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ARTICLE

Fumigant Antitermitic Activity of Plant Essential Oils and Components from Ajowan (*Trachyspermum ammi*), Allspice (*Pimenta dioica*), Caraway (*Carum carvi*), Dill (*Anethum graveolens*), Geranium (*Pelargonium graveolens*), and Litsea (*Litsea cubeba*) Oils against Japanese Termite (*Reticulitermes speratus* Kolbe)

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Plant essential oils from 26 plant species were tested for their insecticidal activities against the Japanese termite, *Reticulitermes speratus* Kolbe, using a fumigation bioassay. Responses varied with source, exposure time, and concentration. Among the essential oils tested, strong insecticidal activity was observed with the essential oils of ajowan (*Trachyspermum ammi*), allspice (*Pimenta dioica*), caraway (*Carum carvi*), dill (*Anethum graveolens*), geranium (*Pelargonium graveolens*), and litsea (*Litsea cubeba*). The composition of six essential oils was identified by using gas chromatography−mass spectrometry. The compounds thus identified were tested individually for their insecticidal activities against Japanese termites. Responses varied in a dose-dependent manner for each compound. Phenol compounds exhibited the strongest insecticidal activity among the test compounds; furthermore, alcohol and aldehyde groups were more toxic than hydrocarbons. The essential oils and compounds described herein merit further study as potential fumigants for termite control.

KEYWORDS: Plant essential oils; *Reticulitermes speratus*; antitermitic activity; fumigant; ajowan; allspice; caraway; dill; geranium; litsea; thymol; carvacrol; eugenol

INTRODUCTION

Due to their wood-eating habits, several termite species cause serious damage to houses and wooden structures. Because they remain well concealed, their presence is often undetected until the timber is severely damaged from within and shows surface changes, which typically appear last. Once termites have entered a building, they damage not only wood but also paper, cloth, carpets, and other cellulosic materials. The annual damage to wooden structures and other cellulosic materials by termites has been estimated to exceed U.S. $3 billion worldwide (*1*). Among termite species, subterranean termites from the genera *Reticulitermes* and *Coptotermes* are the most economically important species worldwide (*2*). The Japanese termite, *Reticulitermes speratus* Kolbe, is the major termite species distributed throughout Korea, Japan, and China. Recently, in Korea, this pest has caused serious damage to important cultural assets such as temples and palaces.

Control of the Japanese termite in Korea is primarily dependent upon continued applications of synthetic pesticides or traditional wood preservatives (*3*). Although effective, there are concerns regarding the use of such pesticides leading to environmental pollution and health disorders. To avoid these problems, there have been efforts to use plant essential oils as potential alternatives to currently used termite control agents because they constitute a rich source of bioactive chemicals (*4*, *5*); moreover, they are known to be safe as they are commonly used as fragrances and flavoring agents for foods and beverages (*6*, *7*). Furthermore, plant essential oils are highly volatile and therefore do not leave toxic residues.

In this study, we selected 26 commercially available plant essential oils that are widely used as fragrances and flavoring agents; we assessed their fumigant toxicity against adult Japanese termites using fumigation bioassays.

MATERIALS AND METHODS

Termites. Japanese termites (*Reticulitermes speratus* Kolbe) were collected from several damaged pine tree logs at Hongneung arboretum, Seoul, Republic of Korea, from March to September 2007 and 2008. Termite-infested wood moistened with distilled water was kept in plastic cages (60×40×40 cm) at 25 ± 1 °C and 80% relative humidity.

Chemicals. Plant essential oils were purchased from Jinarome (USA) (**Table 1**). Information regarding the chemicals used in this experiment is given in **Table 3**.

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Table 1. Plant Essential Oils Tested

oil	source of plant	part	origin
ajowan	*Trachyspermum ammi*	seeds	India
allspice	*Pimenta dioica*	berries	Jamaica
amyris	*Amyris balsamifera*	wood	Caribbean
Artemisia afra	*Artemisia afra*	flowering plant	South Africa
cabreuva	*Myrocarpus fastigiatus*	wood	Brazil
cajeput	*Melaleuca cajuputii*	leaves	Indonesia
cananga	*Cananga odorata*	blossoms	Indonesia
cardamom	*Elettaria cardamomum*	seeds	Equador
carrot seeds	*Daucus carota*	seeds	France
caraway	*Carum carvi*	seeds	Egypt
clementine	*Citrus clementina*	rind	South Africa
copaiva	*Copaifera reticulata*	resin	Brazil
coriander	*Coriandrum sativum*	fruits	Argenitina
davana	*Artemisia pallens*	leaves	India
dill	*Anethum graveolens*	seeds	Bulgaria
elemi	*Canarium luzonicum*	resin	Phillipines
fokienia	*Fokienia hodgensii*	wood	Vietnam
frankincense	*Boswellia carterii*	resin	Ethiopia
galbanum	*Ferula galbaniflua*	resin	Iran
geranium	*Pelargonium graveolens*	leaves	Reunion
gurjum	*Dipterocarpus turbinatus*	resin	Indonesia
hyssop	*Hyssopus officinalis*	flowering plant	France
larch	*Larix europea*	resin	Austria
lavandin	*Lavandula hybrida*	flowering plant	France
litsea	*Litsea cubeba*	fruits	Vitenam
patchouli	*Pogostemon patchouli*	whole plant	Indonesia

