



# Progress in Essential Oils

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## Thyme Oil

Using a combination of TLC and IR as his method of analysis, Popesco (1983) found that a thyme oil (ex. *Thymus vulgaris*) of Romanian origin tentatively contained the following constituents:

$\alpha$ -pinene	p-cymene
$\beta$ -pinene	bornyl acetate
limonene	borneol
1,8-cineole	thymol (31.5-36.0%)

A thyme oil produced in a laboratory from wild plants of *T. vulgaris* was screened for its antimicrobial properties by Panizzi et al. (1993). In addition to confirming the strong antimicrobial properties of the oil, they found that it contained the following components:

$\alpha$ -pinene (0.92%)	linalool (0.44%)
camphene (1.11%)	bornyl acetate (2.34%)
$\beta$ -pinene (0.22%)	$\beta$ -caryophyllene (0.38%)
myrcene (1.71%)	$\alpha$ -terpineol (1.26%)
limonene (0.33%)	borneol (1.35%)
1,8-cineole (0.23%)	geraniol (0.39%)
$\gamma$ -terpinene (11.07%)	thymol (31.39%)
p-cymene (17.01%)	carvacrol (12.36%)
camphor (0.13%)	

An oil of *T. zygis* produced from plants collected in the wild in Morocco was analyzed by GC and GC/MS by Tantaoui-Elaraki et al. (1993). The oil, which because of its composition could not have any economic value, was found to contain the following components:

$\alpha$ -pinene (5.2%)	<i>cis</i> -linalool oxide† (0.5%)
camphene (5.4%)	terpinolene (0.1%)
$\beta$ -pinene (0.7%)	linalool (4.8%)
3-octanol (1.7%)	borneol (5.9%)
myrcene (1.7%)	dihydrocarvone* (t)
$\alpha$ -terpinene (3.9%)	linalyl acetate (0.7%)
p-cymene (50.6%)	methyl carvacrol (0.1%)
$\gamma$ -terpinene (0.2%)	bornyl acetate (0.2%)
<i>cis</i> -sabinene hydrate (0.2%)	thymol (5.0%)
	carvacrol (8.1%)

t = trace (<0.1%)

\* correct isomer not identified

† furanoid form

Pino et al. (1997) used GC/MS to analyze an oil of *T. vulgaris* produced from plants grown in Cuba. The oil was found to contain:

$\alpha$ -thujene (2.97%)	methyl thymol (0.10%)
camphene (0.79%)	$\beta$ -caryophyllene (0.22%)
$\beta$ -pinene (0.60%)	terpinen-4-ol (0.35%)
myrcene (4.00%)	2-undecanone (t)
$\alpha$ -terpinene (0.10%)	borneol (1.14%)
limonene (0.25%)	$\gamma$ -elemene (0.35%)
1,8-cineole (1.94%)	$\delta$ -cadinene (0.85%)
$\gamma$ -terpinene (17.61%)	calamenene* (0.10%)
p-cymene (17.65%)	thymyl acetate (0.29%)
1-octen-3-ol (1.00%)	caryophyllene oxide (0.55%)
<i>trans</i> -sabinene hydrate (0.84%)	thymol (36.60%)
linalool (3.28%)	carvacrol (6.55%)

\*correct isomer not identified

t = trace (<0.01%)

An oil of *T. vulgaris* produced from plants grown in Cameroon was subjected to analysis by Amvan Zollo et al. (1998), and was found to possess the following composition:

$\alpha$ -thujene (1.2%)	thymol (27.2%)
$\alpha$ -pinene (1.0%)	carvacrol (3.3%)
camphene (1.2%)	$\alpha$ -copaene (0.1%)
sabinene (0.4%)	$\beta$ -caryophyllene (3.5%)
$\beta$ -pinene (0.3%)	$\beta$ -gurjunene (0.1%)
myrcene (1.7%)	(E)- $\beta$ -farnesene (0.1%)
$\alpha$ -phellandrene (0.2%)	$\alpha$ -humulene (t)
p-cymene (23.6%)	ar-curcumenene (0.1%)
limonene (1.5%)	germacrene D (0.6%)
$\gamma$ -terpinene (22.7%)	(E,E)- $\alpha$ -farnesene (0.1%)
terpinolene (t)	$\delta$ -cadinene (0.3%)
linalool (5.2%)	caryophyllene oxide (0.6%)
camphor (1.9%)	$\alpha$ -muurolol (0.2%)
terpinen-4-ol (1.3%)	$\alpha$ -cadinol (0.2%)
$\alpha$ -terpineol (0.3%)	

t = trace (<0.1%)

**Comparative percentage composition of an oil and supercritical fluid extract of *Thymus zygis***

**T-1**

Compound	Oil	Supercritical Fluid Extract
$\alpha$ -thujene	1.0	0.5
$\alpha$ -pinene	1.0	0.4
camphene	1.4	1.4
sabinene	0.3	0.1
$\beta$ -pinene	0.3	0.3
myrcene	1.4	0.8
$\alpha$ -phellandrene	1.2	0.1
$\alpha$ -terpinene	0.1	0.2
limonene	1.6	0.7
p-cymene	13.6	15.0
1,8-cineole	0.6	0.1
$\gamma$ -terpinene	13.6	7.0
terpinen-1-ol	0.4	0.3
terpinolene	0.1	-
linalool	2.4	2.4
camphor	0.3	0.3
borneol	2.1	3.0
terpinen-4-ol	0.1	0.2
nerol	0.2	0.1
neral	0.3	0.6
geraniol	12.6	14.0
linalyl acetate	1.1	0.1
bornyl acetate	0.1	0.2
thymol	20.6	11.0
carvacrol	1.3	1.8
geranyl acetate	16.3	17.5
$\beta$ -caryophyllene	3.5	2.0

Also in 1998, Moldao-Martins et al. compared the composition of a water-distilled oil with of a supercritical fluid extract of *T. zygis* ssp. *sylvestris* plants grown in Portugal. A summary of the results of this study can be seen in T-1.

Theuerl et al. (1999) compared the method of GC analysis used to perform the quantitative analysis of thyme oil. They compared conventional temperature programmed GC (which took ca. 40 min) with a temperature programmed fast GC method (which took less than 5 min). Their results can be seen in T-2.

Also in 1999, Lechamo et al. analyzed the oils of five cultivars of *T. vulgaris* grown in Quebec (Canada). A summary of the results of this study can be seen in T-3.

A German commercial sample of thyme oil, presumably of Spanish origin, was analyzed using both GC and GC/MS by Reichling et al. (1999). They found that the oil contained the following components:

**Comparative percentage composition of thyme oil analyzed by conventional and fast GC**

**T-2**

Compound	Conventional GC	Fast GC
$\alpha$ -pinene	0.67	0.72
$\alpha$ -thujene	0.66	0.61
camphene	0.47	0.49
$\beta$ -pinene	0.24	0.26
myrcene	3.08	2.89
$\alpha$ -terpinene	2.91	2.90
$\gamma$ -terpinene	29.76	29.65
p-cymene	22.66	24.10
terpinolene	0.26	0.24
camphor	0.60	0.62
linalool	2.63	2.63
bornyl acetate	0.31	0.31
$\beta$ -caryophyllene	7.95	6.66
terpinen-4-ol	0.07	0.04
$\alpha$ -humulene	0.06	0.07
$\alpha$ -terpineol	0.09	0.11
borneol	0.85	0.85
thymol	17.99	18.79
carvacrol	0.77	0.98

$\alpha$ -pinene (3.05%)	camphor (0.16%)
camphene (1.75%)	borneol (1.14%)
$\beta$ -pinene (0.43%)	terpinen-4-ol (0.15%)
myrcene (0.66%)	$\alpha$ -terpineol (7.63%)
p-cymene (34.20%)	$\gamma$ -terpineol <sup>†</sup> (2.22%)
limonene (1.05%)	thymol (38.55%)
ocimene* (0.17%)	longifolene (0.32%)
terpinolene (0.15%)	$\beta$ -caryophyllene (1.15%)
linalool (4.60%)	$\alpha$ -humulene (0.06%)

\*correct isomer not identified

<sup>†</sup> $\gamma$ -terpineol has not been unequivocally proven to occur naturally, so identity is in question

Another commercial sample of *T. vulgaris* oil that was purchased in Italy was analyzed by GC and GC/MS by Juliano et al. (2000). A summary of its composition is as follows:

$\alpha$ -pinene (1.1%)	3-nonanol (0.2%)
6-methyl-5-hepten-2-one (0.3%)	linalool (1.7%)
camphene (1.2%)	camphor (0.2%)
myrcene (0.7%)	$\alpha$ -campholenic aldehyde (0.1%)
3-octanol (1.0%)	borneol (0.2%)
$\alpha$ -terpinene (0.2%)	linalyl propionate (2.7%)
p-cymene (7.0%)	thymol (27.6%)
limonene (24.2%)	carvacrol (20.5%)
1,8-cineole (0.2%)	
$\gamma$ -terpinene (0.3%)	
cis-sabinene hydrate (0.3%)	

A lab-distilled oil of *T. vulgaris* purchased in the UK was subjected to GC/MS analysis by Dorman et al. (2000). This oil was found to possess the following composition:

tricyclene (0.44%)	terpinolene (0.49%)
$\alpha$ -thujene (0.14%)	linalool (4.56%)

**Comparative percentage composition of the oils of five cultivars of *Thymus vulgaris* grown in Quebec**

**T-3**

Compound	1	2	3	4	5
$\alpha$ -thujene	0.79	0.82	-	0.84	0.88
$\alpha$ -pinene	1.38	1.03	2.53	1.13	0.87
camphene	1.12	1.24	-	1.24	0.42
$\beta$ -pinene	-	-	2.18	0.98	0.56
sabinene	1.56	1.77	0.24	1.44	-
myrcene	0.70	1.03	-	0.90	0.67
$\alpha$ -terpinene	0.69	1.23	2.63	1.02	1.02
limonene	-	4.00	0.38	-	1.62
1,8-cineole	0.34	-	0.26	-	-
p-cymene	20.21	17.03	26.89	17.45	10.27
$\gamma$ -terpinene	3.06	8.62	2.21	6.44	5.42
camphor	-	0.87	0.88	0.21	-
linalool	3.52	3.57	4.76	8.32	3.89
linalyl acetate	-	2.00	4.06	0.55	-
terpinen-4-ol	3.86	0.52	1.08	0.65	0.21
$\beta$ -caryophyllene	2.01	2.30	-	2.55	0.92
lavandulol	-	0.23	2.58	-	-
borneol	3.69	3.39	0.56	2.53	3.20
geraniol	0.67	1.24	3.48	0.59	0.63
bornyl acetate	-	0.23	0.95	0.17	0.62
thymol	38.75	45.99	10.38	51.52	66.94
carvacrol	16.60	2.62	39.85	0.30	0.98

