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Comparison Between the Fresh and Dry Essential Oil of *Helichrysum faradifani* Scott Elliot from Madagascar

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Abstract

Essential oils of *Helichrysum faradifani* were isolated from fresh or dry plant parts by steam distillation with yields from 0.1-0.8%. Analysis of the oils by GC and GC/MS enabled the identification of 49 components, of which α-fenchene (trace-27.3%), β-caryophyllene (14.8-29.2%) and β-himachalene (15.7-36.6%) were most predominant.

Key Word Index

Helichrysum faradifani, Asteraceae, essential oil composition, α-fenchene, β-caryophyllene, β-himachalene.

Introduction

Helichrysum faradifani is one of the endemic *Helichrysum* in Madagascar. This species is widespread in the central and southeastern regions. The plant is used in Malagasy traditional medicine for wound-healing (1). Other endemic species of *Helichrysum* are also known for their pharmacological properties like diuretic, antidiabetic, stimulant, digestive and haemostatic agents (1,2) and their antibacterial activity are also reported (3,4). Even if many species of *Helichrysum* have been investigated (5-8), the chemical composition of *H. faradifani* oils has only been reported by Cavalli et al. (6). They found that *H. faradifani*, collected in the north of the region of Antananarivo, contained mainly β-caryophyllene (34.6%) and linalool (16.1%). Later research using dichloromethane extract of the leaves of this plant rather than a steam distilled oil reported seven compounds; two of which were new: 8-(3-phenyl-2-propen-1-one)-7-hydroxy-5-methoxy-2-dimethylbenzopyran and 2'-4'-6'-trihydroxy-3'-methoxydihydrochalcone (9).

Experimental

Plant material: Samples of the aerial parts of *H. faradifani* were manually collected from wild indigenous plants in the vicinity of Fianarantsoa in south central of Madagascar, 10 times over the course of one year, by the company EPAM ("Extraits et Produits Aromatiques de Madagascar"). *Helichrysum faradifani* is an annual plant. Its vegetative cycle depends on the stations and the climate where it grows (10). However, the climate being variable, in a very hot or a too-dry period, the plants tended to fade or dry, so that gathered plants are rather dry. Plants are sometimes fresh, sometimes partially dried naturally or artificially under sunshine. In the locality of Fianarantsoa, during the inter season (May and June) the weather was dry, so plants were rare. A voucher specimen has been deposited at the Herbarium of the Institut Malgache de Recherches Appliquées (IMRA – Botanical Department).

Analysis: The collected aerial plant parts (300-800 g) were subjected to steam distillation for 2 h. The oil yields ranged from 0.1-0.8%. Yellow oils were obtained from the aerial parts of the fresh or dried plant. GC analysis was carried out on an Instrument CE TOP 8000 chromatograph fitted with a fused-silica capillary column (25 m x 0.32 mm, 0.25 μm coated with CP-WAX 52 CB), with a programmed temperature gradient of 50°-240°C at 5°C/min. Flame ionization detector and injector temperatures were both set at 250°C, with hydrogen as carrier gas. Quantitative data were obtained from FID area counts without the use of correction factors.

GC/MS was performed on the same chromatograph fitted with a fused-silica capillary column Rtx-WAX (30 m x 0.25 mm, 0.25 μm) coupled to a quadrupole Finningan Trace MS, programmed under the above mentioned conditions and equipped with NIST libraries. Hydrogen was used as carrier gas and the

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Table I. Relative percentage composition of the oil from fresh wild *Helichrysum faradifani* collected at different periods from the region of Fianarantsoa, Madagascar