Instrumental Analysis. Gas chromatography (GC) analysis was performed using an Agilent 6890N equipped with a flame ionization detector. Retention times for comparison with authentic compounds were measured with a DB-1MS and a HP-INNOWax column (30 m × 0.25 mm i.d., 0.25 μm film thickness; J&W Scientific, Folsom, CA). The oven temperature was programmed as follows: isothermal at 40 °C for 1 min, raised to 250 °C at 6 °C/min, and maintained at this temperature for 4 min. Helium was used as the carrier gas at a flow rate of 1.5 mL/min. To determine the configurations of α-pinene, β-pinene, and limonene, a chiral column—Beta DEX120 (30 m × 0.25 mm i.d., 0.25 μm; Supelco, Bellefonte, PA)—was used. The oven temperature was maintained at 100 °C for 20 min, and the flow rate of the carrier gas was 1.0 mL/min. For the chiral GC separation of carvone, a Beta DEX 225 (30 m × 0.25 mm i.d., 0.25 μm; Supelco) was used. The temperature program was as follows: 130 °C for 10 min raised to 200 °C at a rate of 10 °C/min. The carrier gas had a flow rate of 1.0 mL/min. GC−mass spectrometry (GC-MS) analysis was performed on an Agilent 7890A coupled with a 5975C mass selective detector (MSD). A DB-5MS (30 m ×0.25 mm i.d., 0.25 μm film thickness; J&W Scientific) was used for the separation of the analytes. The oven temperatures were identical to those used for GC. Helium was the carrier gas, at a flow rate of 1.0 mL/min. Infrared (IR) spectra were recorded on a Nicolet FT-IR (Thermo Fisher Scientific Inc.) spectrometer.

Synthesis. Citronellyl acetate, neryl acetate, 2-phenylethyl acetate, and rose acetate (2,2,2-trichloro-1-phenylethyl acetate) were obtained by acetylation of the corresponding alcohol with acetic acid anhydride and pyridine by using *p*-toluenesulfonic acid as catalyst in CH_2Cl_2. The corresponding alcohol of rose acetate, 2,2,2-trichloro-1-phenylethanol, was prepared according to a previously reported method (*8*). The structure was confirmed by comparison of its mass spectrum with data from the NIST mass spectrum library and the IR spectrum.

Neryl acetate: yield, 93.9%; purity, 99.2%; IR (neat, cm^{-1}) 2972 (m), 1743 (s), 1379 (m), 1240 (m), and 1026 (m); GC-MS (*m*/*z*, %) 196 (M^+, 0.1), 154 (2.3), 153 (0.2), 136 (16.9), 121 (22.7), 107 (7.7), 93 (57.7), 80 (21.9), 69 (100), and 53 (10.7).

2-Phenylethyl acetate: yield, 89.6%; purity, >99.0%; GC-MS (*m*/*z*, %) 163 ($M^+ - H$, 0.01), 134 (0.02), 121 (0.2), 105 (11.0), 104 (100), 91 (17.7), 78 (5.5), 77 (5.4), 66 (0.4), and 43 (27.4).

Citronellyl acetate: yield:, 98%; purity, 96.5%; GC-MS (*m*/*z*, %) 155 ($M^+ - OAc$, 0.1), 138 (47.3), 123 (74.0), 109 (36.7), 95 (97.6), 81 (100), 69 (96.1), 67 (65.1), 55 (46.3), and 43 (77.7).

2,2,2-Trichloro-1-phenylethanol: KOH (1.84 g, 33.0 mmol; Wako) was added to a solution of benzaldehyde (3.18 g, 30.0 mmol; TCI) and $CHCl_3$ (4 mL) at 0 °C. The solution was stirred for 1 h at 40−50 °C, and 2% H_2SO_4 solution was then poured. The solution was diluted with diethyl ether and washed with $NaHCO_3$, H_2O, and brine and dried over $MgSO_4$. After purification by SiO_2 column chromatography and distillation, 2.44 g of the desired alcohol was obtained (yield, 36.3%; purity, 98.4% based on GC). GC-MS (*m*/*z*, %) values were as follows: 224 (M^+, 0.1), 125 (10.0), 107 (B, 100), 79 (52.7), 77 (32.6), and 51 (9.0). IR (neat, cm^{-1}) values were as follows: 3600−3100 (br s), 3062 (s), 3032 (s), 2980 (s), 1497 (s), 1454 (s), 820 (s), and 700 (s).

Rose acetate: yield, 74.5%; purity, >99% based on GC; GC-MS (*m*/*z*, %) 266 (M^+, 0.3), 230 (0.6), 209 (0.7), 190 (2.5), 172 (6.0), 149 (40.0), 125 (4.2), 107 (100), 79 (13.6), and 43 (45.9).

Antitermitic Activity. Antitermitic activity was evaluated using a fumigation bioassay. A paper disk (8 mm, Advantec) treated with the essential oil or compound being tested was placed in the bottom lid of a glass cylinder (5 cm diameter×10 cm) with a wire sieve fitted 3.5 cm above the bottom; thereafter, the lid was sealed. Ten adult worker termites were placed on the sieve. This prevented the direct contact of the termites with the test plant oils and compounds. Filter paper soaked with water was supplied as food. The insects were maintained at 25 ± 1 °C and 80% relative humidity. The adult termites were considered to be dead if appendages did not move when prodded with a brush. Cumulative mortalities were determined 2 and 7 days after treatment. All treatments were replicated five times.