$\alpha$ -pinene (2.12%)  
camphene (0.17%)  
 $\beta$ -pinene (0.10%)  
myrcene (1.22%)  
 $\alpha$ -terpinene (0.44%)  
p-cymene (25.50%)  
limonene (1.14%)  
 $\gamma$ -terpinene (4.44%)

\*correct isomer not identified

borneol (0.86%)  
 $\beta$ -terpineol\* (0.28%)  
terpinen-4-ol (0.23%)  
 $\alpha$ -terpineol (0.94%)  
bornyl acetate (1.91%)  
bornyl acetate (1.91%)  
carvacrol (2.95%)  
 $\beta$ -caryophyllene (0.98%)

1,8-cineole (0.9%)  
 $\gamma$ -terpinene (14.9%)  
p-cymene (10.0%)  
terpinolene (0.1%)  
hexanal (0.1%)  
3-octanol (0.2%)  
1-octen-3-ol (1.0%)  
*trans*-sabinene hydrate (1.1%)  
camphor (1.0%)  
linalool (2.3%)  
*cis*-sabinene hydrate (0.4%)  
bornyl acetate (0.2%)  
 $\beta$ -caryophyllene (2.3%)

\*correct isomer not identified

t = trace (<0.1%)

carvone (0.2%)  
 $\delta$ -cadinene (0.2%)  
(E)-anethole (0.1%)  
geraniol (0.2%)  
caryophyllene oxide (0.4%)  
methyl eugenol (0.1%)  
cubenol (t)  
spathulenol (0.1%)  
eugenol (0.2%)  
T-cadinol (0.2%)  
thymol (51.1%)  
carvacrol (3.6%)  
 $\alpha$ -cadinol (0.2%)

An oil, a methylene chloride extract and a number of supercritical fluid (SCF) extracts of *T. vulgaris* produced at 40°C with a flow rate of CO<sub>2</sub> of 97.725 L/h and a range of pressures from 80-400 bar were analyzed by Zekovic et al. (2000). The results of this study can be found in T-4. As can be seen, the authors did not find any p-cymene or  $\gamma$ -terpinene in either the oil or the extracts. Also, the characterization of menthone and menthol seems highly questionable.

Thyme oil produced from *T. vulgaris* plants grown in Mongolia were analyzed by Shatar and Altantsetseg (2000) using a combination of GC and GC/MS. The authors found that the oil possessed the following composition.

$\alpha$ -pinene (0.4%)  
camphene (0.2%)  
 $\beta$ -pinene (0.1%)  
myrcene (1.0%)  
 $\alpha$ -phellandrene (0.1%)  
 $\alpha$ -terpinene (1.2%)  
limonene (0.4%)  
 $\beta$ -phellandrene (0.1%)

terpinen-4-ol (0.2%)  
dihydrocarvone\* (0.1%)  
 $\beta$ -farnesene\* (0.2%)  
methyl chavicol (0.1%)  
 $\alpha$ -humulene (0.1%)  
 $\alpha$ -terpineol (0.3%)  
borneol (1.1%)  
geraniol (0.1%)

As part of a screening study for the fungitoxicity of some oils produced from Greek aromatic plants, Dalferera et al. (2000) determined (by GC/MS) that the main constituents of thyme ex. *T. vulgaris* oil were:

$\alpha$ -pinene (0.3%)  
 $\alpha$ -terpinene (1.0%)  
p-cymene (23.5%)  
 $\gamma$ -terpinene (4.3%)  
borneol (1.4%)

terpinen-4-ol (0.6%)  
carvacrol (2.2%)  
thymol (63.6%)  
 $\beta$ -caryophyllene (1.3%)

More recently, Echeverrigaray et al. (2001) compared the composition of oils produced from *T. vulgaris* of various

Compound	Oil	CH <sub>2</sub> Cl <sub>2</sub> Extract	SFC Extracts		
			100 bar	200 bar	400 bar
menthone	t	-	-	-	-
camphor	2.94	0.74	0.49	0.63	0.65
borneol	t	-	-	-	-
menthol	8.14	1.31	2.84	1.90	1.31
2,6-dimethylundecane	t	-	-	-	-
methyl thymol	t	-	-	-	-
dodecane	3.02	1.96	1.83	2.08	2.15
methyl carvacrol	t	-	-	-	-
thymol	50.06	16.11	37.29	22.31	20.01
carvacrol	1.15	0.36	0.86	0.49	0.36
tetradecane	16.11	47.18	26.56	37.54	38.92
β-caryophyllene	0.72	1.14	1.38	0.95	0.98
2-methylundecane	0.62	1.33	0.66	1.14	1.06
pentadecane	2.59	6.50	3.76	5.30	5.25
hexadecane	3.18	5.49	3.35	4.69	4.64
heptadecane	2.98	4.06	2.37	3.33	3.29
octadecane	2.02	2.08	1.27	1.76	1.74
phytol	2.90	-	1.39	-	0.38
t = trace (<0.01%)					
SFC = supercritical fluid CO <sub>2</sub>					

commercial seed sources using GC/MS. As a result of this study, the authors found that the oils varied as follows:

α-thujene (1.35-1.97%)  
 α-pinene (0.78-0.92%)  
 camphene (0.36-0.74%)  
 β-pinene (0.20-0.26%)  
 myrcene (1.63-1.92%)  
 α-phellandrene (0.20-0.26%)  
 α-terpinene (2.06-2.58%)  
 p-cymene (11.68-13.99%)  
 1,8-cineole (0.23-0.41%)  
 γ-terpinene (11.47-22.28%)  
 cis-sabinene hydrate (0.68-1.21%)  
 linalool (1.61-2.55%)  
 borneol (0.68-1.29%)  
 terpinen-4-ol (0.47-0.73%)  
 thymol (41.96-50.47%)  
 carvacrol (0-2.64%)  
 thymyl acetate (0-0.07%)  
 β-caryophyllene (0.59-0.87%)  
 germacrene D (0.17-0.36%)

H. Popesco, *The chemical composition of some essential oils from plants of Romanian provenance*. Clujul Medical, 56(2), 168-170 (1983).

L. Panizzi, G. Flamini, P. L. Cioni and I. Morelli, *Composition and antimicrobial properties of essential oils of four Mediterranean Lamiaceae*. J. Ethnopharm., 39, 167-170 (1993).

A. Tantaoui-Elaraki, N. Lattaoui, A. Errifi and B. Benjilali, *Composition and antimicrobial activity of the essential oils of Thymus broussonettii, T. zygis and T. satureioides*. J. Essent. Oil Res., 5, 45-53 (1993).

J. A. Pino, M. Estarron and V. Fuentes, *Essential oil of Thyme (Thymus vulgaris L.) grown in Cuba*. J. Essent. Oil Res., 9, 609-610 (1997).

P. H. Amvam Zollo, L. Biyiti, F. Tchoumboungang, C. Menut, G. Lamaty and Ph. Bouchet, *Aromatic plants of Tropical Central Africa. Part XXXII. Chemical composition and antifungal activity of thirteen essential oils from aromatic plants of Cameroon*. Flav. Fragr. J., 13, 107-114 (1998).

M. Moldao-Martins, R. Trigo, M. A. Nolasco, M. G. Bernardo Gil and M. L. Beirao de Costa, *Influence of extraction procedure on the aroma composition of Thymus zygis and Mentha pulegium*. In: *Food Flavors: Formation, Analysis and Packaging Influences*. Edits., E. T. Conti, C-T. Ho, C. J. Mussinan, T. H. Pailment, F. Shahidi and A. M. Spainer, p. 133-141, Elsevier Science B. V., Amsterdam (1998).

T. Theuerl, R. Blanke and W. Kleiböhmer, *Schnelle Gaschromatographie durch widerstandsbeheizte Kapillarsäulen. Analyse von ätherischen Ölen aus Gewürzpflanzen*. GITSpeziale-Separation (1), 40-43 (1999).

W. Letchamo, A. Gosselin, J. Hoelzl and R. Marquard, *The selection of Thymus vulgaris cultivars to grow in Canada*. J. Essent. Oil Res., 11, 337-342 (1999).

J. Reichling, M. Harkenthal and R. Saller, *Wirkung ausgewählter ätherischer Öle*. Erfahrungsheilkunde, 6, 357-366 (1999).

C. Juliano, A. Mattana and M. Usai, *Composition and in vitro antimicrobial activity of the essential oil of Thymus herbarum Loisel growing wild in Sardinia*. J. Essent. Oil Res., 12, 516-522 (2000).

H. J. D. Dorman, P. Surai and S. G. Deans, *In vitro antioxidant activity of a number of plant essential oils and phytoconstituents*. J. Essent. Oil Res., 12, 241-248 (2000).

Z. Zekovic, Z. Lepojevic and Dj. Vajic, *Supercritical extraction of Thyme (Thymus vulgaris L.)*. Chromatographia, 51, 175-179 (2000).

C. Juliano, A. Mattana and M. Usai, *Composition and in vitro antimicrobial activity of the essential oil of Thymus herbarum*

*Loisel growing wild in Sardinia*. J. Essent. Oil Res., 12, 516-522 (2000).

S. Shatar and S. Altantsetseg, *Essential oil composition of some plants cultivated in Mongolian climate*. J. Essent. Oil Res., 12, 745-750 (2000).

D. J. Daferera, B. N. Ziogas and M. G. Polissiou, *GC-MS analysis of essential oils from some Greek aromatic plants and their fungitoxicity on Penicillium digitatum*. J. Agric. Food Chem., 48, 2576-2581 (2000).

