

Original Research Article

Essential Oil Composition of Two *Grammosciadium* DC Species, *G. platycarpum* (Boiss et Hausskn) Schischk and *G. macrodon* Boiss (Apiaceae), from Turkey

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Abstract

Purpose: To investigate and compare the essential oil composition of two *Grammosciadium* species obtained by hydrodistillation

Methods: The essential oil of the aerial parts of two species was obtained by hydrodistillation and analyzed by gas chromatography (GC) and gas chromatography - mass spectrometry (GC-MS).

Results: In *G. platycarpum*, twenty-seven compounds were identified representing 99.3 % of the total oil. The major constituents of the oil were limonene (28.4 %), β -pinene (16.1 %), β -selinene (15.9 %), δ -3-carene (11.2 %), α -farnesene (6.0 %) and *p*-cymene (4.7 %). The oil of *G. macrodon* contained thirty-seven components representing 94.4 % of the total oil, with caryophyllene oxide (15.9 %), germacrene - D (12.4 %), β -caryophyllene (11.5 %), δ -3-carene (10.1 %) and cyclohexane (4.6 %) as the main constituents.

Conclusion: The essential oil composition of *G. macrodon* was analysed for the first time, the most abundant constituents being caryophyllene oxide, and cyclohexane.

Keywords: *Grammosciadium macrodon*, *Grammosciadium platycarpum*, Essential oil, Limonene, Caryophyllene oxide, Germacrene - D, β -Caryophyllene, δ -3-Carene

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INTRODUCTION

Grammosciadium genus is belongs to the family Apiaceae (Umbellifereae). The family includes 97 genera and 402 species in the Flora of Turkey [1]. *Grammosciadium* was established by De Candolle as a new genus containing two species, *G. daucoides* DC. and *G. meoides* DC., in 1829 [2]. The members of genus *Grammosciadium* are distributed in Turkey and also in Transcaucasica, Iran and Armenia [1-5]. Generally, they occur in temperate or temperate cold pasture lands and are considered to be attractive animal feed. The genus occurs also in Iraq, *G. platycarpum* and grows naturally in the mountainous regions of the

country. It is a perennial plant with a very strong fragrance [6].

Essential oils are valuable natural products used as raw materials in many fields such as perfumes, cosmetics, aromatherapy, and food industry [7]. There is an increasing attempt to screen plants for studying the biological activities of essential oils from chemical and pharmacological investigations to therapeutic aspects [8-13]. The essential oil of members of *Grammosciadium* genus, have antibacterial activity [14,15], free radical scavengers and antioxidant activity [4-16]. *In vitro* antibacterial and antioxidant activity of the essential oil and its

two main components of *G. scabridum* Boiss growing wild in Iran, as well as the composition of its essential oil were studied [15].

The results of the essential oils of two *Grammosciadium* species will give some clues on the chemotaxonomic relationships of family and genus patterns. In the present study, we report the yield and chemical composition of the essential oil isolated from the aerial parts of *G. platycarpum* and *G. macrodon*. This is the first report on the essential oil composition of *G. macrodon* and for *G. platycarpum* from Turkey origin.

EXPERIMENTAL

Plant materials

The plant samples were collected from their natural habitats and identified by Mehmet Yavuz Paksoy. *G. platycarpum* (voucher specimen no. 7998 G) was collected from Muş (Turkey), Malazgirt, Kaz lake surroundings, step, 1800 m, on 25 June 2012, *G. macrodon* (voucher specimen no. 7985) was collected from Van (Turkey), Bahcesaray, Karabel geçiti, 2400 m, on 29 May 2012. The voucher specimens were kept in FUH (Firat University Herbarium).

Extraction of essential oils

Hundred grams of air-dried aerial parts of the plant materials were extracted to hydrodistillation using a Clevenger-type apparatus for 3 h for extraction of the essential oil.

Gas chromatography (GC) analysis

The essential oil was analyzed using HP 6890 GC equipped with and FID detector and an HP-5 MS column (30 m x 0.25 mm i.d., film thickness 0.25 µm) capillary column was used. The column and analysis conditions were the same as in GC - MS. The percentage composition of the essential oils was computed from GC - FID peak areas without correction factors.