Statistical Analyses. Mortality of termites was transformed to arcsine square root values for analysis of variance (ANOVA). Mean values for treatment data were compared and separated by Scheffé test (*9*).

RESULTS

Antitermitic Activity of Plant Essential Oils. When 26 plant essential oils were bioassayed, termite mortalities varied according to the oil type, dose, and exposure time (**Table 2**). Of these, 6 essential oils, ajowan (*Trachyspermum ammi*), allspice (*Pimenta dioica*), caraway (*Carum carvi*), dill (*Anethum graveolens*), geranium (*Pelargonium graveolens*), and litsea (*Litsea cubeba*), achieved >80% mortality 2 days after treatment at 2 mg/filter paper. *Artemisia afra*, cajept (*Melaleuca cajuputii*), cananga (*Cananga odorata*), cardamom (*Elettaria cardamomum*), coriander (*Coriandrum sativum*), elemi (*Canarium luzonicum*), hyssop (*Hyssopus officinalis*), larch (*Larix europea*), and lavindin (*Lavandula hybrid*) showed >80% antitermitic activity 7 days after treatment. Plant essential oils showing >80% mortality at 2 mg/filter paper were tested at lower concentrations. The insecticidal activity of allspice essential oil was 88% 7 days after treatment at 0.5 mg/filter paper; however, this decreased to 0% at 0.25 mg/filter paper. Antitermitic activities of ajowan, caraway, and litsea were 86, 100, and 96%, respectively, 7 days after treatment at 1 mg/filter paper; however, these decreased to 2, 22, and 2%, respectively, at 0.5 mg/filter paper. Essential oils of dill and geranium produced 92 and 84% insecticidal activities at 1.5 mg/filter paper but showed 76 and 78% mortality at 1 mg/filter paper, respectively.

Chemical Analyses of Active Essential Oils. The chemical compositions of the six active essential oils—ajowan (*T. ammi*), allspice (*P. dioica*), caraway (*C. carvi*), dill (*A. graveolens*), geranium (*P. graveolens*), and litsea (*L. cubeba*)—are shown in **Table 3**. Retention indices were obtained using an equation proposed by van Den Dool and Kratz (*12*). We determined the configurations of α-pinene, β-pinene, and limonene by using a chiral column (Beta DEX120). α-Pinene in litsea essential oil consisted of (*S*)-(−)-α-pinene (0.51%) and (*R*)-(+)-α-pinene (0.71%). Only (*R*)-(+)-α-pinene was detected in ajowan oil. Ratios of (+)-β-pinene and (−)-β-pinene were 0.31 and 0.75% in litsea oil, respectively. Only (+)-β-pinene was identified in ajowan oil. In the case of limonene, two isomers were identified in ajowan and litsea oils, but only (*R*)-(+)-limonene was determined

Table 2. Fumigant Antitermitic Activity of Essential Oils against Japanese Termite

essential oil	concn, mg/filter paper	mortality (%, mean ± SEM, $N = 5$) 2 days	7 days
ajowan	2	100.0 a[a]	100.0 a
	1.5	72.0 ± 4.9 abcde	100.0 a
	1	18.0 ± 5.8 ghi	86.0 ± 2.4 abcd
	0.5	0.0 i	2.0 ± 2.0 i
allspice	2	100.0 a	100.0 a
	1.5	94.0 ± 4.0 a	100.0 a
	1	82.0 ± 3.7 abc	100.0 a
	0.5	20.0 ± 3.2 fghi	88.0 ± 5.8 abcd
	0.25	0.0 i	0.0 i
amyris	2	10.0 ± 5.5 hi	46.0 ± 7.5 bcdefghi
Artemisia afra	2	76.0 ± 7.5 abcd	100.0 a
	1.5	6.0 ± 2.4 hi	72.0 ± 4.9 abcdef
cabreuva	2	10.0 ± 3.2 hi	72.0 ± 6.6 abcdef
cajept extra	2	24.0 ± 7.5 efghi	100.0 a
	1.5	0.0 i	28.0 ± 8.6 fghi
cananga	2	18.0 ± 5.8 ghi	94.0 ± 4.0 ab
	1.5	0.0 i	8.0 ± 3.7 hi
cardamom	2	52.0 ± 10.2 abcdefgh	100.0 a
	1.5	2.0 ± 2.0 i	16.0 ± 4.0 ghi
carrot seed	2	0.0 i	56.0 ± 4.0 abcdefgh
caraway	2	88.0 ± 4.9 ab	100.0 a
	1.5	64.0 ± 2.4 abcdefg	100.0 a
	1	10.0 ± 6.3 hi	100.0 a
	0.5	0.0 i	22.0 ± 5.8 ghi
clementine	2	16.0 ± 5.1 ghi	60.0 ± 3.2 abcdefg
copaiva	2	10.0 ± 3.2 hi	76.0 ± 4.0 abcdef
coriander	2	78.0 ± 5.8 abc	96.0 ± 2.4 a
	1.5	4.0 ± 4.0 hi	42.0 ± 5.8 defghi
davana	2	28.0 ± 3.7 defghi	44.0 ± 2.4 cdefghi
dill	2	88.0 ± 4.9 ab	100.0 a
	1.5	28.0 ± 5.8 defghi	92.0 ± 3.7 abc
	1	0.0 i	76.0 ± 2.4 abcdef
	1.5	0.0 i	4.0 ± 2.4 i
elemi	2	0.0 i	86.0 ± 6.8 abcd
	1.5	0.0 i	32.0 ± 3.7 efghi
fokieniawood	2	2.0 ± 2.0 i	28.0 ± 6.6 fghi
frankincense	2	0.0 i	32.0 ± 3.7 efghi
galbanum	2	2.0 ± 2.0 i	30.0 ± 5.5 efghi
geranium	2	98.0 ± 2.0 a	98.0 ± 2.0 a
	1.5	44.0 ± 6.0 bcdefghi	84.0 ± 5.1 abcd
	1	22.0 ± 3.7 fghi	78.0 ± 5.8 abcde
gurjum	2	6.0 ± 2.4 hi	8.0 ± 2.0 hi
hyssop	2	68.0 ± 3.7 abcdef	100.0 a
	1.5	26.0 ± 6.8 efghi	64.0 ± 4.0 abcdefg
larch	2	38.0 ± 7.3 cdefghi	100.0 a
	1.5	2.0 ± 2.0 i	72.0 ± 8.0 abcdef
lavandin	2	34.0 ± 4.0 cdefghi	94.0 ± 2.4 ab
	1.5	6.0 ± 2.4 hi	30.0 ± 7.1 efghi
litsea	2	100.0 a	100.0 a
	1.5	78.0 ± 5.8 abc	100.0 a
	1	42.0 ± 4.9 bcdefghi	96.0 ± 2.4 a
	0.5	0.0 i	2.0 ± 2.0 i
patchouli	2	2.0 ± 2.0 i	56.0 ± 4.0 abcdefgh
control		0.0 i	0.0 i