S. Echeverrigaray, G. Agostini, L. Atti-Serfini, N. Paroul, G. F. Pauletti and A. C. Atti dos Santos, *Correlation between the chemical and genetic relationships among commercial thyme cultivars*. J. Agric. Food Chem., 49, 4220-4223 (2001).

## Origanum oil

In 1993, Vokou et al. sampled *Origanum vulgare* ssp. *hirtum* from across Greece. Oils produced from each of the collections were analyzed by GC and GC/MS. The authors found that of the 23 collections, the oils of 11 were found to be carvacrol-rich, and four were found to be thymol-rich, while the remaining eight contained both thymol and carvacrol in amounts greater than 10% (see T-5.). Furthermore, the authors found that the sum of the four major constituents ( $\gamma$ -terpinene, p-cymene, thymol and carvacrol) were affected by climate because higher concentrations were found in collections from hotter climates; also, the lower the altitude of the collection site, the higher the total oil content.

Melegari et al. (1995) examined the oils of *O. vulgare* L. ssp. *hirtum* (Link) Ietswaart from different areas of cultivation in Italy. Their results can be seen in T-6. From these results, it is readily noticeable that the Grecia biotype grown at Policoro produced a most desirable carvacrol-rich oil of *O. vulgare* ssp. *hirtum* while, other than the biotype grown at Biotonto, which possessed an oil with both carvacrol and thymol, the other biotypes were all less desirable chemotypes because of their paucity in thymol. In the same report, Melegari et al. found that two oils of *O. vulgare* ssp. *glandulosum* (Desfontaines) Ietswaart were both rich in carvacrol as shown below:

sabinene (t)	terpinolene (t)
myrcene (0.61-0.85%)	terpinen-4-ol (t)
$\alpha$ -terpinene (0.48-1.06%)	thymol (0.12-5.32%)
p-cymene (5.11-5.40%)	carvacrol (79.27-83.81%)
limonene (t-0.10%)	$\beta$ -caryophyllene (0.63-0.80%)
1,8-cineole (t-0.15%)	$\alpha$ -humulene (t-0.10%)
$\gamma$ -terpinene (5.70-7.09%)	$\gamma$ -elemene† (t-0.17%)

† incorrect identification based on elution order

t = trace (<0.1%)

Also in 1995, Bourrel et al. reported that an oil of origanum contained the following constituents:

$\alpha$ -thujene (0.1%)	linalool (5.3%)
$\alpha$ -pinene (0.4%)	borneol (1.3%)
camphene (0.2%)	terpinen-4-ol (0.8%)
$\beta$ -pinene (0.1%)	methyl thymol (0.4%)
myrcene (0.6%)	thymol (2.8%)
$\alpha$ -terpinene (0.5%)	carvacrol (71.7%)
p-cymene (8.1%)	$\beta$ -caryophyllene (0.5%)

1,8-cineole (0.6%)	acoradiene* (0.8%)
$\gamma$ -terpinene (2.5%)	$\delta$ -cadinene (0.1%)

\*correct isomer not identified

These data were obtained as a result of a GC/MS analysis. Zhu et al. (1995) analyzed an oil of *O. vulgare* produced in Wenchuan (Sichuan, China) using GC/MS. They found that the oil contained the following constituents:

2-hexenal* (0.16%)	terpinen-4-ol (2.65%)
3-heptanone (0.10%)	p-cymen-8-ol (4.42%)
7-hepten-4-ol <sup>o</sup> (1.89%)	$\alpha$ -terpineol (0.40%)
3-octanone (2.21%)	myrtenol (0.26%)
3-octanol (0.24%)	methyl carvacrol (0.13%)
$\alpha$ -terpinene (0.10%)	cuminaldehyde (0.76%)
p-cymene (11.14%)	thymoquinone (1.73%)
1,8-cineole (0.25%)	carvacrol (38.95%)
$\gamma$ -terpinene (0.28%)	thymol (8.50%)
<i>trans</i> -sabinene hydrate (0.88%)	$\beta$ -caryophyllene (0.29%)
<i>cis</i> -linalool oxide† (0.10%)	thymohydroquinone (0.80%)
<i>trans</i> -linalool oxide† (0.47%)	tetradecanol (0.34%)
linalool (1.22%)	$\alpha$ -farnesene* (1.12%)
<i>cis</i> -pinene hydrate† (0.28%)	nerolidol* (0.12%)
<i>trans</i> -pinocarveol (0.46%)	spathulenol (1.58%)
<i>trans</i> -verbenol (0.47%)	caryophyllene oxide (3.01%)
borneol (1.75%)	palmitic acid (0.62%)

\* correct isomer not identified

† furanoid form

‡ probably misidentification for *cis*-p-menth-2-en-1-ol

<sup>o</sup> corroboration for the identification of this unusual component is needed

Sivropoulou et al. (1996) compared the composition of an oil of *O. vulgare* ssp. *hirtum* with that of a commercial oregano oil available in Greece. The results of the GC/MS study can be found in T-7. As can be seen, the oil of *O. vulgare* ssp. *hirtum* studied was considered to be much more useful as an oregano oil rather than the so-called commercial oil which was too rich in p-cymene as well as containing thymol as the major phenol rather than the desired carvacrol.

Oils of *O. vulgare* ssp. *hirtum* produced from plants collected in late autumn from six different localities in three distinct regions in Greece were analyzed for major constituents by Kokkini et al. (1997). These major constituents were found to vary as follows:

$\gamma$ -terpinene (0.6-3.6%)	thymol (0.2-42.8%)
p-cymene (17.3-51.3%)	carvacrol (1.7-69.6%)

Furthermore, the authors found that oils of plants collected in the north of Greece were found to be thymol-rich,



Comparative percentage composition of *Origanum vulgare* ssp. *hirtum* oils of Greek origin

T-5

Compound	Carvacrol-Rich Oils (11) <sup>a</sup>	Thymol-Rich Oils (4)	Mixed Oils (8)
α-thujene	0-1.0	0-1.1	0-1.7
α-pinene	0-1.0	0-0.6	0-1.0
camphene	0-0.1	-	0-0.5
1-octen-3-ol	0-2.0	0.6-0.8	0.3-0.9
3-octanol	0-0.2	0-0.4	0.1-0.3
myrcene	0-2.2	0-2.2	0-2.6
α-phellandrene	0-0.2	0-0.1	0-0.2
α-terpinene	0-1.6	0-0.9	0.1-2.5
1,8-cineole	-	0-4.4	-
limonene + β-phellandrene + (Z)-β-ocimene	0-0.5	0-0.2	0-0.8
γ-terpinene	0-10.5	0.6-16.4	1.8-16.0
p-cymene	2.2-12.7	3.2-15.8	2.8-13.9
trans-sabinene hydrate	0.2-1.2	0.4-0.6	0.1-1.3
terpinolene	-	-	0-0.2
cis-sabinene hydrate	0.2-1.4	0.1-0.4	0-0.4
borneol	0-1.0	0-1.2	0-1.6
naphthalene	-	0-0.2	0-0.4
terpinen-4-ol	0.1-1.3	0-0.8	0-0.7
α-terpineol	0-0.1	0-1.2	0-0.2
cis-piperitone oxide	0-0.6	-	-
methyl thymol	0-2.9	0-2.4	0-3.5
trans-piperitone oxide	-	-	0-1.2
piperitenone oxide	-	-	0-0.3
thymyl acetate	0-0.4	-	-
thymol	0-9.5	52.2-90.2	27.1-55.1
carvacrol	74.2-93.8	2.3-5.9	11.9-62.9
β-caryophyllene	0-2.2	0-0.9	0-2.0
α-humulene	-	0-0.3	0-0.2
farnesene*	0-2.4	0-0.3	0-0.8
δ-cadinene	0-3.3	-	-
spathulenol	0-0.4	-	-
caryophyllene oxide	0-1.7	0-1.3	0-0.9

\* correct isomer not identified  
<sup>a</sup> = number of oil accessions

whereas those oils from plants collected in the south of Greece were carvacrol-rich.

In 1998, Adam et al. used a combination of GC and GC/MS to analyze an oil of *O. vulgare* ssp. *hirtum* produced from plant material collected in central Greece. The constituents found in this oil were:

α-thujene (0.04%)	trans-sabinene hydrate (0.38%)
α-pinene (0.65%)	
β-pinene (0.98%)	cis-sabinene hydrate (0.53%)
sabinene (0.11%)	
myrcene (0.12%)	linalool (0.18%)
α-phellandrene (0.06%)	terpinen-4-ol (0.03%)
α-terpinene (1.63%)	β-caryophyllene (0.97%)
limonene (1.21%)	isoborneol (0.06%)
1,8-cineole (0.21%)	α-humulene (0.20%)
(Z)-β-ocimene (0.16%)	p-cymen-8-ol (0.50%)
γ-terpinene (5.54%)	spathulenol (0.13%)

(E)-β-ocimene (0.04%)  
p-cymene (7.35%)  
terpinolene (0.05%)

thymol (45.22%)  
carvacrol (33.05%)

This same year, Russo (1998) examined the composition of oils produced from the inflorescences of 23 collections of *O. vulgare* ssp. *hirtum* from Calabria (Italy). A summary of the results of the GC and GC/MS analyses are presented in T-8. As can be seen, this reviewer has grouped the oils according to their main components.

An oil of *O. vulgare* grown in Chile was found to contain the following composition (Montes et al. 1998).