Gas chromatography/mass spectrometry (GC-MS)

The oils were analyzed by GC - MS, using a Hewlett Packard system. HP- Agilent 5973 N GC - MS system with 6890 GC in Plant Products and Biotechnology Res. Lab. (BUBAL) in Firat University. HP-5 MS column (30 m x 0.25 mm i.d., film thickness 0.25 µm) was used with helium as the carrier gas. Injector temperature was 250 °C, split flow was 1 ml/min. The GC oven temperature was kept at 70 °C for 2 min.

and programmed to 150 °C at a rate of 10 °C/min and then kept constant at 150 °C for 15 min to 240 °C at a rate of 5 °C/min. Alkanes were used as reference points in the calculation of relative retention indices (RRI). MS were taken at 70 eV and a mass range of 35 - 425. Component identification was carried out using spectrometric electronic libraries (Wiley and Nist library).

RESULTS

The compounds identified in the oils of *Grammosciadium* sp. studied are listed in Table 1 in elution order from the HP-5MS column. The hydro distillation of the aerial parts of *G. platycarpum* and *G. macrodon* yielded 0.2 % and 0.4 % of pale yellowish oils, respectively. The extracted essential oils were complex mixture of non-terpenes, monoterpenes and sesquiterpenes; 49 components were identified out of which 15 are common to both oils (Table 1).

In the essential oil of *G. platycarpum*, the monoterpene hydrocarbons made up the high contribution (ca. 65 %) limonene being the most abundant compound. In contrast, this fraction was reduced ca. 20 % in the essential oil of *G. macrodon* and caryophyllene oxide (15.9 %) was assigned as the main sesquiterpene.

In this study, twenty-seven components in the essential oil of *G. platycarpum* were identified. The most abundant constituents were limonene (28.4 %), β-pinene (16.1 %), β-selinene (15.9 %), δ-3-carene (11.2 %), α-farnesene (6.0 %) and p-cymene (4.7 %) (Table 1). In the oil of *G. macrodon*, thirty-seven components were characterized representing 94.4 % of the total oil, with caryophyllene oxide (15.9 %), germacrene - D (12.4 %), β-caryophyllene (11.5 %), δ-3-carene (10.1 %) and cyclohexane (4.6 %) as the main constituents (Table 1). Both oils revealed limonene (28.4 %, 3.5 %), β-pinene (16.1 %, 2.8 %), δ-3-carene (11.2 %, 10.1 %), p-cymene (4.7 %, 2.5 %) and spathulenol (2.7 %, 3.8 %) as the main and common constituents in both species essential oils respectively (Table 1). α-farnesene (6.0 %), undecane (2.3 %), 2,6,10 - dodecatrien-1 - ol, 3,7,11 - trimethyl (1.2 %) and 6 methyl - 5 - hepten - 2 - one (1.1 %) were another major components in the *G. platycarpum* oil but they were not detected in the *G. macrodon* oil. Germacrene D (12.4 %), cyclohexane (4.6 %), cis - calamenene (3.5 %), caryophyllene II (2.3 %), isolongifolene (1.7 %), dodeconoic acid (1.6 %), salvia - 4(14) - en - 1 - one (1.3 %), 1,4-methanazulene (1.2 %) and α-humulene (1.0 %) were only determined in the *G. macrodon* oil.

Table 1: Constituents of the essential oils of (a) *G. platycarpum* and (b) *G. macrodon*