[a]Means within a column followed by the same letters are not significantly different ($P = 0.05$, Scheffe test).

in allspice oil. The main components of caraway oil were (*S*)-(+)-carvone (48.7%), (*R*)-(+)-limonene (24.2%), *cis*-carveol (0.4%), and *trans*-carveol (0.3%). The most abundant compound in dill oil was (*S*)-(+)-carvone (35.56%) followed by (*R*)-(+)-limonene (20.21%), myrcene (4.89%), dill ether (4.58%), and *p*-cymene (2.34%). Ratios of the other compounds were < 1%. A total of 17 compounds were identified in geranium oil. Geraniol (10.25%) was the most abundant followed by citronellol (8.63%), 2-phenylethanol (8.03%), geranyl acetate (7.25%), dihydrocitronellol (3.33%), and rose acetate (3.01). The other compounds were present in amounts of < 2%.

Antitermitic Activity of Essential Oil Components. The antitermitic activity of individual compounds 2 days after treatment is shown in **Table 4**. Fumigant toxicities varied according to compounds, application dose, and exposure time. The insecticidal activity of two phenols (thymol and carvacrol) was stronger than that of the alcohol group. The insecticidal activities of verbenol, 2-phenylethanol, nerol, and geraniol were 100, 100, 90, and 76% at 0.25 mg/filter paper, but decreased to 96, 0, 4, and 44% at 0.125 mg/filter paper, respectively. The other alcohol compounds exhibited < 70% mortality at 0.25 mg/filter paper. In a test with phenylpropanoids, the insecticidal activities of eugenol and acetyleugenol was higher than those of methyleugenol, isoeugenol, and methyl isoeugenol. Ketone compounds showed strong insecticidal activity against Japanese termite at 1 mg/filter paper, except for 6-methyl-5-hepten-2-one. The toxicity of (+)-carvone was moderately higher than that of (−)-carvone. 2-Phenylethyl acetate exhibited the strongest insecticidal activity among the acetate group. There was a significant difference in the insecticidal activity of members in the aldehyde group, with neral and geranial being more toxic than citronellal. Compounds belonging to the hydrocarbon group exhibited weak insecticidal activity as compared with the compounds from other functional groups, with toxicity never higher than 50% at 2 mg/filter paper. The antitermitic activities of all individual compounds tested at 7 days after treatment is shown in **Table 5**. Most compounds showed higher insecticidal activity at 7 days than at 2 days. The mortality of some compounds, such as rose acetate, carveol, 6-methyl-5-hepten-2-one, isoeugenol, and methylisoeugenol, demonstrated a significant increase in toxicity over the bioassay duration.

DISCUSSION

Many phytochemicals have been reported to possess antitermitic, antifeedant, or repellent activities against a variety of insects and pests. Reported antitermitic phytochemicals include plumbagin, isodiospyrin, and microphyllone from *Diospyros sylvatica* (*13*), 7-methyljuglone from *Diopyros virginiana* (*14*), and chamaecynone from *Chamaecyparis pisifera* (*15*). Several plant essential oils and essential oil components have been also reported to show antitermitic or repellent activity. Cedrol and α-cadinol from *Taiwania cryptomerioides* heartwood oil (*3*) and eugenol and sulfide compounds from clove and garlic oils (*5*) have exhibited strong insecticidal activity against termites. Other studies have confirmed that certain essential oils, such as those extracted from cedarwood (*16*), *L. cubeba* (*17*), and *Cinnamomum* spp. (*18*), are repellent to termites. In our study, strong insecticidal activity against Japanese termite was achieved with essential oils of ajowan (*T. ammi*), allspice (*P. dioica*), caraway (*C. carvi*), dill (*A. graveolens*), geranium (*P. graveolens*), and litsea (*L. cubeba*).

