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The Volatile Oil of Saudi *Pulicaria crispa*

Mohamed A. Al-Yahya, Aly M. El-Sayed, [1]Mahmoud M.A. Hassan and Ibrahim El-Meshal

Medicinal, Aromatic and Poisonous Plants Research Center, College of Pharmacy, King Saud University, P.O. Box 2457, Riyadh 11451, Saudi Arabia
[1]Al Ali General Hospital, P.O. Box 16110, Riyadh 11464, Saudi Arabia

ABSTRACT. The aerial parts of *Pulicaria crispa*, grown wild in Saudi Arabia, were subjected to steam distillation. Analysis of the combined GC/MS spectra of the volatile oil revealed several terpenoids. β-caryophyllene and its oxide were tentatively characterized as the major constituents along with other sesquiterpenoids not previously reported in this plant. The activity of the oil on several microorganisms was also explored.

The annual herb, *Pulicaria crispa* Sch. Bip. (syn. Francoeuria crispa Forssk., Cass.; compositae) is well recognized in the folk medicine of the central region of the Kingdom of Saudi Arabia by the name "gethgath". It is oftenly prescribed for treating inflammation and some times used as an insect repellent (Al-Yahya *et al.*, 1984). A previous study on the phytochemical screening gave evidence of several natural constituents of which flavonoids and volatile oil constituents are prevalent (Al-Yahya *et al.*, 1984). The cytotoxic principals of "gethgath" were established to be among the sesquiterpenoid mixture and 2 α-hydroxyalantolactone was isolated (Al-Yahya *et al.*, 1984). More recent experiments isolated the novel, 5, 6-epoxy-2-α-hydroxyalantolactone which was markedly cytotoxic on the 9KB cell culture (Al-Yahya *et al.*, 1988). Axillarin was found to be a potential anticarcinogenic flavone in "gethgath" using cultural hamster embryo cells exposed to benzo [α] pyrene (Al-Yahya *et al.*, 1988).

This investigation is a continuation of on going systematic study of medicinal plants of Saudi Arabia. It presents the combined GC/MS analysis of the volatile

materials and hence sheds light on their chemical composition. It demonstrates the inhibitory activity of the oil on some pathogenic organisms, not previously reported.

Experimental

A) Materials: The flowering aerial parts of *Pulicaria crispa* were collected from the central region of Saudi Arabia. Authentication of the collection was performed by Dr. Sultan ul-Abedin, College of Pharmacy, King Saud University. A voucher herbarium specimen is deposited at the Medicinal, Aromatic and Poisonous Plants Research Center, College of Pharmacy, King Saud University, Riyadh.

B) Methods: Distillation of the aerial parts of *P. crispa* was performed on freshly collected samples. The acid value and ester value were determined according to the B.P., 1980. Optical rotation was measured on a Perkin Elmer polarimeter 241 mc. The refractive index was determined on a Zeiss Abbe Refractometer.

GC/MS analysis was performed on a Finnigan Mat 5100 using a 25 M OV-1 glass capillary column. A satisfactory separation was achieved using the following GC parameters: oven temperature 250°C, injector temperature 275°C and column increasing temperature 50 to 275°C. The MS was operated with a 0.1 L split 100:1, EI mode, with mass 40 to 500 units. Unknown spectra were matched by computer[1] with a reference library of compounds on most of the largest peaks.

TLC separation was performed on silica gel PF254 precoated plates using toluene: ethylacetate, 92:8, as eluent. Visualization of spots was carried out with vanillin: sulfuric acid spray reagent aided with gentle heat on a hot plate.

The antimicrobial screening was performed according to Mitscher *et al.* (1972).

Results and Discussions

The steam volatile portion of *F. crispa* is a light-yellow oil possessing a characteristic faint aroma (yield, 0.05% v/w), specific gravity 0.9171, $[\alpha]_D^{25}$ + 6.636 and positive refractive index 1.48, acid number 13.60, ester number 132.34. It inhibited the pathogenic organisms *Staphyllococcus aureus* at 500 µg/ml and *Bacillus subtilis* at 250 µg/ml. However, marked resistance were observed with *Escherishia coli, Pseudomonas aeruginosa, Proteus vulgaris,* and *Candida albicans* at 2000 µg/ml.