Compound	Sept. 00	Nov. 00	Feb. 01	Mar. 01	July 01	Aug. 01
tricyclene	0.8	t	0.5			0.7
α-pinene	0.7	0.7	0.7	1.3	1.1	0.4
α-fenchene	17.7	13.7	14.1	27.3	20.8	13.1
camphene	0.2	0.2	0.2	0.3	0.5	0.1
β-pinene	0.7	0.5	0.6	0.9	0.9	0.7
myrcene	0.8	0.7	0.8	0.2	0.6	0.5
α-terpinene	0.3	0.3	0.2		0.2	0.2
limonene	3.8	3.8	4.5	1.9	3.0	2.3
β-phellandrene	0.4	0.4	0.4	0.1	0.4	0.4
1,8-cineole	0.7	0.5	0.2	0.5		
γ-terpinene	0.5	0.5	0.5	0.1	0.4	0.4
(E)-β-ocimene	0.2	0.3	0.3		0.1	0.1
2-heptyl acetate	0.2	0.2	t	0.2	0.2	0.2
p-cymene	0.4	0.3	0.7	0.4	0.3	0.3
terpinolene	0.5	0.5	0.5	0.1	0.4	0.4
α-fenchyl acetate	0.3	0.4	0.5	0.2	0.2	0.3
α-copaene	0.8	1.3	0.9	0.8	1.9	0.8
α-cedrene	1.2	1.0	1.3	1.3	1.0	1.1
linalool	2.3	2.5	3.3	1.2	1.5	1.7
linalyl acetate		0.2	0.1	0.2	0.1	0.1
bornyl acetate	4.6	3.5	5.2	2.5	3.5	3.4
α-bergamotene[a]	0.3	0.5		0.5	0.6	0.5
α-fenchol	0.4	0.5			0.2	
β-caryophyllene	19.7	18.2	20.5	17.6	14.8	15.6
aromadendrene	t	0.2			0.1	
terpinen-4-ol	1.1	1.1	1.3	0.6	0.9	0.8
unknown[b]	2.8	2.8	3.9	2.6	2.7	3.7
allo-aromadendrene		0.2			0.2	
α-himachalene	0.2	0.3	0.2		0.3	0.2
α-humulene	1.6	1.9	1.7	1.5	1.2	1.4
(Z)-β-farnesene	0.4	0.6	0.7		0.3	0.8
methyl chavicol	0.2	0.3			0.3	
γ-muurolene	0.4	0.6	0.7	0.5	0.3	0.4
β-himachalene	25.4	26.3	17.1	15.7	27.2	32.8
α-terpineol	2.0	2.8	3.2[c]	0.9	3.1	1.8
borneol		0.2			0.4	0.1
β-selinene		0.3	0.2		0.2	0.2
γ-cadinene		0.4	0.3	0.1	0.2	0.2
zingiberene		0.2				
β-bisabolene	1.3	1.6	2.3	1.9	1.1	2.1
β-cadinene		0.3	0.1			0.1
geranial	1.4	1.8	1.1	0.9	1.5	1.7
δ-cadinene	1.0	1.2	0.9	1.0	2.0	0.7
ar-curcumene	2.2	2.3	5.6	7.9	2.1	2.5
geraniol				0.2		
caryophyllene oxide			1.0	1.2	t	0.2
cadinol[a]		0.4	0.2		0.2	
eudesmol[a]	0.6	0.6	1.0	0.4	1.0	1.8
bisabolol[a]	0.4	0.6	1.1	0.4	0.4	1.9
(Z,Z)-farnesol		0.1			t	

[a]correct isomeric form not identified; [b]unknown = 196(M+), 93 (100), 79(68), 41 (35), 55(15), 69(20), 107(19), 121(28), 136(10); [c]mix of α-terpineol and borneol; t = trace (<0.1%)

mass spectrometer operated in EI mode at 70 eV.

Results and Discussion

The identification of the compounds was based on their retention indices and on comparison of their mass spectral data with those obtained from authentic samples and/or NIST library as well as with literature data (11,12).

By means of these analyses more than 40 components of the oil of *H. faradifani* from Fianarantsoa were identified. The main compounds were β-himachalene (15.7-36.6%), β-caryophyllene (14.8-29.2%) and α-fenchene (trace-27.3%). The identified components from fresh plant and dried plant are listed in Table I and II, respectively.