Various compounds, including alcohols, aldehydes, fatty acid derivatives, terpenoids, and phenolics, exist in plant essential oils. Jointly or independently, they contribute to the insecticidal, antifungal, and nematicidal activities of these oils (*5*, *11*, *19*). Chemical analysis of ajowan, allspice, and litsea has been reported in a previous study (*19*). In this study, we determined the configurations of α-pinene, *β*-pinene, and limonene by using a chiral column. Only (*R*)-(+)-α-pinene was found in ajowan oil, but both of the optical isomers were identified in litsea oil. Ratios of (+)-*β*-pinene and (−)-*β*-pinene were 0.31 and 0.75 in litsea oil, respectively. However, only (+)-*β*-pinene was found in ajowan

Table 3. Chemical Analysis of Active Plant Essential Oils

			retention index (std compd)		composition ratio (%, w/w)					
no.	compound	purity, company[a]	1MS	INNOWax	ajowan	allspice	carvi	dill	geranium	litsea
1	α-pinene		928 (928)[f]	1021 (1021)	0.87			0.35	0.08	1.22
	(*S*)-(-)-α-pinene	95%, T								0.51
	(*R*)-(+)-α-pinene	99%, A			0.87			0.35		0.71
2	camphene	80%, T	940 (940)	1064 (1064)	0.10					0.27
3	sabinene[b]		963 (−)	1128 (−)						1.51
4	6-methyl-5-hepten-2-one	99%, A	964 (964)	1344 (1344)						1.34
5	β-pinene	94%, T	967 (967)	1108 (1108)	1.26					1.06
	(+)-β-pinene				1.26					0.31
	(−)-β-pinene									0.75
6	myrcene	95%, A	981 (981)	1165 (1167)	0.48			0.17		0.73
7	α-phellandrene	98%, T	995 (994)	1165 (1166)				4.89		
8	α-terpinene	85%, F	1007 (1007)	1181 (1181)	0.13					
9	*p*-cymene	95%, F	1012 (1012)	1275 (1275)	24.40			2.34[g]		
10	1,8-cineole	99%, F	1018 (1018)	1209 (1208)	0.32	0.10		0.96[g]		1.21
11	limonene		1020 (1019)	1200 (1200)	0.44	0.18	24.20	20.48		14.64
	(*S*)-(−)-limonene	97%, T			0.08			0.27		0.34
	(*R*)-(+)-limonene	97%, A			0.36	0.18	24.20	20.21		14.30
12	γ-terpinene	97%, F	1050 (1050)	1248 (1248)	27.77					
13	2-phenylethanol	98%, T	1082 (1081)	1929 (1929)					8.03	
14	linalool	98%, W	1084 (1084)	1556 (1557)					1.14	1.38
15	verbenol	95%, A	1121 (1121)	1665 (1665)						0.35
16	isopulegol	99%, A	1127 (1128)	1533 (1538)					0.28	
17	citronellal	85%, A	1130 (1130)	1486 (1486)						0.57
18	terpine-4-ol	97%, A	1159 (1159)	1610 (1611)	0.32					
19	dill ether[b]		1165 (−)	1484 (−)				4.58		
20	*trans*-dihydrocarvone[c]	98%, A	1168 (1170)	1616 (1616)				0.83		
21	α-terpineol	95%, T	1170 (1170)	1707 (1707)					0.56	0.44
22	*cis*-dihydrocarvone[c]	98%, A	1174 (1175)	1744 (1744)				0.86		
23	dihydrocitronellol[b]								3.33	
24	*cis*-carveol[c]	97%, A	1196 (1196)	1848 (1848)			0.40			
25	*trans*-carveol[c]	97%, A	1207 (1207)	1878 (1879)			0.30			
26	citronellol	95%, A	1209 (1211)	1777 (1776)					8.63[g]	
27	nerol	97%, A	1209 (1211)	1811 (1811)					2.17[g]	0.27
28	neral	75%, S[e]	1216 (1216)	1690 (1690)						30.27
29	2-phenylethyl acetate	99%, S	1223 (1225)	1828 (1828)					1.57	
30	carvone		1214 (1213)	1745 (1744)			48.70	35.56		
	(*R*)-(−)-carvone	96%, A								
	(*S*)-(+)-carvone	96%, A					48.70	35.56		
31	geraniol	98%, F	1236 (1236)	1858 (1858)					10.25	0.50
32	linalyl acetate	95%, T	1239 (1240)	1563 (1563)					1.03	
33	geranial	85%, S[e]	1245 (1245)	1742 (1742)						39.23
34	thymol	99%, F	1273 (1273)	2207 (2207)	41.77					
35	carvacrol	95%, T	1278 (1278)	2235 (2236)	0.55					
36	citronellyl acetate	96%, S	1333 (1335)	1668 (1668)					0.74	
37	eugenol	95%, W	1334 (1334)	2186 (2186)		86.44				
38	neryl acetate	99%, S	1341 (1344)	1733 (1734)					1.26	
39	geranyl acetate	97%, F	1360 (1360)	1764 (1764)					7.25	
40	methyleugenol	98%, W	1370 (1370)	2025 (2026)		3.87				
41	diphenyl ether	98%, T	1372 (1370)	2026 (2026)					1.78	
42	β-caryophyllene	90%, T	1414 (1414)	1600 (1600)		7.70			0.25	0.60
43	α-humulene	98%, F	1447 (1447)	1673 (1673)		0.99				
44	rose acetate[d]	99%, S	1522 (1522)	2222 (2223)					3.01	