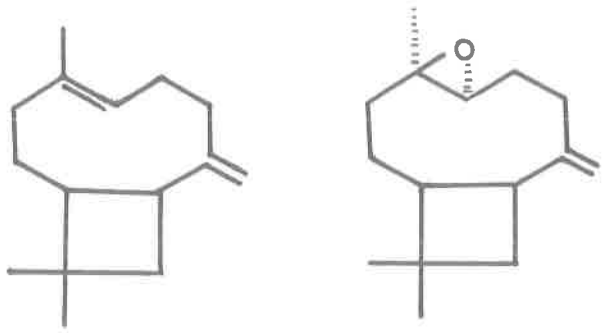
[1] Incos 2000 data system.

Separation of the oil components on TLC indicated several spots visualized as varing colors from pink to dark-brown. The major terpenoids were located at R_f 0.83, 0.69 and 0.34, respectively. The GC/MS data presented in Table 1 reveals the major constituents. β-caryophyllene [1] together with its oxide [2] (Bohlmann *et al.*, 1978) were present in 88.33 and 100% relative abundance in the sesquiterpenoid mixture, respectively.

Table 1. GC/MS Data of the major components of volatile oil of *Pulicaria crispa*

Compound No.	Percentage relative abundance	Molecular formula	Retention time	Assignment
1	12.7	$C_{10}H_{18}O$	4.1	Citronellal$^+$
2	40.5	$C_{10}H_{18}O$	4.5	Borneal$^+$
3	8.8	$C_{12}H_{20}O_2$	6.25	Linalyl acetate
4	21.12	$C_{10}H_{18}O$	6.5	Linalol$^+$
5	40.69	$C_{15}H_{24}$	13.45	β-maaliene
6	88.33	$C_{15}H_{24}$	14.48	β-caryophyllene
7	13.47	$C_{13}H_{22}O$	15.28	Geranyl acetone
8	10.42	$C_{15}H_{24}$	15.35	α-Humulene
9	28.35	$C_{15}H_{24}$	16.29	ɣ-Gurjuene
10	23.24	$C_{14}H_{24}O_2$	16.39	Geranylbutyrate
11	27.83	$C_{15}H_{24}$	17.08	ɣ-cadinene
12	8.68	$C_{15}H_{24}$	17.25	ɣ-cadinene
13	3.88	$C_{15}H_{24}O_2$	18.15	unknown M^+236
14	100%	$C_{15}H_{24}O$	18.39	β-caryophyllene oxide
15	13.01	$C_{15}H_{24}O$	19.09	unknown M^+220
16	25.51	$C_{15}H_{24}$	20.29	unknown M^+204
17	31.86	$C_{15}H_{24}O$	20.48	Santalol
18	10.74	$C_{15}H_{24}O$	21.02	unknown M^+220
19	7.1	$C_{15}H_{24}O$	21.06	unknown M^+220
20	14.81	$C_{15}H_{24}O$	21.18	unknown M^+220
21	15.39	$C_{15}H_{24}O$	21.59	unknown M^+220
22	6.18	–	22.51	unknown M^+246
23	4.18	–	23.40	unknown M^+248
24	6.52	$C_{15}H_{18}O_4$	25.15	unknown M^+262
25	2.83	$C_{15}H_{18}O_3$	25.39	unknown M^+248
26	4.1	$C_{15}H_{30}O$	26.21	E,E-Farnesylacetone
27	1.33	$C_{15}H_{20}O_4$	27.38	unknown M^+264
28	15.05	$C_{20}H_{40}O$	30.36	Phytol

+ Identity confirmed by MS and GC comparisons with authentic standard.



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Their structures along with the other oil components were assigned by computer matching with standard spectra in the data bank system and with those mass data published by Stenhagen *et al.*, 1975.