α-pinene (3.74%)	linalyl acetate (1.36%)
β-pinene (0.23%)	myrtenal (0.75%)
sabinene (0.22%)	borneol (0.36%)
myrcene + α-terpinene (8.14%)	isoborneol (0.07%)
	α-terpineol (0.06%)

**Comparative percentage composition of *Origanum vulgare* ssp. *hirtum* oils of different Italian origin**
**T-6**

Compound	Policoro <sup>a</sup> Bitonto <sup>b</sup>	Policoro Grecia	Policoro Castellaneta	Policoro Zagabria	Policoro Policoro	Monreale Monreale	Villalba Monreale
$\alpha$ -thujene	t	0.24	0.45	t	0.35	0.83	0.98
$\alpha$ -pinene	0.20	0.24	t	t	t	0.11	0.21
sabinene	t	t	0.60	t	t	t	t
myrcene	1.63	1.34	1.73	t	1.49	2.43	1.79
$\alpha$ -terpinene	1.23	0.40	2.77	0.17	3.82	3.73	3.93
p-cymene	6.72	10.65	8.95	2.96	7.89	11.54	11.91
limonene	t	t	0.13	t	0.19	0.63	0.81
1,8-cineole	t	t	1.70	t	1.00	0.66	0.28
$\gamma$ -terpinene	8.21	3.46	21.63	7.17	25.88	16.43	16.86
linalool	0.10	t	t	t	0.67	t	t
terpineol-4-ol	t	t	t	t	t	t	t
thymol	41.28	t	54.92	79.30	39.42	54.62	58.92
carvacrol	37.67	81.11	1.76	6.50	1.90	0.91	0.61
$\beta$ -caryophyllene	t	t	0.14	0.90	7.73	1.14	0.21
$\alpha$ -humulene	t	t	t	t	0.47	t	t
$\beta$ -cubebene†	t	t	t	t	5.94	t	t
$\gamma$ -elemene†	t	0.99	t	0.93	t	1.62	0.4

† misidentifications based on elution order

<sup>a</sup> area where grown Policoro, 28m (Matera province), Monreale, 310M (Palermo province) and Villalba, 600m (Caltanissetta province)

<sup>b</sup> biotypes

t = trace (&lt;0.1%)

**Comparative percentage composition of *Origanum vulgare* ssp. *hirtum* oil and a commercial oregano oil**
**T-7**

Compound	<i>Origanum</i> <i>vulgare</i> ssp. <i>hirtum</i> oil	Commercial Oregano Oil	Compound	<i>Origanum</i> <i>vulgare</i> ssp. <i>hirtum</i> Oil	Commercial Oregano Oil
$\alpha$ -thujene	-	0.13	<i>cis</i> -sabinene hydrate	0.22	4.32
$\alpha$ -pinene	0.88	2.40	linalool	0.12	-
camphene	0.15	5.36	linalyl acetate	-	0.06
$\beta$ -pinene	0.08	0.34	terpinen-4-ol	-	t
sabinene	0.04	1.00	$\beta$ -caryophyllene	1.50	0.62
myrcene	0.61	0.75	methyl carvacrol	0.05	0.14
$\alpha$ -phellandrene	0.07	0.12	<i>trans</i> -dihydrocarvone	0.03	0.13
$\alpha$ -terpinene	0.62	1.53	<i>cis</i> -dihydrocarvone	t	0.02
limonene	0.14	4.12	isoborneol	0.10	0.09
1,8-cineole	0.18	0.68	$\alpha$ -terpineol	0.42	2.31
$\beta$ -phellandrene	0.08	t	$\gamma$ -elemene	0.20	0.07
$\gamma$ -terpinene	2.07	1.32	$\beta$ -bisabolene	0.15	0.09
$\beta$ -ocimene*	0.09	t	$\gamma$ -cadinene	0.02	-
p-cymene	8.76	40.15	<i>trans</i> -carveol	0.10	0.06
terpinolene	0.05	0.40	calamenene*	0.03	-
6-methyl-3-heptanol	-	0.11	p-cymen-8-ol	0.08	-
nonanal	-	0.09	carvacryl acetate	0.36	-
1-octen-3-ol	0.37	0.04	spathulenol	0.05	0.05
<i>trans</i> -sabinene hydrate	0.15	t	thymol	2.45	31.80
$\beta$ -bourbonene	-	0.05	carvacrol	79.58	0.43

\*correct isomer not identified

t = trace (0.01%)

Compound	Thymol Rich (14) <sup>a</sup>	Carvacrol Rich (3)	Mixed (6)
α-thujene	0.76-1.65	0.92-1.48	1.01-1.77
α-pinene	0.37-0.78	0.48-0.63	0.47-0.86
camphene	0.08-0.15	0.09-0.11	0.07-0.14
sabinene	0.05-0.53	0.06-0.14	0.10-0.19
β-pinene	0.14-0.28	0.11-0.30	0.13-0.16
1-octen-3-ol	0.06-0.22	0.03-0.15	0.12-0.26
3-octanone	0.04-0.10	0-0.07	0.04-0.11
myrcene	1.38-2.69	1.72-2.37	1.82-2.36
3-octanol	0-0.02	-	0-0.04
α-phellandrene	0.21-0.40	0.22-0.31	0.31-0.38
δ-3-carene	0.06-0.11	0.08-0.09	0.08-0.13
α-terpinene	2.23-4.11	1.87-2.52	2.11-3.24
p-cymene	3.57-8.89	3.82-4.38	3.63-9.74
limonene	0.50-0.74	0.43-0.50	0.43-0.61
1,8-cineole	0-0.18	0-0.15	0-0.16
(Z)-β-ocimene	0.20-0.79	0.16-0.31	0.22-0.48
(E)-β-ocimene	0.18-1.28	0.19-0.22	0.12-1.47
γ-terpinene	15.98-32.59	12.64-17.61	16.14-25.94
cis-p-menth-2-en-1-ol†	0.39-1.31	0.07-1.10	0.70-1.02
terpinolene	0.08-0.17	0.09-0.15	0.08-0.13
trans-p-menth-2-en-1-ol†	0.05-0.17	0.02-0.18	0-0.15
linalool	0.18-1.73	0.07-0.23	0.09-0.60
borneol	0.02-0.40	0.08-0.21	0.08-0.18
terpinen-4-ol	0.16-0.59	0.39-0.53	0.35-0.44
α-terpineol	0.08-1.03	0.10-0.96	0.09-1.79
cis-dihydrocarvone	0-0.11	0.08-0.13	0-0.06
methyl thymol	0.33-1.26	0.06-0.19	0.15-0.73
carvone	0-0.39	0-0.08	0-0.02
methyl carvacrol	1.99-3.77	2.81-4.67	2.89-4.20
linalyl acetate	0-0.02	-	-
bornyl acetate	0-0.10	0-0.05	0-0.28
thymol	38.76-55.46	7.91-9.24	19.00-45.69
carvacrol	0.27-8.43	50.40-56.63	10.02-36.43
cuminyl alcohol†	0-0.14	-	-
carvacryl acetate	0-1.24	0-0.18	0-0.63
thymyl acetate	0-0.90	0.26-0.46	0.06-0.50
α-copaene	0-0.06	0-0.20	0-0.06
β-bourbonene	0-0.04	-	-
β-caryophyllene	0.75-4.40	1.11-1.66	0.07-2.63
β-cubebene†	0-0.16	0-0.07	0-0.13
aromadendrene	0-0.03	0-0.09	0-0.21
α-humulene	0.13-0.80	0.20-0.40	0.25-0.80
allo-aromadendrene†	0-0.12	0-0.02	0-0.25
γ-murolene	0.06-0.28	0.06-0.13	0.09-0.39
germacrene D	0.11-1.86	0.08-0.69	0.32-1.27
valancene†	0.07-0.26	0.11-0.18	0.13-0.26
α-murolene	0-0.10	0.03-0.06	0.03-0.29
β-bisabolene	0.37-3.00	0.63-1.29	0.75-2.09
γ-cadinene	0.12-0.62	0.04-0.13	0.07-0.51
δ-cadinene	0.12-0.51	0.15-0.68	0.23-0.53
germacrene D-4-ol†	0-0.36	0-0.18	0.04-0.13
caryophyllene oxide	0-0.44	0.04-0.25	0.06-0.91
α-murolol†	0-0.22	0.09-0.21	0-0.47
α-cadinol	0-0.44	0-0.4-0.08	0.04-0.53

† tentative identification

<sup>a</sup> number of analyses of separate collections



limonene (0.12%)	geranial (0.06%)
p-cymene (33.57%)	geraniol (0.06%)
γ-terpinene (11.38%)	carvone (0.26%)
1-octen-3-ol (1.77%)	pulegone† (0.11%)
linalool + β-caryophyllene† (4.56%)	carvacrol (29.35%)

† incorrect identity based on elution order

Tiziana Baratta et al. (1998) used GC/MS to analyze a lab-distilled oil produced from oregano leaves obtained from the spice shelf in the UK. The components identified in this oil were as follows:

tricyclene (t)	α-p-dimethylstyrene (0.1%)
α-thujene (1.2%)	terpinolene (0.1%)
α-pinene (0.7%)	linalool (0.1%)
benzaldehyde (0.1%)	terpinen-4-ol (1.2%)
camphene (0.2%)	α-terpineol (0.3%)
sabinene (0.1%)	cis-carveol (0.1%)
β-pinene (2.0%)	methyl carvacrol (4.8%)
myrcene (0.9%)	thymol (32.4%)
α-phellandrene (0.2%)	carvacrol (16.7%)
δ-3-carene (0.1%)	thymyl acetate (0.3%)
α-terpinene (1.8%)	eugenol (t)
p-cymene (11.5%)	carvacryl acetate (0.1%)
β-phellandrene (0.4%)	α-copaene (0.1%)
limonene (0.3%)	β-caryophyllene (0.4%)
(Z)-β-ocimene (t)	β-funebrene (0.1%)
(E)-β-ocimene (0.1%)	trans-α-bergamotene (0.1%)
γ-terpinene (10.4%)	germacrene D (0.3%)
β-terpineol*† (0.4%)	α-cadinene (0.5%)

\*correct isomer not identified

t = trace (<0.1%)

† incorrect identity based on elution order

This reviewer realizes that oregano oil is usually rich in carvacrol; however, when *O. vulgare* ssp. *hirtum* is harvested from the wild, mixed chemotypes can easily be collected. The above is an example of mixed thymol/carcacrol chemotype.

Kanias et al. (1998) reported that the oil yield of Greek *O. vulgare* ranged from 2.85-16.60%. The authors also determined that the percentage composition of the major components ranged as follows:

α-pinene (0.17-2.47%)	p-cymene (2.68-31.60%)
camphene (0.04-0.86%)	linalool (0.05-0.88%)
β-pinene (0.03-0.48%)	δ-cadinene (0.19-0.81%)
myrcene (0.18-1.59%)	thymol (0.22-64.80%)
α-terpinene (0.31-3.74%)	carvacrol (2.41-90.30%)
γ-terpinene (0.12-14.50%)	

Also in 1998, Karpouhtsis et al. compared the composition of carvacrol-rich oregano oils produced from *O. vulgare* ssp. *hirtum* and *Coridothymus capitatus* (L.) Reichenb. growing wild on Mt. Taygetos (Greece). A summary of the results of this study are shown in T-9.