[a]A, Aldrich; F, Fluka; S, synthesized in our laboratory; T, TCI; W, Wako. [b]Structure was tentatively identified by comparison of mas spectrum in the library. [c]Mixture of *cis*- and *trans*-, *trans*- and *cis*- isomers were determined from the literature report (*10*). [d]IUPAC name of rose acetate is 2,2,2-trichloro-1-phenylethanyl acetate. [e]Previously prepared neral and geranial were used in this experiment (*11*). [f]RI of standard compound. [g]Composition ratio of citronellol and nerol, limonene, and 1,8-cineole were determined on HP-INNOWax column.

oil. The chemical analysis data of caraway, dill, and geranium oils have been well studied (*20*, *21*). Similar results were observed therein, albeit with differences in the ratios and the minor components. Galambosi and Peura (*22*) have reported that the composition of essential oil differs widely with production conditions, such as harvesting date or storage time, as well as climate and soil factors.

The structure−activity relationships (SAR) of plant compounds against insect pests and nematodes have been studied in depth. The SAR of monoterpenoids and fumigant activity against adult *Acanthoscelides obtectus* (Say) was investigated by Regnault-Roger and Hamraoui (*23*). They found that oxygenated structures, especially carvacrol, linalool, and terpineol, exhibited the strongest activity. In our study, the antitermitic activities of verbenol, carvone, thymol, and carvacrol were higher than those of α-pinene and limonene. Verbenol, carvone, thymol, and carvacrol are oxygenated compounds of α-pinene, limonene, and *p*-cymene, respectively. Choi et al. (*24*) reported that phenol,

Table 4. Fumigant Antitermitic Activity of Compounds from Six Active Essential Oils against Japanese Termite at 2 Days after Treatment

	mortality of termite at 2 days after treatment with synthetic compound at each concn[b] (%, mean ± SEM, $N = 5$)							
compound	2	1.5	1	0.5	0.25	0.125	0.062	0.031
hydrocarbons								
camphene	2.0 ± 42.0 f[c]	[d]						
(+)-α-pinene	20.0 ± 4.5 def	0 e						
(−)-α-pinene	40.0 ± 4.5 cd	10.0 ± 3.2 cde						
β-pinene	8.0 ± 5.8 ef							
myrcene	6.0 ± 2.4 f							
α-phellandrene	0 g							
α-terpinene	20.0 ± 3.2 def							
p-cymene	44.0 ± 2.4 cd	6.0 ± 2.4 cde						
(*R*)-(+)-limonene	6.0 ± 4.0 f	4.0 ± 2.4 de	4.0 ± 2.4 c					
(*S*)-(−)-limonene	10.0 ± 4.5 ef	6.0 ± 4.0 cde	4.0 ± 2.4 c					
γ-terpinene	46.0 ± 2.4 c							
α-humulene	32.0 ± 5.8 cde	22.0 ± 4.9 bcd	8.0 ± 3.7 c					
β-caryophyllene	24.0 ± 4.0 cdef	18.0 ± 2.0 cde	2.0 ± 2.0 c					
aldehydes								
citronellal	100 a	100 a	82.0 ± 2.0 a	2.0 ± 2.0 e				
neral	100 a	100 a	100 a	100 a	80.0 ± 5.5 abc	12.0 ± 5.8 cde		
geranial	100 a	100 a	100 a	100 a	92.0 ± 4.9 ab	38.0 ± 3.7 bc	0 b	
acetates								
2-phenylethyl acetate	100 a	100 a	100 a	100 a	96.0 ± 4.0 a	0 e		
linalyl acetate	100 a	100 a	100 a	58.0 ± 3.7 b	24.0 ± 5.1 gh			
citronellyl acetate	100 a	100 a	100 a	78.0 ± 4.9 ab	24.0 ± 5.1 gh			
neryl acetate	100 a	100 a	100 a	100 a	52.0 ± 3.7 cdefg	0 e		
geranyl acetate	100 a	100 a	100 a	100 a	60.0 ± 3.2 bcdef	0 e		
rose acetate	100 a	100 a	100 a	56.0 ± 6.0 bc	42.0 ± 4.9 defg	32.0 ± 11.0 bcd	0 b	
alcohols								
2-phenylethanol	100 a	100 a	100 a	100 a	100 a	0 e		
linalool	100 a	100 a	100 a	56.0 ± 4.0 bc	20.0 ± 7.1 gh			
verbenol	100 a	100 a	100 a	100 a	100 a	96.0 ± 2.4 a	8.0 ± 5.8b	
isopulegol	100 a	100 a	100 a	100 a	32.0 ± 8.0 efgh	2.0 ± 2.0 e		
terpinen-4-ol	100 a	100 a	100 a	30.0 ± 3.2 d	26.0 ± 2.4 fgh	6.0 ± 6.0 de		
α-terpineol	100 a	100 a	100 a	94.0 ± 6.0 a	66.0 ± 4 abcde	4.0 ± 4.0 de		
carveol[a]	100 a	100 a	100 a	100 a	20.0 ± 5.5 gh	14.0 ± 4.0 cde	2.0 ± 2.0 b	2.0 ± 2.0 c
citronellol	100 a	100 a	100 a	0 e	0 h			
nerol	100 a	100 a	100 a	100 a	90.0 ± 4.5 ab	4.0 ± 4.0 de		
geraniol	100 a	100 a	100 a	100 a	76.0 ± 2.4 abcd	44.0 ± 6.0 b	0 b	
ketones								
6-methyl-5-hepten-2-one	32.0 ± 5.8 cde	24.0 ± 6.8 bc	20.0 ± 7.1 b					
dihydrocarvone[a]	100 a	100 a	100 a	66.0 ± 2.4 b	50.0 ± 4.5 cdefg	4.0 ± 2.4 de		
(*R*)-(−)-carvone	100 a	100 a	100 a	100 a	100 a	2.0 ± 2.0 e	0 b	
(*S*)-(+)-carvone	100 a	100 a	100 a	100 a	100 a	44.0 ± 2.4 b	2.0 ± 2.0 b	
phenols								
thymol	100 a	100 a	100 a	100 a	100 a	100 a	100 a	68.0 ± 2.0 a
carvacrol	100 a	100 a	100 a	100 a	100 a	100 a	100 a	12.0 ± 4.9 c
eugenol	100 a	100 a	100 a	100 a	100 a	100 a	100 a	48. 0 ± 4.9 b
methyleugenol	100 a	100 a	100 a	100 a	100 a	100 a	2.0 ± 2.0 b	0 c
isoeugenol	100 a	100 a	100 a	28.0 ± 2.0 d	24.0 ± 2.4 gh	20.0 ± 3.2 bcde	6.0 ± 4.0 b	
methylisoeugenol	100 a	100 a	100 a	34.0 ± 5.1 cd	28.0 ± 7.3 fgh	22.0 ± 5.8 bcde	10.0 ± 3.2 b	2.0 ± 2.0 c
acetyleugenol	100 a	100 a	100 a	100 a	100 a	100 a	84.0 ± 4.0 a	2.0 ± 2.0 c
others								
diphenyl ether	100 a	100 a	100 a	100 a	98.0 ± 2.0 a	92.0 ± 5.8 a	18.0 ± 5.8 b	0 c
1,8-cineole	72.0 ± 3.7b	38.0 ± 3.7b	16.0 ± 6.0c					
control	0 f	0 e	0 c	0 e	0 h	0 e	0 b	0 c