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دراسة الزيت الطيار المحضر من نبات الجثجاث السعودي

محمد عبدالعزيز اليحيى و علي محمد السيد و[1] محمود علي حسن
و ابراهيم المشعل

مركز أبحاث النباتات الطبية والعطرية والسامة ـ كلية الصيدلة ـ جامعة الملك سعود
ص. ب: ٢٤٥٧ الرياض ١١٤٥١
و[1] مستشفى العلي العام ـ ص. ب (١٦١١٠) الرياض ١١٤٦٤ ـ المملكة العربية السعودية

يعتبر نبات الجثجاث التابع للعائلة المركبة من النباتات الهامة في الطب الشعبي بالمملكة العربية السعودية فهو كثيراً ما يستخدم في علاج الالتهابات وأحياناً يستعمل في طرد الحشرات. وقد أثبتت الدراسات الكيميائية من قبل على هذا النبات أنه يحتوي على فلافونيدات وزيت طيار وكذلك على مواد سامة لها فاعلية على خلايا السرطان الخبيثة مثل مادة الفاهيدروكسي الانتولاكتون واكسيده. كذلك وجدت مادة الاكسيلارين التي لها خواص مانعه لنمو الخلايا الخبيثة على حيوانات التجارب المعروفة. ونظراً لأهمية هذا النبات كان موضوع دراسة هذا البحث هو الزيت الطيار الذي تم تحضيره بطريقة التقطير المائي المعروفة ووجد أنه يوجد بنسبة (٠٥, ٪) وله كثافة ٠٩١٧١, ، معدل دوران ضوئي موجب ٦,٦٣٦ وكذلك ثابت ضوئي موجب ١٠٤٨، رقم حامض ١٣,٦٠ ورقم استير ١٣٢,٣٤. كذلك تم دراسة فاعلية هذا الزيت الطيار على الميكروبات ووجد أن له تأثير مثبط على كل من الجراثيم الكروية بنسبة ٥٠٠ ميكروجرام/سم والميكروبات العصوية بنسبة ٢٥٠ ميكروجرام/سم. وكذلك على خمائر الكانديدا بنسبة ٢٠٠٠ ميكروجرام/سم. كما شمل البحث تحليل هذا الزيت بكروماتوجرافيا الغاز المتصلبه بمطياف الكتلة وتم معرفة عديد من مكوناته الكيمائيه مثل مادة البيتا ـ كاريوفيللين (٨٨ ٪) وبيتاكاريوفيللين اكسيد (١٠٠ ٪)، سترونيلال (١٢,٧ ٪) بورنيال (٤٠,٥ ٪) لينالیل اسيتات (٨,٨ ٪) لينالول (٦,٥ ٪) بيتاماليين (٤٠,٦٩ ٪) جيرانيل اسيتون

(٤٦, ١٣ ٪)، الفاهيميولين (٣٥, ١٥ ٪)، جاماجيرجين (٢٤, ٢٣ ٪) جاماكادينين (٨٣, ٢٧ ٪) دلتا كادينين (٢٥, ١٧ ٪) سانتالول (٨, ٣١ ٪) فارينسيل اسيتون (٠١, ٤ ٪)، فيتول (٠٥, ١٥ ٪) وعديد من التربينولات التي لم يكتشف بعد تركيبها البنائي ولكن التركيب الجزيئي لهذه المركبات يشير بأنها تتبع مجموعة السسكيتربين الواسعة الانتشار في عائلة هذا النبات. كذلك شملت دراسة هذا الزيت دراسة مقارنة لكل من السترونيلال، بورنيال، لينالول مع مركبات مرجعية للتأكد من وجودها مع المركبات الاخرى في هذا الزيت الطيار بطريقة كروماتوجرافيا الغاز. والجدير بالذكر بأن هذه أول مره يدرس فيها الزيت الطيار لنبات الجثجاث من الناحية الكيميائية ومعرفة تأثيره المثبط على العديد من الجراثيم المنتشره.