An oil produced from *O. vulgare* ssp. *hirtum* plants that were collected in Crete was analyzed by Daferera et al. (2000) using GC/MS. The main constituents found in this oil were:

α-thujene (0.3%)	borneol (0.5%)
α-pinene (0.6%)	terpinen-4-ol (0.3%)

Comparative percentage composition of two carvacrol-rich oils

T-9

Compound	<i>Origanum vulgare</i> ssp. <i>hirtum</i> Oil	<i>Coridothymus capitatus</i> Oil
α-thujene	0.06	-
α-pinene	1.22	0.65
camphene	0.10	0.16
β-pinene	0.10	0.07
sabinene	0.06	0.05
myrcene	1.35	0.94
α-phellandrene	0.13	0.06
α-terpinene	1.04	0.67
limonene	0.17	0.13
1,8-cineole	0.19	0.14
β-phellandrene	0.03	-
γ-terpinene	5.92	2.25
(E)-β-ocimene	0.17	-
p-cymene	9.71	6.41
terpinolene	0.05	0.08
3-octanol	-	0.07
trans-sabinene hydrate	0.41	0.27
linalool	0.12	0.61
linalyl acetate	-	0.07
terpinen-4-ol	0.12	0.03
cis-sabinene hydrate	0.30	0.13
β-caryophyllene	1.37	1.91
methyl carvacrol	0.09	0.03
trans-dihydrocarvone	0.04	0.06
cis-dihydrocarvone	0.06	-
isoborneol	0.16	0.61
α-terpineol	0.60	0.08
γ-elemene	-	0.06
β-bisabolene	-	0.05
γ-cadinene	0.05	0.04
calamenene*	0.04	-
p-cymen-8-ol	-	0.61
carvacryl acetate	0.20	-
spathulenol	-	0.10
thymol	0.71	1.46
carvacrol	74.56	81.46
* correct isomer not identified		

β-pinene (0.5%)	carvacrol (7.8%)
α-terpinene (1.0%)	thymol (63.3%)
p-cymene (9.9%)	β-caryophyllene (0.5%)
γ-terpinene (12.7%)	β-bisabolene (0.4%)
linalool (0.6%)	

This is an example of the thymol-rich chemotype of *O. vulgare* ssp. *hirtum*. Dorman et al. (2000) using GC and GC/MS analyzed an oil of *O. vulgare*. It was found to possess the following composition:

camphene (0.53%)	linalool (0.61%)
myrcene (1.45%)	borneol (0.43%)

$\delta$ -3-carene (0.14%)	terpinen-4-ol (0.82%)
$\alpha$ -terpinene (1.01%)	$\alpha$ -terpineol (0.75%)
p-cymene (12.70%)	$\gamma$ -terpineol† (0.15%)
limonene (0.77%)	bornyl acetate (0.11%)
$\gamma$ -terpinene (2.09%)	thymol (1.36%)
terpinolene (0.27%)	carvacrol (69.40%)

† identity requires corroboration because  $\gamma$ -terpineol has not been unequivocally identified as occurring naturally

D'Antuono et al. (2000) analyzed oils of numerous accessions of *O. vulgare* from Liguria (northern Italy). They found that the oils could be subdivided into three groups; however, none possessed the desired composition of *O. vulgare* ssp. *hirtum* (see T-10).

This same year, Mastelic et al. (2000) determined that an oil of *O. vulgare* ssp. *hirtum* that was collected near Split (Croatia) possessed the following composition:

$\alpha$ -thujene (0.5%)	2-isopropyl-1-methoxy-
$\beta$ -pinene (0.3%)	4-methylbenzene (0.4%)
sabinene (0.2%)	borneol (1.1%)
1-octen-3-ol (2.0%)	p-cymen-8-ol (0.1%)
$\delta$ -3-carene (0.3%)	thymol (49.3%)
$\beta$ -phellandrene (0.1%)	carvacrol (24.6%)
$\gamma$ -terpinene (2.0%)	$\beta$ -caryophyllene (0.7%)
p-cymene (11.8%)	$\alpha$ -humulene (0.1%)
terpinolene (0.3%)	$\alpha$ -muurolene (0.3%)
bornyl acetate (0.1%)	$\beta$ -farnesene* (0.3%)
terpinen-4-ol (1.0%)	$\beta$ -bisabolene (0.7%)
methyl thymol (0.5%)	$\delta$ -cadinene (0.2%)

\* correct isomer not identified

The authors also examined the bound glycosides and, using either enzyme isolation ( $\beta$ -glucosidase) or acid hydrolysis (HCl); the volatiles released were found to be as follows:

3-hexenol* (t-3.4%)	p-cymen-8-ol (0.8-1.2%)
1-octen-3-ol (1.3-5.4%)	eugenol (0-0.4%)
benzaldehyde (0.8-0.9%)	thymol (6.8-7.5%)
terpinen-4-ol (0-0.6%)	carvacrol (1.9-5.5%)
thymoquinone (40.2-47.0%)	1H-indole (0.8-2.2%)
	1H-indole (0.8-2.2%)
methyl salicylate (0-0.6%)	xethanol (0-1.3%)
benzyl alcohol (2.0-8.9%)	butyl phthalate† (t-3.0%)
2-phenethyl alcohol (0.8-5.6%)	

\* correct isomer not identified

† artifact

D. Vokou, S. Kokkini and J.-M. Bessière, *Geographic variation of Greek oregano (Origanum vulgare ssp. hirtum) essential oils*. Biochem. Syst. Ecol., 21, 287-295 (1993).

M. Melegari, F. Severi, M. Bertoldi, S. Benvenuti, G. Circetta, I. Morone Fortunato, A. Bianchi, C. Leto and A. Carrubba, *Chemical characterization of essential oils of some Origanum vulgare subspecies of various origin*. Rivista Ital. EPPOS, (16), 21-28 (1995).

C. Bourrel, G. Vilarem, G. Michel and A. Gaset, *étude des propriétés bactériostatiques et fongistatiques en Milieu solide de 24 huiles essentielles préablement analysées*. Rivista Ital. EPPOS, (16), 3-12 (1995).

L.-F. Zhu, Y.-H. Li, B.-L. Li, B.-Y. Lu and W.-L. Zhang, *Aromatic plants and essential constituents. Supplement 1*. South China Institute Botany, Chinese Acad. Sciences, Hai Feng Publ. Co., Peace Book Co., Hong Kong (1995).

A. Sivropoulou, E. Papanikolaou, G. Nikolaou, S. Kokkini, T. Lanaras and M. Arsenakis, *Antimicrobial and cytotoxic activities of Origanum essential oils*. J. Agric. Food Chem., 44, 1202-1205 (1996).

S. Kokkini, R. Karousou, A. Dardioti and T. Lanaras, *Autumn essential oils of Greek oregano*. Phytochemistry, 44, 883-886 (1997).

K. Adam, A. Sivropoulou, S. Kokkini, T. Lanaras and M. Arsenakis, *Antifungal activities of Origanum vulgare subsp. hirtum, Mentha spicata, Lavandula angustifolia and Salvia fruticosa essential oils against human pathogenic fungi*. J. Agric. Food Chem., 46, 1739-1745 (1998).

M. Russo, G. C. Galletti, P. Bocchini and A. Carnacini, *Essential oil chemical composition of wild populations of Italian oregano spice (Origanum vulgare ssp. hirtum (Link) Ietswaart): A preliminary evaluation of their use in chemotaxonomy by cluster analysis I. Inflorescences*. J. Agric. Food Chem., 46, 3471-3746 (1998).

M. A. Montes, T. Wilkomirsky and H. Bello, *Antibacterial activity of essential oils from aromatic plants growing in Chile*. Fitoterapia, 69, 170-172 (1998).

M. Tiziana Baratta, H. J. D. Dorman, S. G. Deans, D. M. Biondi and G. Ruberto, *Chemical composition, antimicrobial and antioxidant activity of laurel, sage, rosemary, oregano and coriander essential oils*. J. Essent. Oil Res., 10, 618-627 (1998).

K. D. Kanas, C. Souleles, A. Loukis and E. Philotheou-Panou, *Trace elements and essential oil composition in chemotypes of the aromatic plant Origanum vulgare*. J. Radioanal. Nucl. Chem., 227, 23-29 (1998).

I. Karpouhtsis, E. Pardali, E. Feggou, S. Kokkini, Z. G. Scouras and P. Mavragani-Tsipidou, *Insecticidal and genotoxic activities of oregano essential oils*. J. Agric. Food Chem., 46, 1111-1115 (1998).

D. J. Daferera, B. N. Ziogas and M. G. Polissiou, *GC-MS analysis of essential oils from some Greek aromatic plants and their fungitoxicity on Penicillium digitatum*. J. Agric. Food Chem., 48, 2576-2581 (2000).

H. J. D. Dorman, P. Surai and S. G. Deans, *In vitro antioxidant activity of a number of plant essential oils and phytoconstituents*. J. Essent. Oil Res., 12, 241-248 (2000).

L. F. D'Antuono, G. C. Galletti and P. Bocchini, *Variability of essential oil content and composition of Origanum vulgare L. Populations from a North Mediterranean Area (Liguria Region, Northern Italy)*. Ann. Bot., 86, 471-478 (2000).

J. Mastelic, M. Milos and I. Jerkovic, *Essential oil and glycosidically bound volatiles of Origanum vulgare L. ssp. hirtum (Link) Ietswaart*. Flav. Fragr. J., 15, 190-194 (2000).