[a]Mixture of *cis*- and *trans*-isomers. [b]Milligrams per filter paper. [c]Means within a column followed by the same letters are not significantly different ($P = 0.05$, Scheffe test). [d]No entry indicates the compound was not tested.

alcohol, and aldehyde compounds were generally more toxic to the pine wood nematode (*Bursaphelenchus xylophilus*) than were compounds from other monoterpenoid groups such as ketones and hydrocarbons. In this study, phenol compounds exhibited the strongest insecticidal activity among the compounds tested, and alcohol and aldehyde groups were more toxic than the hydrocarbon group.

Among the aldehyde and ketone compounds, α,β-unsaturated carbonyl compounds (nerol, geranial, and carvones) were more active than the others at 0.5 mg/filter paper. Lee et al. (*11*) and Kim et al. (*25*) reported that α,β-unsaturated carbonyl compounds were responsible for antifungal and nematicidal activities, respectively. These results suggested that the double bond at the α,β position in carbonyl compounds enhanced insecticidal activity.

Lee at al. (*11*) and Choi et al. (*24*) reported that primary alcohols were more active than secondary and tertiary alcohols in antifungal and nematicidal activities, respectively. However, such

Table 5. Fumigant Antitermitic Activity of Compounds from Six Active Essential Oils against Japanese Termite at 7 Days after Treatment