No	Compound	KI	Concentration	
			a	b
1	Nonane	996	0.2	--
2	α -pinene	1021	0.5	1.5
3	Verbenone	1039	0.2	--
4	Bicyclo(3,1,0)hex-2-ene	1052	--	0.9
5	β -pinene	1055	16.1	2.8
6	6 methyl-5-hepten-2-one	1060	1.1	--
7	β -myrcene	1064	0.8	0.6
8	Octanal	1075	0.9	--
9	l-phellandrene	1078	--	0.2
10	p-cymene	1091	4.7	2.5
11	Limonene	1095	28.4	3.5
12	Cis-ocimene	1100	2.3	0.2
13	δ -3-carene	1108	11.2	10.1
14	γ -terpinene	□□□□	1.2	0.2
15	Undecane	1148	2.3	--
16	Cyclohexene	1157	--	0.9
17	2-Cyclohexen-1-one	1210	--	0.3
18	2,6-dimethyl-1,3,5,7-octatetraene	1223	--	0.3
19	2-decanal	1263	0.3	0.2
20	Acetic acid	1283	--	0.2
21	α -copaene	1360	--	0.7
22	2-venyl-2-cyclopent-1-ol	1364	0.3	--
23	Cyclohexane	1371	--	4.6
24	β -Caryophyllene	1394	0.2	11.5
25	β -cubebene	1400	--	0.3
26	1,6,10-dodecatriene	1416	0.3	4.5
27	α -humulene	1418	--	1.0
28	Germacrene D	1436	--	12.4
29	β -selinene	1441	15.9	0.3
30	Bicyclogermacrene	1445	0.7	0.6
31	α -farnesene	1449	6.0	--
32	Germacrene A	1453	--	0.9
33	Cis-calamenene	1460	--	3.5
34	2,6,10-dodecatrien-1-ol, 3,7,11-trimethyl	1465	1.2	--
35	Sesquirofuran	1476	0.2	--
36	Cis-farnesol	1478	0.6	--
37	Dodeconoic acid	1484	--	1.6
38	1,5-epoxysalvial-4(14)-ene	1490	--	0.6
39	Spathulenol	1495	2.7	3.8
40	Caryophyllene oxide	1498	0.5	15.9
41	Salvial- 4(14)-en-1-one	1504	--	1.3
42	Isolongifolene	1518	--	1.7
43	Azulene	1526	0.2	--
44	α -cadinol	1539	0.3	--
45	1,4-Methanazulene	1541	--	1.2
46	Caryophyllene II	1548	--	2.3
47	Cyercene	1558	--	0.8
48	Piperitenone	1598	--	0.2
49	1-deoxycapsidol	1621	--	0.3
Total (%)			99.3	94.4

Note: KI, Kovatz index

DISCUSSION

Regarding the quantitative terpenoid composition, higher amounts of monoterpenoids in comparison with sesquiterpenoids are present in the essential oil of fruit, while in the leaf essential oil, sesquiterpenoids are the main group in *G. platycarpum* [4] from Iran. In another report it is declared that, the monoterpenes made up the higher contribution in *G. platycarpum* fruit essential oil from Iran [18]. However, it is sometimes usual to have such variations in the composition of the essential oils of plants resulting from geographical differences, growing conditions and variations in climate.

A review of the chemical constituents of *Grammosciadium* showed that the phytochemical composition of the genus has been addressed by a few studies in the past. Sonboli *et al* [14] reported that the essential oil from the whole aerial parts of *G. platycarpum* consisted mainly of linalool (79.0 - 81.8 %) and limonene (5.8 - 10.0 %). Also, Sonboli *et al* [15] analyzed the volatile constituents of the aerial parts of *G. scabridum*. This oil was characterized by high amounts of δ -terpinene (73.5 %), p - cymene (14.2 %), and (E) - β - farnesene (5.3 %). Nickavar *et al* [18] reported that the major components of the hydrodistilled essential oils obtained from dried leaves and fruits of *G. platycarpum* were linalool (26.1 and 53.9 %), (E,E) - α - farnesene (24.1 and 20.4 %) and (Z) - β - santalol (10.6 and 10.9 %), respectively.

There are some differences between this study and the previous investigation on the essential oil of *G. platycarpum*. Based on the present study and that of Sonboli *et al* [14], limonene was found as the main constituent. Moreover, α -farnesene, the major compound of *G. platycarpum* was also reported as the predominant compound by Nickavar *et al* [18]. The major compound, limonene, present in both species' essential oils has also been reported to exist in *G. scabridum* [19]. Some similarities were found in view of the major essential oil compounds within the same family (Apiaceae) patterns. In the essential oil of *Artemisia squamata*, α - pinene (57.8 %), camphene (9.0 %), β - myrcene (5.7 %), δ -3-carene (5.3 %) and limonene (5.3 %) were determined as the major constituents and also butanoic acid (37.1 %), caryophyllene oxide (13.4 %), germacrene D (10.0 %) and β - caryophyllene (5.4 %) were determined as major compounds in the essential oil of *Malabaila secacul* [20]. The majority of some common compounds in the essential oils may supply some contributions to the chemotaxonomy of the family patterns.

CONCLUSION

There are some qualitative and quantitative differences between both plant essential oils and *G. platycarpum* from other countries. This study demonstrates the occurrence of limonene/ β -pinene chemotype of *G. platycarpum* and caryophyllene oxide/germacrene D chemotype of *G. macrodon* in Anatolian Region of Turkey. Members of the genus *Grammosciadium* are among the most important aromatic plants and the essential oils of this genus have great commercial value.

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