	*trans*-Myrtanol	0.42 ± 0.11	**0.47 ± 0.00**	**0.32 ± 0.01**	**0.36 ± 0.02**	**0.27 ± 0.00**	**0.05 ± 0.00**
29	26.28	Isobornyl acetate	0.57 ± 0.30	0.11 ± 0.01	0.05 ± 0.01	0.06 ± 0.02	0.04 ± 0.00	0.01 ± 0.00
35	30.17	*α*-Copaene	**3.72 ± 1.33**	**0.44 ± 0.09**	**0.17 ± 0.08**	**0.19 ± 0.08**	0.01 ± 0.02	0.01 ± 0.00
40	31.73	*α*-Cedrene	**4.25 ± 3.29**	**0.55 ± 0.08**	**0.27 ± 0.04**	**0.28 ± 0.02**	**0.20 ± 0.01**	**0.05 ± 0.01**
43	32.47	Thujopsene	1.26 ± 0.95	0.29 ± 0.02	0.12 ± 0.01	0.10 ± 0.02	**0.09 ± 0.00**	0.01 ± 0.00
45	33.36	*α*-Humulene	**2.34 ± 1.06**	0.31 ± 0.07	0.12 ± 0.06	0.13 ± 0.05	0.01 ± 0.00	

Top three major VOCs in bold

As seen in Table 8, the initial top three VOCs emitted were *α*-cedrene, *α*-copaene, and *α*-humulene at quantities of 4.25, 3.72, and 2.34 mg/m^2, respectively. Murugesan et al. [25] reported that *α*-cedrene was the major component (92.40%) of hexane extracts from *Coleus forskohlii* roots and exhibited antimicrobial/insecticidal activities. *α*-Copaene has been found to be the constituent present in the bioactive compounds of Thailand-grown red and white guava fruit with prebiotic activity [26]. It also has the potential to serve as a chemical cue to facilitate orientation of Mediterranean fruit flies, *Ceratitis capitate*, to enhance their success of mating [27]. Table 5 shows that *α*-copaene is among the top three VOCs emitted by *C. formosensis, C. japonica*, and *T. cryptomerioides* at quantities of 6.58, 3.72, and 3.70 mg/m^2, which dropped to 0.01, 0.01, and 0 mg/m^2 at 12 weeks, respectively. In other words, over time, *C. formosensis, C. japonica,* and *T. cryptomerioides* no longer gives off the characteristic aroma of *α*-copaene. Previous studies have also found that *α*-humulene present in essential oil of *Abies balsamea* exhibited activity against solid tumor cell lines tested through inducing dose- and time-dependent decrease in content of glutathione and increase in production of reactive oxygen species [28].

Figure 6 compares the changes in quantities of *α*-cedrene and thujopsene emitted by *C. lanceolata* and *T. cryptomerioides* over time. Again, the declining trend