	mortality of termite at 2 days after treatment with synthetic compound at each concn[b] (%, mean ± SEM, $N = 5$)								
compound	2	1.5	1	0.5	0.25	0.125	0.062	0.031	0.015
hydrocarbons									
camphene	8.0 ± 5.8 ef[c]	[d]							
(+)-α-pinene	80.0 ± 3.2 ab	16.0 ± 5.1 c							
(-)-α-pinene	100 a	22.0 ± 4.9 c							
β-pinene	30.0 ± 3.2 de								
myrcene	30.0 ± 9.5 de								
α -phellandrene	100 a	54.0 ± 2.4 b							
α-terpinene	44.0 ± 2.4 cd								
p-cymene	84.0 ± 2.4 ab	10.0 ± 10.0 c							
(*R*)-(+)-limonene	100 a	76.0 ± 5.1 ab	4.0 ± 2.4 f						
(*S*)-(−)-limonene	100 a	92.0 ± 4.9 a	64.0 ± 2.4 b						
γ-terpinene	64.0 ± 2.4 bc								
α-humulene	88.0 ± 2.0 a	84.0 ± 2.4 a	44.0 ± 6.0 c						
β-caryophyllene	94.0 ± 4.0 a	90.0 ± 4.5 a	10.0 ± 4.5 f						
aldehydes									
citronellal	100 a	100 a	100 a	14.0 ± 6.0 b					
neral	100 a	100 a	100 a	100 a	82.0 ± 4.9 a	34.0 ± 2.4 cd			
geranial	100 a	100 a	100 a	100 a	100 a	94.0 ± 4.0 a	4.0 ± 2.4 e		
acetates									
2-phenylethyl acetate	100 a	100 a	100 a	100 a	100 a	2.0 ± 2.0 cd			
linalyl acetate	100 a	100 a	100 a	100 a	30.0 ± 4.5b				
citronellyl acetate	100 a	100 a	100 a	100 a	30.0 ± 4.5 b				
neryl acetate	100 a	100 a	100 a	100 a	88.0 ± 4.9 a	4.0 ± 2.4 cd			
geranyl acetate	100 a	100 a	100 a	100 a	100 a	2.0 ± 2.0 cd			
rose acetate	100 a	100 a	100 a	100 a	100 a	100 a	100 a	92.0 ± 4.9 ab	40.0 ± 4.5 a
alcohols									
2-phenylethanol	100 a	100 a	100 a	100 a	100a	4.0 ± 2.4 cd			
linalool	100 a	100 a	100 a	100 a	10.0 ± 4.4b				
verbenol	100 a	100 a	100 a	100a	100a	100a	22.0 ± 5.8de		
isopulegol	100 a	100 a	100 a	100a	98.0 ± 2.0a	10.0 ± 6.3 cd			
terpinen-4-ol	100 a	100 a	100 a	88.0 ± 4.9 a	84.0 ± 2.4a	10.0 ± 7.7 cd			
α-terpineol	100 a	100 a	100 a	100 a	82.0 ± 2.0a	22.0 ± 3.7 cd			
carveol[a]	100 a	100 a	100 a	100 a	100a	100a	68.0 ± 4.9 cb	28.0 ± 9.2 de	
citronellol	100 a	100 a	100 a	100 a	30.0 ± 8.9 b				
nerol	100 a	100 a	100 a	100 a	100 a	8.0 ± 8.0 cd			
geraniol	100 a	100 a	100 a	100 a	100 a	80.0 ± 20.0 ab	4.0 ± 4.0 e		
ketones									
6-methyl-5-hepten-2-one	100 a	100 a	22.0 ± 5.8 de						
dihydrocarvone[a]	100 a	100 a	100 a	100 a	100 a	46.0 ± 4.0 bc			
(*R*)-(−)-carvone	100 a	100 a	100 a	100 a	100 a	80.0 ± 3.2 ab	40.0 ± 3.2 d		
(*S*)-(+)-carvone	100 a	100 a	100 a	100 a	100 a	100 a	44.0 ± 4.0 cd		
phenols									
thymol	100 a	100 a	100 a	100 a	100 a	100 a	100 a	98.0 ± 2.0 a	10.0 ± 6.3 b
carvacrol	100 a	100 a	100 a	100 a	100 a	100 a	100 a	68.0 ± 3.7 bc	
eugenol	100 a	100 a	100 a	100 a	100 a	100 a	100 a	96.0 ± 4.0 ab	0 b
methyleugenol	100 a	100 a	100 a	100 a	100 a	100 a	100 a	20.0 ± 3.1 de	
isoeugenol	100 a	100 a	100 a	100 a	100 a	94.0 ± 2.4 a	20.0 ± 7.1 de		
methylisoeugenol	100 a	100 a	100 a	100 a	100 a	100 a	76.0 ± 2.4 ab	8.0 ± 2.0 e	
acetyleugenol	100 a	100 a	100 a	100 a	100 a	100 a	100 a	84.0 ± 6.8 ab	2.0 ± 2.0 b
others									
diphenyl ether	100 a	100 a	100 a	100 a	100 a	100 a	72.0 ± 6.6 b	40.0 ± 3.2 cd	
1,8-cineole	100 a	92.0 ± 4.9 a	38.0 ± 5.8 cd						
control	0 f	0 c	0 f	0 c	0 c	0 d	0 e	0 e	0 b

[a]Mixture of *cis*- and *trans*-isomers. [b]Milligrams per filter paper. [c]Means within a column followed by the same letters are not significantly different ($P = 0.05$, Scheffe test). [d]No entry indicates that the compound was not tested.

a tendency was not clearly found in this study although alcohol compounds showed a generally higher activity. This might indicate that the position of the hydroxy group in alcohol is not crucial for antitermitic activity.

There was a significant difference in the insecticidal activity among phenylpropanoid compounds. Park et al. (*19*) and Kim et al. (*26*) reported that the functional group at the C1 position of the benzene ring or the position of the double bond of the propenyl group was important in nematicidal or antifungal activity. There was a significant difference in the nematicidal or insecticidal activity of *cis*−*trans* diastereomers such as *cis*- and *trans*-asarone (*25*, *27*). Nematicidal or insecticidal activities of *cis*-asarone were stronger than those of *trans*-asarone. However, significant difference in antitermitic activity was not observed in enantiomers such as (*R*)-(+)-limonene, (*S*)-(−)-limonene, (*R*)-(+)-carvone, and (*S*)-(−)-carvone.

Although little is known regarding the physiological actions of essential oils on insects, various oils or their components cause

symptoms that suggest a neurotoxic mode of action (*28*, *29*). Eugenol has been claimed to exert its insecticidal activity by binding to octopamine receptors (*30*, *31*). Price and Berry (*32*) insisted that geraniol and citral showed greater similarities to octopamine than did eugenol. Elucidating the mode of action of the lethality of these oils toward insect pests is important for the safety of humans and other veterbrates (*32*). Roeder (*33*) and Enan (*30*) have noted the facts that vertebrates possess very few octopamine receptors and that specific octopamine receptor binding might not cause any adverse effect in vertebrates exposed to the oils. However, the exact mode of action of essential oils and constituents remains unclear.

Our results indicate that ajowan, allspice, caraway, dill, geranium, and litsea and their components could be developed as fumigants against Japanese termites. For the practical use of these oils and their constituents as novel fumigants, the safety of oils and their constituents in humans and nontarget organisms needs to be further evaluated; moreover, the development of formulations with improved efficacy and stability and reduced costs necessitates further studies.

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