## Armoise oil

Vernin and Merad (1994) used a combination of GC/MS and retention indices on both a polar and a non-polar capillary column to characterize 10 chrysanthenyl compounds in armoise (*Artemisia herba-alba* Asso) oil of Algerian origin. The compounds identified were as follows:

cis-chrysanthenol  
cis-chrysanthenyl acetate  
cis-chrysanthenyl propionate  
cis-chrysanthenyl isobutyrate

Compound	Group 1	Group 2	Group 3
2-hexenal*	0.02-0.06	0.03-0.09	0.23-0.47
3-hexenol*	0.01-0.03	0-0.02	0.07-0.11
$\alpha$ -thujene	0.57-1.37	0.26-0.56	0.18-0.27
$\alpha$ -pinene	0.33-0.60	0.13-0.18	0.07-0.39
camphene	0.04-0.07	0.02-0.03	0.04-0.27
sabinene	0.10-0.37	0.47-1.06	0.97-5.54
1-octen-3-ol	0.81-1.20	0.63-0.94	1.60-1.73
3-octanone	0.07-0.18	0.16-0.19	0.16-0.43
myrcene	0.82-1.50	0.43-0.62	0.01-0.37
3-octanol	0.02-0.10	0.25-0.33	0.43-1.32
$\alpha$ -phellandrene	0.11-0.22	0.05-0.07	0.03
$\delta$ -3-carene	0.04-0.07	0.03	-
$\alpha$ -terpinene	1.19-3.37	0.66-1.66	0.41-1.08
p-cymene	5.74-17.78	1.43-3.01	1.83-3.50
limonene	0.15-0.28	0.11-0.51	1.69-2.09
(Z)- $\beta$ -ocimene	0.17-1.77	0.69-3.71	0.32-2.11
(E)- $\beta$ -ocimene	0.06-1.38	0.16-0.80	0.26-1.74
$\gamma$ -terpinene	0.83-26.83	2.17-7.22	0.97-2.66
cis-sabinene	0.18-0.76	0.17-0.85	1.39-5.71
cis-linalool oxide†	0-0.02	0.05-0.44	0.33-0.59
terpinolene + trans-linalool oxide†	0.05-0.12	0.07-1.01	0.71-0.77
trans-sabinene hydrate	0.05-0.54	0.04-1.66	0.83-0.85
linalool	0.07-1.83	18.39-43.06	2.06-5.78
trans-pinene hydrate‡	0.02-0.04	0.01-0.37	0.35-0.37
p-mentha-1,3,8-triene	0.02-0.04	0-0.04	0.02-0.06
borneol	0.06-0.11	0.02-0.09	0.64-1.01
terpinen-4-ol	0.31-0.74	0.28-5.31	4.11-4.14
p-cymen-8-ol	0.02-0.10	0-0.08	0.13-0.33
$\alpha$ -terpineol	0.08-0.19	0.11-1.92	2.02-3.41
cis-dihydrocarvone	0-0.04	0-0.02	0-0.11
myrtenyl acetate	0.01-0.02	0-0.05	0.10-0.19
methyl thymol	0.02-2.15	0.10-0.64	0.10-0.15
carvone	0-0.02	0-0.01	0.03-0.13
methyl carvacrol	1.63-5.40	1.07-1.51	0.08
bornyl acetate	-	0-0.02	0.07-0.31
indole	0-0.04	0.01-0.02	0.03-0.04
thymol	0.68-47.82	3.96-20.37	0.38-1.06
carvacrol	0.77-62.86	0.30-2.41	0.04-0.24
eugenol + thymyl acetate	0-0.02	0.02	0.08-0.35
$\alpha$ -cubebene	0.01-0.03	0.02-0.14	0-0.04
$\alpha$ -copaene	0.05-0.15	0.05-0.16	0.20-0.52
$\beta$ -bourbonene	0.05-0.32	0.26-0.65	4.21-13.40
$\beta$ -cubebene	0-0.06	0.05-0.16	0.24-1.29
$\beta$ -elemene	0.01-0.06	0.08-0.17	0.18-0.56
jasnone*	0.01	0.01-0.02	0-0.02
$\beta$ -caryophyllene	2.54-9.21	6.63-18.28	5.51-7.84
$\beta$ -gurjunene	0.07-0.10	0.08-0.12	0.54-1.62
aromadendrene	0.03-0.16	0.04-0.19	0.03-0.05
$\alpha$ -cadinene	0-0.02	0.02-0.04	0.25-0.80
$\alpha$ -humulene	0.23-1.17	0.81-1.78	0.16-0.53
allo-aromadendrene	0.02-0.06	0.12-0.24	0.52-0.86
(Z)- $\beta$ -farnesene	0.02-0.05	0.03-0.07	0.07-0.08
$\gamma$ -muurolene	0.38-7.90	7.39-15.94	5.14-12.31

(T-10 continues on following page)

Compound	Group 1	Group 2	Group 3
germacrene D	0.24-1.18	1.10-4.02	1.14-2.09
$\alpha$ -muurolene	0.04-0.08	0.03-0.09	0.10-0.12
$\beta$ -bisabolene	1.38-5.09	2.65-4.25	0.15-4.57
$\delta$ -cadinene	0.56-0.91	0.60-0.93	0.97-0.99
(E)- $\gamma$ -bisabolene	0-0.06	0.03-0.32	0.98-1.55
germacrene D-4-ol	0.13-0.56	0.73-2.83	5.39-16.00
caryophyllene oxide	0.30-0.94	0.80-6.55	10.17-21.48
globulol	0-0.04	0.09-0.29	0.57-1.29
T-cadinol	0.03-0.14	0.11-0.33	0.02-0.35
$\alpha$ -muurolol	0.02-0.03	0.01-0.10	0.27-0.30
$\alpha$ -cadinol	0.06-0.27	0.22-0.76	0.83-1.22
* correct isomer not identified			
† furanoid form			
‡ probably misidentification of <i>trans</i> -p-menth-2-en-1-ol			

*cis*-chrysanthenyl butyrate  
*cis*-chrysanthenyl isobutyrate  
*cis*-chrysanthenyl tiglate  
*cis*-chrysanthenyl phenylacetate  
*trans*-chrysanthenol  
*trans*-chrysanthenyl acetate

It is worth noting that the author presented mass spectra and retention indices for each of the above listed compounds.

A year later, Vernin et al. (1995) reported a detailed analysis of armoise oil produced from plants collected in Algeria. They found that the oil contained the following constituents:

1-octene	carvone
dimethyl-1-heptene*	piperitone
1,2-dimethylcycloheptene†	(Z)-jasnone
1-decene	2-hexadecanone
artemisiatriene	ethyl isobutyrate
tricyclene	ethyl isovalerate
$\alpha$ -thujene	<i>cis</i> -chrysanthenyl acetate
$\alpha$ -pinene	dehydrocarvyl acetate†
camphene	bornyl acetate
$\alpha$ -fenchene†	<i>cis</i> -chrysanthenyl propionate
sabinene	bornyl propionate
$\beta$ -pinene	<i>cis</i> -chrysanthenyl butyrate
p-menthadiene*	<i>cis</i> -chrysanthenyl isovalerate
alkylbenzene*	<i>cis</i> -chrysanthenyl 2-methylbutyrate
2,3-bis-methylene-bicyclo[3.2.1]octane†	<i>cis</i> -chrysanthenyl valerate
$\alpha$ -phellandrene	bornyl isovalerate
$\alpha$ -terpinene	<i>cis</i> -chrysanthenyl tiglate
$\alpha$ -p-dimethylstyrene	<i>cis</i> -chrysanthenyl phenylacetate
trimethylbenzene*	1-octen-3-ol
m-cymene	<i>cis</i> -chrysanthenol
p-cymene	
1,8-cineole	
$\beta$ -ocimene*	

limonene	borneol
$\gamma$ -terpinene	thujyl alcohol*
terpinolene	terpinen-4-ol
$\alpha$ -cubebene	myrtenol
$\alpha$ -copaene	$\alpha$ -terpineol
$\gamma$ -muurolene	<i>cis</i> -p-menth-2-en-7-ol
$\beta$ -caryophyllene	<i>cis</i> -piperitol
$\alpha$ -humulene	<i>trans</i> -carveol
aromadendrene	p-mentha-1,5-dien-7-ol
germacrene D	<i>cis</i> -myrtanol
germacrene B‡	<i>trans</i> -pinocarveol
$\alpha$ -muurolene	p-mentha-1,4-dien-8-ol
calamenene*	p-mentha-1,4-dien-7-ol
$\delta$ -cadinene	humulene epoxide*
2-pentanone	palustrol
2-isopentanone	spathulenol
hexanal	caryophyllene oxide
3-octanone	calamenenol*
2,6,6-trimethyl-1,3-cyclohexadiene-1-carboxaldehyde†	caryophyllenol*
filifolone†	2-methylfuran
$\alpha$ -thujone	2-ethylfuran
$\beta$ -thujone	2,3-dehydro-1,8-cineole
chrysanthenone	2-pentylfuran
camphor	chrysanthenone oxide*†
myrtenal	verbenone oxide†
verbenone	chrysanthenone oxide*†
cuminaldehyde	eugenol
	methyl eugenol

\*correct isomer not identified  
† tentative identification

The authors also removed the acids and phenols from the oil, and after methylation they characterized the methyl esters of the following acids and phenols:

heptanoic acid	pentadecanoic acid
nonanoic acid	hexadecanoic acid
campholenic acid	octadecanoic acid
myrtanic acid	4-isopropylanisole
decanoic acid	methyl chavicol
undecanoic acid	t-butyl anisole*

dodecanoic acid  
tridecanoic acid  
tetradecanoic acid

cumenic acid  
methyl isoeugenol\*  
acetovanillone

\*correct isomer not identified

In addition, Vernin et al. examined the composition of a number of oils produced from plants collected from different areas in Algeria. These oils could be categorized according to their major constituents such as oils with the following ratio of major constituents:

1. camphor >  $\beta$ -thujone >  $\alpha$ -thujone > chrysanthenone
2. camphor > chrysanthenone >  $\alpha$ -thujone >  $\beta$ -thujone
3. camphor > chrysanthenone >  $\beta$ -thujone >  $\alpha$ -thujone
4. camphor >  $\beta$ -thujone > chrysanthenone >  $\alpha$ -thujone
5.  $\alpha$ -thujone > camphor > chrysanthenone >  $\beta$ -thujone

A summary of the quantitative data on the oils in these categories can be seen in T-11.