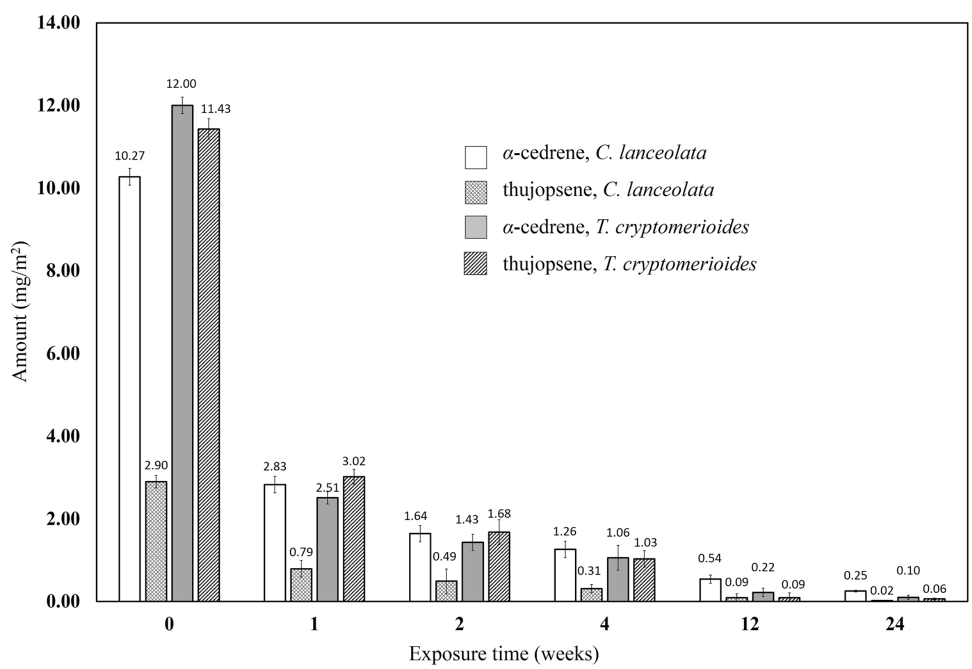

Fig. 6 Changes in amounts of *α*-cedrene and thujopsene emitted from *C. lanceolata* and *T. cryptomerioides* at ambient temperature over time (mg/m^2)

over prolonged exposure is obvious and significant. After 24 weeks of exposure, the emission quantities of *α*-cedrene and thujopsene from *C. lanceolata* were 0.25 and 0.02 mg/m^2, respectively; while those from *T. cryptomerioides* became insignificant at 0.10 and 0.06 mg/m^2, respectively.

Note that *α*-cedrene and thujopsene are the top two major VOCs of both *C. lanceolata* and *T. cryptomerioides*. While the emission quantities of *α*-cedrene in the two plantation timbers are comparable (10.27 and 12.00 mg/m^2, respectively), those of thujopsene are significantly different (2.90 and 11.43 mg/m^2, respectively), thus accounting for their distinctively different aroma. α-Cedrene in essential oil of *C. lanceolata* shows antimite activity [23]. Jeong et al. [29] reported that thujopsene exhibited anti-inflammatory, antispasmodic, tonic, astringent, diuretic, sedative, insecticidal, and antifungal activities. Not only does thujopsene have the potential to be developed into botanical drugs, it can provide a fragrant and healthy living environment.

Conclusions

Analysis of essential oils extracted from *Chamaecyparis formosensis, Cryptomeria japonica, Cunninghamia lanceolata, Chamaecyparis obtusa* var. *formosana*, and *Taiwania cryptomerioides* revealed the highest yield from *Chamaecyparis obtusa* var. *formosana*, followed by *C. formosensis* with *C. japonica* giving the lowest yield. The main compound of highest relative contents in essential oils of these five plantation timbers was oxygenated sesquiterpene while their main constituents were *trans*-myrtanol, 1-*epi*-cubenol, cedrol, *α*-cadinol, and *α*-cadinol, respectively. *C. obtusa* var. *formosana* emitted a greater variety of VOCs than the other four timbers, while emissions of sesquiterpene and oxygenated monoterpene from *C. formosensis* were markedly higher than those from the other four timbers. In terms of emission quantity of top VOC for each species, the results showed the order of *C. formosensis* (myrtenal) > *T. cryptomerioides* (thujopsene) > *C. lanceolata* (α-cedrene) > *C. obtusa* var. *formosana* (*α*-pinene) > *C. japonica* (*α*-cedrene). *C. formosensis* had a greater amount of VOCs emitted and hence gave off more fragrance than *C. obtusa* var. *formosana* initially. However, after 4-week indoor exposure, the VOC emission quantity of *C. obtusa* var. *formosana* exceeded that of *C. formosensis*. In all five plantation timbers, the same trend of the longer the indoor exposure, the smaller the amount of VOC emitted was observed. They also showed good antifungal, antimicrobial, antibacterial, and antitermitic properties, making them ideal materials for interior decoration. Not only do they have strong bioactivities, they can also provide a fragrant and healthy living environment.

Acknowledgements This study was supported by a Grant (107-A04-3) from the Experimental Forest, College of Bioresource and Agriculture, National Taiwan University, Taiwan, ROC. We also thank the Forestry Bureau for financial support.

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