The results were based primarily on the state of freshness of the distilled plants and not the seasonal variation because of the vegetative cycle of the plants. The results obtained illustrate that the oil composition between the fresh and dried

Table II. Relative percentage composition of the oil from naturally dried aerial plant parts of *Helichrysum faradifani* collected at different periods from the region of Fianarantsoa, Madagascar

Compound	Aug. 00	Oct. 00	Dec. 00	Mar. 01*	Apr. 01	Jul. 01*
tricyclene						
α-pinene				0.7		0.2
α-fenchene	0.4	t	0.4	1.8	0.1	0.7
camphene						
β-pinene				0.2		
myrcene				0.2		
α-terpinene				0.1		
limonene	0.2		0.6	2.2		0.3
β-phellandrene	0.2		0.2	3.0		t
1,8-cineole						
γ-terpinene				0.4		t
(E)-β-ocimene				t		
2-heptyl acetate						
p-cymène				0.2		
terpinolene				0.5		
α-fenchyl acetate	0.2		0.3	0.4	0.2	0.2
α-copaene	0.4	0.8	1.2	1.1	0.2	1.6
α-cedrene	1.5	1.2	1.2	1.2	1.8	0.9
linalool	1.3	0.1	0.8	2.6	0.8	0.7
linalyl acetate	0.2	0.3		0.2	0.1	0.1
bornyl acetate	2.3	0.3	2.7	5.1	1.7	1.5
α-bergamotene[a]	0.3	0.6	0.8	0.3	1	0.8
α-fenchol	0.3	0.2	0.8	0.5	0.4	t
β-caryophyllene	22.9	25.0	29.2	15.4	21.8	27.2
terpinen-4-ol	0.8	0.2	0.9	1.4	0.7	0.4
unknown[b]	3.8	1.2	4.3	4.8	3.9	2.1
allo-aromadendrene				0.1	0.2	0.2
α-himachalene	0.3	0.2		0.3	0.2	0.2
α-humulene	2.0	2.5	2.5	1.5	2.3	2.3
(Z)-β-farnesene	0.4	0.7	0.7	1.0	1.3	0.4
methyl chavicol		0.2				
γ-muurolene	0.4	0.2	0.6	1.1	0.6	0.4
β-himachalene	35.5	33.3	30.0	30.8	31.4	36.6
α-terpineol	2.4	2.9	3.5	6.1[c]	1.2	1.0
borneol	0.1	0.2	0.3		2.2	t
β-selinene		0.6	1.0	0.4	0.4	
γ-cadinene	0.2	0.7	0.2	t	0.3	0.3
zingiberene			0.2	t	0.3	
β-bisabolene	1.8	3.7	2.4	2.1	2.9	2.5
β-cadinene	0.1	0.3		0.2	0.3	
geranial	2.0	2.0	2.0	2.0	2.3	1.9
δ-cadinene	0.9	1.3	1.9	1.0	1.3	1.1
ar-curcumene	3.3	3.2	2.8	3.1	3.7	2.5
geraniol	0.1			0.1	0.1	
caryophyllene oxide	0.2	0.3	0.3	t	0.3	0.2
cadinol[a]	0.4	1.0			0.2	0.1
eudesmol[a]	2.7	2.8	4.2	2.1	4.1	3.4
bisabolol[a]	2.9	4.0	1.4	1.5	2.8	3.2
(Z,Z)-farnesol	0.6	0.3		t	0.5	

*aerial plants dried in sunshine after collecting (same material than fresh oil); [a]correct isomeric form not identified; [b]unknown = 196(M+), 93 (100), 79(68), 41 (35), 55(15), 69(20), 107(19), 121(28), 136(10); [c]mix of α-terpineol and borneol; t = trace (< 0.1%)

plants is quite different. We noted that the relative content of the α-fenchene depended on the state of freshness of the plant at the time of distillation. When the plant parts were fresh, the content of α-fenchene ranged between 13.1-27.3%. For partially fresh or dry plants, the content of α-fenchene was relatively low (trace-1.8%), while β-himachalene, cadinol, eudesmol and bisabolol increased. The comparison of the results of the samples of the oils obtained starting from fresh plants (March 2001 and July 2001) with the same samples dried in sunshine illustrated the fall of the volatile compounds contents, particularly α-fenchene, which passed from 27.3-1.8% for March 2001 samples, to 13.1-0.7% for July 2001 batch. The results also showed that the percentage of β-himachalene increased from 15.7-30.8% for the March 2001 samples, to 32.8-36.6% for the July 2001 samples.

The oil of *H. faradifani* was also characterized by ar-curcumene (2.1-7.9%), β-bisabolene (1.1-3.7%) and bornyl acetate (0.3-5.2%). Noteworthy is the observation that the chemical

composition of the oil of *H. faradifani* in this present study was rather different from that which was already published by Cavalli et al. because they did not detect any α-fenchene or β-himachalene.

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