Lamiri et al. (1997) collected *A. herba-alba* plants from different regions of Morocco to determine the geographical distribution of the numerous chemotypes of armoise. They found that in Region A (Imintamout, Tizin'test, Talioune regions) camphor,  $\alpha$ -thujone,  $\alpha$ -thujone/ $\beta$ -thujone and  $\alpha$ -thujone/camphor chemotypes and in Region B (Tizin'tichka, Ouarzazate, Tansikkt, Iminoulaoun regions)  $\alpha$ -thujone,  $\beta$ -thujone, chrysanthenone, camphor, *cis*-chrysanthenyl acetate,  $\alpha$ -thujone/ $\beta$ -thujone, davanone,  $\alpha$ -thujone/camphor,  $\beta$ -thujone/camphor,  $\alpha$ -thujone/chrysanthenone,  $\alpha$ -thujone/

davanone, chrysanthenone/davanone, davanone/camphor and 1,8-cineole/camphor chemotypes. The chemotypes found in Region C (Midelt, Boumia, Tounfite regions) were  $\alpha$ -thujone,  $\beta$ -thujone, camphor, davanone,  $\alpha$ -thujone/ $\beta$ -thujone,  $\alpha$ -thujone/camphor,  $\beta$ -thujone camphor,  $\beta$ -thujone/davanone,  $\alpha$ -thujone/davanone and davanone/camphor, whereas in Region D (Errachidia, Boudrib, Goulimina and Erfoud regions) only the  $\alpha$ -thujone chemotype could be found.

Vernin and Párkányi (2001) analyzed a petroleum ether/methylene chloride (1:1) mixture non-polar extract and an ethanol polar extract of *A. herba alba* leaves/branches of Algerian origin. A quantitative analysis of the non-polar extract was reported and it is as follows:

camphor (30.0%)	C <sub>29</sub> and C <sub>31</sub> alkanes (1.0%)
1,8-cineole (12.5%)	
camphene (3.0%)	C <sub>18</sub> unsaturated fatty acids (1.0%)
$\alpha$ -thujone (4.0%)	
$\beta$ -thujone (16.0%)	epoxy terpenoid ketones (1.0%)
chrysanthenone (0.3%)	
verbenone (0.2%)	

In addition to the compounds reported in Vernin's 1995 report on *A. herba-alba*

Compound	1(4)	2(4)	3(6)	4(1)	5(2)
artemisiatriene	0.26-1.05	0.21-0.60	0.10-1.50	0.70	0.05-0.46
tricyclene	0.14-0.27	0.30-0.38	0.10-0.49	0.31	0.23-0.40
$\alpha$ -pinene	0.56-1.20	0.60-1.80	0.30-0.70	0.35	1.20-1.28
camphene	2.3-4.4	3.72-5.80	1.70-7.90	6.27	3.00-6.50
sabinene	0.12-0.79	0.16-0.62	0.13-0.52	0.13	0.30-0.70
$\beta$ -pinene	0.13-0.26	0.13-0.31	0.12-0.35	0.12	0.20-0.30
p-cymene	0.26-0.67	0.44-0.67	0.15-1.00	0.65	0.70-1.20
1,8-cineole	8.25-18.13	8.00-19.10	5.00-17.00	10.80	9.00-9.80
$\gamma$ -terpinene	0.13-0.79	0.14-0.76	0.10-0.55	0.30	0.13-0.30
filifone†	0.70-1.70	1.30-3.60	0.65-4.00	0.67	0.70-4.10
$\alpha$ -thujone	11.60-15.60	4.37-12.00	2.02-12.00	1.55	22.00-26.70
$\beta$ -thujone	19.6-20.8	1.65-5.24	2.35-9.30	6.20	8.60-14.20
chrysanthenone	0-10.0	9.20-22.50	6.80-21.20	5.00	7.30-11.50
camphor	23.00-29.00	23.00-36.00	30.00-41.00	48.00	19.00-19.50
<i>cis</i> -chrysanthenol	0.12-0.70	0.26-0.80	0.10-0.75	0.23	0.30-0.70
borneol	0.52-1.47	0.53-1.20	0.20-0.96	1.50	0.84-0.90
myrtenol + terpinen-4-ol	2.35-4.80	0.60-3.35	2.70-4.83	3.10	1.70-2.20
verbenone	0.70-0.92	0.23-1.00	0.10-0.80	0.30	0.43-0.83
<i>cis</i> -chrysanthenyl acetate	0.17-0.30	0.20-0.28	0.15-1.10	0.10	0.25-0.27
bornyl acetate	0.15-0.80	0.17-1.56	0.12-0.66	0.10	0.17-0.40

Numbers in parentheses refer to number of oils found in the various categories; † tentative identification

oil, the following additional compounds were identified:

5-methyl-2-ethylfuran	<i>trans</i> -sabinol
2,4-dimethylhexane	<i>trans</i> -piperitol
octane	<i>cis</i> -carveol
p-xylene	<i>cis</i> -verbenyl acetate
verbenene	filifolide A†
cumene	filifolide B†
$\delta$ -3-carene†	dibutyl phthalate°
$\beta$ -phellandrene	ethyl octadecanoate
3-hexen-2-one	methyl 9,12-
1-(1-cyclohexyl)-ethanone	octadecadienoate
pinocarvone	dihydro-oplophenone†
chrysanthenal†	isopathulenol
verbenal†	5-methoxycoumarin
(E)-tagetone	
1,2-dimethyl-2-isopropylcyclopentanol†	

°correct isomer not identified

° artifact

† tentative identification

In addition, a number of hydrocarbons ( $C_{13}$ - $C_{32}$ ) were also found in the non-polar extract.

Also in 2001, Salido et al. examined the composition of an oil of *A. herba-alba* ssp. *valentina* (Lam.) Marcl., which was lab-distilled from plants collected near Quesada, Jaén province (Spain). Using a combination of GC (for retention indices

and quantitative data) and GC/MS (for component identity confirmation), the oil composition was found to be as follows:

2,5-diethenyl-2-methyltetrahydrofuran (0.1%)  
 tricyclene (t)  
 $\alpha$ -thujene (0.1%)  
 $\alpha$ -pinene (1.7%)  
 ethyl tiglate (0.3%)  
 camphene (0.4%)  
 5,5-dimethyl-2(5H)-furanone (0.2%)  
 sabinene (0.2%)  
 $\beta$ -pinene (0.1%)  
 myrcene (5.1%)  
 $\alpha$ -phellandrene (t)  
 $\alpha$ -terpinene (0.6%)  
 p-cymene (13.5%)  
 limonene (1.6%)  
 1,8-cineole (10.2%)  
 lavender lactone (t)  
 (E)- $\beta$ -ocimene (0.1%)  
*cis*-arbusculone (0.5%)  
 $\gamma$ -terpinene (5.5%)  
*trans*-arbusculone (0.3%)  
*cis*-linalool oxide† (0.1%)  
 terpinolene + *trans*-linalool oxide† (0.3%)  
 linalool (0.9%)  
 isochrysanthenone† (0.5%)  
 3,7-dimethyl-1,5,7-octatrien-3-ol (0.2%)  
*cis*-p-menth-2-en-1-ol (0.2%)  
 chrysanthenone (6.7%)  
*trans*-pinocarveol (0.1%)  
*trans*-p-menth-2-en-1-ol (0.2%)  
 camphor (4.0%)  
*cis*-chrysanthenol (0.5%)



borneol (0.6%)  
 lavandulol (0.9%)  
 terpinen-4-ol (2.1%)  
 p-cymen-8-ol (0.3%)  
 $\alpha$ -terpineol (1.5%)  
*cis*-piperitol (0.1%)  
*trans*-piperitol (0.1%)  
 nordavanone (0.2%)  
 ascaridole (0.1%)  
 cuminaldehyde (0.2%)  
*cis*-chrysanthenyl acetate (5.6%)  
 bornyl acetate (0.4%)  
 p-cymen-7-ol (0.2%)  
 lavandulyl acetate (0.2%)  
 thymol (0.2%)  
 carvacrol (0.2%)  
 $\alpha$ -terpinyl acetate (0.8%)  
 eugenol (t)  
 $\alpha$ -copaene (t)  
 $\beta$ -caryophyllene (0.2%)  
 $\gamma$ -muurolene (1.5%)  
 davana ether (0.9%)  
 $\delta$ -cadinene (0.2%)  
 nerolidol\* (0.3%)  
 spathulenol (0.6%)  
 caryophyllene oxide (0.2%)  
 davanone (18.1%)  
 selin-11-en-4 $\alpha$ -ol (0.4%)  
 davanone-2 $\beta$ -ol (t)

t = trace (<0.1%)

† furanoid form

\* correct isomer not identified

## Large Cardamom or Nepalese Cardamom Oil

Large cardamom or Nepalese cardamom oil has become a small item of commerce from Nepal over the past few years. It is obtained from fruit of *Amomum subulatum* Roxb., a member of the Zingiberaceae family.

A survey of the literature reveals that in 1960 and 1961, Nigam and Purohit used the resorcinol method to determine that the major component of *A. subulatum* oil was 1,8-cineole (64.94%). Using

- G. Vernin and L. O. Merad, *Mass spectra and Kovats indices of some new cis-chrysanthenyl esters found in the essential oil of Artemisia herba-alba from Algeria*. J. Essent. Oil Res., 6, 437-448 (1994).
- G. Vernin, O. Merad, G. M. F. Vernin, R. M. Zamkotsian and C. Párkányi, *GC/MS analysis of Artemisia herba-alba Asso essential oils from Algeria*. In: *Food Flavors: generation, analysis and process influence*. Edit., G. Charalambous, p. 147-204. Elsevier Sci. BV, Amsterdam (1995).
- A. Lamiri, A. Belanger, M. Berrada, M. M. Ismaili-Alaoul and B. Benjilali, *Origine du polymorphisme chimique de L'armoise blanche (Artemisia herba-alba Asso) da Maroc*. In: *Plantes Aromatiques et Médicinales et leurs huiles essentielles, Congress Proceedings*. Edits., B. Benjilali, M. Ettalibi, M. M. Ismaili-Alaoui and S. Zrira, p. 81-93, Editions Acetes, Inst. Agronomique Veterinaire Hassan II, Rabat, Morocco (1997).
- G. Vernin and C. Párkányi, *GC/MS analysis of Artemisia herba-alba Asso from Algeria; Non-polar and polar extracts*. Rivista Ital. EPPS (32), 3-16 (2001).
- S. Salido, J. Altarejos, M. Nogueras and A. Sanchez, *Chemical composition of the essential oil of Artemisia herba-alba Asso ssp. valentina (Lam.) Marcl. J. Essent. Oil Res.*, 13, 221-224 (2001).

derivatization of components separated from the cineole-free oil and GC (peak enrichment), the authors inferred that the oil also contained  $\alpha$ -terpineol (7.15%),  $\alpha$ -terpinyl acetate (5.1%), sabinene (6.6%) a terpinene (10.7%) and a bisabolene (3.6%).

Nine years later, Lawrence (1970) determined that a lab-distilled oil of *A. subulatum* contained the following constituents:

$\alpha$ -pinene (2.0%)	$\gamma$ -terpinene (0.2%)
$\beta$ -pinene (2.4%)	p-cymene (0.2%)
sabinene (0.2%)	terpinen-4-ol (2.0%)
myrcene (0.3%)	$\delta$ -terpineol (0.8%)
$\alpha$ -terpinene (0.2%)	$\alpha$ -terpineol (5.6%)
limonene (10.2%)	(E)-nerolidol (1.0%)
1,8-cineole (74.0%)	

Although this analysis was performed before GC/MS became a routine analytical procedure, the components were isolated from the oil by preparative GC and their identities were confirmed by IR and retention times.

Fourteen years later, Gupta et al. (1984) analyzed some lab-distilled oils of the fruit of *A. subulatum* collected in Sikkim. Using peak enrichment techniques on a packed GC column, the authors tentatively determined that the range of main constituents of the oil were as follows:

sabinene (3.11-4.19%)
$\gamma$ -terpinene (4.72-8.73%)
1,8-cineole (77.00-89.00%)
$\alpha$ -bisabolene* (1.09-7.05%)
$\beta$ -terpineol (0-7.05%)
$\alpha$ -terpineol + $\alpha$ -terpinyl acetate (3.52-6.38%)

\*correct isomer not identified

In 1993, Kaur et al. also used peak enrichment techniques to compare the composition of an oil with a liquid CO<sub>2</sub> extract of *A. subulatum*. The components tentatively identified in this study were as follows:

$\alpha$ -pinene (1.48%, t <sup>b</sup> )	terpinolene (0.06%, 0)
camphene (0.04%, 0*)	linalool (0.11%, 0.10%)
$\beta$ -pinene (3.53%, t)	$\beta$ -terpineol* (2.76%, 1.45%)
myrcene (0.53%, 0)	$\alpha$ -terpinyl acetate (8.05%, 21.58%)
limonene (0.1%, 0)	neryl acetate (0.89%, 5.66%)
1,8-cineole (71.21%, 0.07%)	geranyl acetate (0.87%, 4.15%)
$\gamma$ -terpinene (0.32%, 0)	geraniol (0, 2.75%)
p-cymene (0.52%, 0.01%)	

\* oil

<sup>b</sup>liquid extract

\* correct isomer not identified

Zhu et al. (1993) reported that an oil of *A. subulatum* produced from dried fruit harvested in China was analyzed by GC/MS and found to contain:

$\alpha$ -pinene (0.56%)	terpinen-4-ol (1.86%)
$\beta$ -pinene (0.52%)	$\alpha$ -terpineol (4.06%)
myrcene (0.82%)	myrtenol (1.97%)
1,8-cineole (40.56%)	$\beta$ -bisabolene (0.72%)
linalool (0.57%)	nerolidol* (2.43%)

\*correct isomer not identified

In 1996, Gurudutt et al. used GC/MS to analyze an oil of large cardamom. The composition of this lab-distilled oil was found to be:

$\alpha$ -thujene (0.32%)	verbenone (0.05%)
$\alpha$ -pinene (3.79%)	cis-carveol (0.23%)
camphene (0.10%)	cuminaldehyde (0.10%)
$\beta$ -pinene (8.85%)	carvone (0.08%)
myrcene (1.15%)	linalyl acetate (0.11%)
p-cymene (0.10%)	hydroxycitronellal† (0.12%)
1,8-cineole (61.31%)	$\alpha$ -cubebene (0.03%)
$\gamma$ -terpinene (1.95%)	neryl acetate (0.08%)
linalool (0.67%)	$\alpha$ -copaene (0.10%)
myrcen-8-ol (0.17%)	$\beta$ -cubebene (0.10%)
$\alpha$ -campholenic aldehyde (0.19%)	$\delta$ -elemene† (1.19%)

norbornyl acetate† (0.19%)	longifolene (0.25%)	myrtenol (0.2%)
dihydrolinalool‡ (0.59%)	β-cedrene (0.08%)	verbenone (0.1%)
pinocarveol* (0.11%)	allo-aromadendrene (3.17%)	cis-carveol† (0.2%)
β-terpineol* (0.21%)	aromadendrene (0.38%)	carvone† (0.1%)
δ-terpineol (1.60%)	α-bisabolene* (0.35%)	cuminaldehyde† (0.1%)
terpinen-4-ol (1.29%)	β-bisabolene (0.29%)	linalyl acetate (0.1%)
α-terpineol (7.92%)	nerolidol* (0.32%)	hydroxycitronellal° (0.1%)
myrtenol (0.19%)		α-cubebene (t)

\*correct isomer not identified

† incorrect identification based on elution order

‡ incorrect identifications; these components do not occur naturally

neryl acetate (0.1%)  
 α-copaene (0.1%)  
 β-cubebene (0.1%)  
 δ-elemene‡ (1.2%)  
 longifolene† (0.3%)  
 β-cedrene† (0.1%)  
 allo-aromadendrene‡ (0.4%)

In 1998, Adegoke et al. reported on the comparative composition of oils of *Aframomum danielli* (Hook. f.) K. Schum. and *A. subulatum* Roxb.; however, the data reported for the *A. subulatum* oil was that of Gurudutt et al., even though none of the authors of the Adegoke et al. paper was an author of the Gurudutt et al. paper.

The following year, Atta-ur-Rahman et al. (1999) screened a number of oils of Pakistani spices for their composition and antifungal characteristics. The oil analyses were performed using both GC (FID) (for quantitation and retention indices) and GC/MS (for initial component identifications). The components identified in the oil of *A. subulatum* of Pakistani origin were as follows:

α-pinene (1.1%)	p-cymene (0.3%)
β-pinene (2.7%)	terpinolene (t)
myrcene (0.3%)	terpinen-4-ol (4.7%)
limonene (2.9%)	δ-terpineol (1.0%)
1,8-cineole (72.7%)	α-terpineol (13.3%)
γ-terpinene (0.4%)	(E)-nerolidol (0.5%)

t = trace (<0.1%)

Mohan Rao (2000) used GC/MS and retention indices to analyze a sample of *A. subulatum* oil of Indian origin. The oil composition was found to be as follows:

α-thujene (0.3%)  
 α-pinene (3.8%)  
 camphene (0.1%)  
 β-pinene (8.9%)  
 myrcene (1.2%)  
 p-cymene (0.1%)  
 limonene + 1,8-cineole (61.3%)  
 γ-terpinene (2.0%)  
 linalool (0.7%)  
 myrcen-8-ol (0.2%)  
 α-campholenic aldehyde (0.2%)  
 norbornyl acetate° (0.2%)  
 dihydrolinalool° (0.6%)  
 trans-pinocarveol (0.1%)  
 β-terpineol\* (0.2%)  
 δ-terpineol (1.6%)  
 terpinen-4-ol (1.3%)  
 α-terpineol (7.9%)

calamene<sup>\*</sup> (0.4%)  
 $\alpha$ -bisabolene<sup>\*</sup> (0.3%)  
 $\beta$ -bisabolene + nerolidol<sup>\*</sup> (0.3%)  
 t = trace (<0.1%)  
<sup>\*</sup> correct isomer not identified  
<sup>†</sup> identity requires confirmation  
<sup>‡</sup> incorrect identity based on elution order  
<sup>°</sup> does not exist naturally (identity incorrect)

- S. S. Nigam and R. M. Purohit, *Chemical examination of the essential oil derived from the seeds of Amomum subulatum Roxb.* Perfum. Essent. Oil Rec., 51, 121-123 (1960).
- S. S. Nigam and R. M. Purohit, *Chromatography of the essential oil of Amomum subulatum Roxb.* Indian Perfum., 5, 3-7 (1961).
- B. M. Lawrence, *Terpenes in two Amomum oils.* Phytochemistry, 9, 665 (1970).
- P. N. Gupta, A. Naqvi, L. N. Misra, T. Sen and M. C. Nigam, *Gas chromatographic evaluation of the essential oils of different strains of Amomum subulatum growing wild in Sikkim.* Parfum. Kosmet., 65, 528-529 (1984).
- S. Kaur, S. N. Naik and R. C. Maheshwari, *Composition of the essential oil of Amomum subulatum (Roxb.) (Cardamom large) isolated by liquid CO<sub>2</sub> extraction and steam distillation.* Indian Perfum, 37, 249-252 (1993).
- L-F. Zhu, L-F. Zhu, -H. Li, B-L. Li, B-Y. Lu and N-H. Xia, *Aromatic plants and essential constituents.* p. 193, South China Institute of Botany, Chinese

Academy of Sciences, Hai Feng Publish. Co. distributed by Peace Book Co., Ltd., Hong Kong, (1993).

- K. N. Gurudutt, J. P. Naik, P. Srinivas and B. Ravindranath, *Volatile constituents of large cardamom (Amomum subulatum Roxb.).* Flav. Fragr. J., 11, 7-9 (1996).
- G. O. Adegoke, L. J. Mohan Rao and N. B. Shankaracharya, *A comparison of the essential oils of Aframomum danielli (Hook. f.) K. Schum. and Amomum subulatum Roxb.* Flav. Fragr. J., 13, 349-352 (1998).
- Attar-ur-Rahman, M. I. Choudhary, A. Farooq, A. Ahmed, M. Z. Iqbal, B. Demirci, F. Demirci and K. H. C. Baser, *Antifungal activities and essential oil constituents of some spices from Pakistan.* 3rd Internat. Electronic Conf. Synth. Org. Chem. (ECSOD-3), p. 1-10, www.reprints.net/ecsoc-3.htm. September 1-30, (1999).
- L. J. Mohan Rao, *Quality of essential oils and processed materials of selected spices and herbs.* J. Med. Arom. Plant Sci., 22, 808-816 (2000). ■