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Original Article

Chemical Analysis of Essential Oils from Different Populations of *Ferulago angulata* subsp. *carduchorum* in Iran

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Abstract

Ferulago angulata subsp. *carduchorum* (Boiss. & Hausskn.) D.F. Chamb is a perennial species of the family Apiaceae and is locally used as a flavouring agent and for some medicinal properties. The essential oil of this plant if found to possess antibacterial and antifungal activities. The present study is aimed to determine constituents of essential oils from inflorescence of four *F. angulata* subsp. *carduchorum* populations. Air-dried samples were subjected to hydrodistillation using a Clevenger-type apparatus and extracted oils were chemically analyzed by means of GC and GC-MS. Results: With the average of 1.99% (w/w), the oil content of studied populations varied from 0.86% (Dalahoo) to 2.39% (Azgale). All oils were characterized by the high amounts of monoterpene hydrocarbons (71.1-83.6%), and the low levels of sesquiterpene hydrocarbons (2.0-7.8%) and especially oxygenated ones (absent to 3.5%). *α*-Pinene (7.1-29.8%), (*Z*)-*β*-ocimene (14.7-45.9%), *allo*-ocimene (5.9-16.7%), *γ*-terpinene (2.0-12.2%) and bornyl acetate (3.0-7.3%) were found to be the principal volatiles of populations in question. There were significant differences in the chemical composition of studied populations, which, considering constituents of an essential oil determine its flavor and biological activities, enabled selection of favored populations for use in different industries.

Key words: *Ferulago angulata* subsp. *carduchorum*, Essential oils, *α*-pinene, (*Z*)-*β*-ocimene

Introduction

Growing awareness of residual toxicity or potential side effects of chemical preservatives have resulted in increasing pressure on food industry to reduce or eliminate the use of these synthetic compounds. Among other alternative methods, essential oils or their isolated components have received a special interest to develop as natural preservatives for extension of food product's shelf life and maintenance of their quality [1-3]. In fact, natural origin, known antioxidant and antimicrobial activities and reduced risk of microbial resistance [2,4-6] are special properties of these aromatic liquids that make them promising for this purpose. Due to their therapeutic and preservative properties and pleasant tastes and aromas, essential oils are also intensely screened and used in medicine, perfumery, cosmetics etc. in addition to food flavoring and preservation. It has been shown that composition of essential oils and relative concentrations of oil components, and so their biological properties, vary among species and relate to the factors such as geographical origin, season, genetic background, type of plant organ, environmental conditions and stage of development [7-9].

Ferulago W.D.J. Koch. is a member of Apiaceae (Umbelliferae) family and comprises approximately 35-40 species of herbaceous plants with wide distribution in the temperate zone of both hemispheres, especially in Central Asia and Mediterranean area [10,11]. Among other groups of secondary metabolites, the plants have been

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reported to contain essential oil with significant biological activities [10,12-19]. In traditional systems of medicine, some *Ferulago* species are used as sedative, anti-parasitic, tonic, digestive and aphrodisiac and for the treatment of intestinal worms, hemorrhoids, ulcers, snake bites, headaches and diseases of the spleen. In addition, incision of the roots of several *Ferulago* species yields a gum, which is used as a spice and for flavouring purposes [11,12, 20].

Seven species of the genus *Ferulago* are found in the flora of Iran, of which *F. contracta* Bois. & Husskn. ex Boiss. *F. phialocarpa* Rech.f. & Riedl and *F. angulata* Boiss. subsp. *carduchorum* (Boiss. & Hausskn.) (syn. *F. Carduchorum* Boiss. & Hausskn.) are endemic [21,22]. The latest is a perennial species and naturally occur in west and south of the country including Kurdistan, Kermanshah, Isfahan, Bakhtiari, Luristan, Kerman and Fars provinces [21]. The plant is locally used as a flavouring agent and for some medicinal properties. Previously, the plant has been the subject of phytochemical investigations and *α*-phellandrene, *β*-phelandrene, (*Z*)-*β*-ocimene, terpinolene, *α*-pinene, *α*-phellandrene, *β*-phellandrene and germacrene D were identified as the most abundant components of its aerial parts and seeds essential oils [19,20,23]. These oils were found to possess antibacterial and antifungal activities in a previous study [20]. To the best of our knowledge, there is no comprehensive investigation concerning the essential oil composition of different populations of this aromatic plant and, therefore, the present study was aimed to identify components of the essential oils of *F. angulata* subsp. *carduchorum* plants collected from four different locations.

Materials and Methods

Plant Materials

Ferulago angulata subsp. *carduchorum* plants of different origins were collected at the full flowering stage from Ghalage, Kerend, Dalahoo and Azgale, Kermanshah province, Iran (Table 1) in June to July 2010. Plant materials were identified at the Herbarium of Research Institute of Forests and Rangelands, Tehran, Iran. Voucher specimen (Number 3054) has been deposited at Herbarium of Agriculture and Natural recourses Research Center of Kermanshah.

Oil Extraction Procedure

Fifteen grams of inflorescences of each population were individually air-dried in the shade at the room temperature, roughly ground to a fine powder and then subjected to hydrodistillation using a Clevenger-type apparatus for 3 h. Resulted oils were dried over anhydrous sodium sulfate and stored in tightly closed vials at 4 °C before chemical analyses.

GC/MS Analysis

GC analysis was performed using a Younglin Acm 600 gas chromatograph equipped with a flame ionization detector (FID) and a HP-5MS fused silica column (30 m × 0.25 mm i.d.; film thickness 0.25 µm). Helium was used as the carrier gas at a flow rate of 0.8 ml/min. The injector and detector temperatures were kept at 290 °C. Oven temperature program was kept at 50 °C for 5 min, then raised to 240 °C at the rate of 3 °C/min then raised to 300 ºC with the ramp of 15 ºC/min and held isothermally for 3 min. GC–MS analysis was carried out by an Agilent 6890 gas chromatograph equipped with the same column as mentioned above and coupled with an Agilent 5973 mass spectrometer. Helium was used as the carrier gas at a flow rate of 0.8 ml/min in split ratio of 1:25. Ionization voltage was 70 eV.

Table 1 Some ecological features of the studied populations of *F. angulata* subsp. *carduchorum*.

Descriptions	Galage	Kerend	Dalahoo	Azgle
Longitudes (E)	46º 15′ 57″	46º 14′ 29.2″	46º 4′ 42.9″	46º 54′ 8.3″
Latitudes (N)	34º 2′ 57″	34º 11′ 36.4″	34º 37′ 45.4″	34º 44′ 18″
Altitudes (m)	1710	1812	1797	968
Collection dates	2010.Jul.1	2010.June.30	2010.Jul.15	2010.Jul.26
Soil pH	7.8	7.2	7.9	8
Soil texture	Clay	Clay-Sandy	Clay-Sandy	Sandy-Clay
OC%	1.44	1.08	1.14	0.97
TNV%	33	39	32	37
EC (ds/m)	0.69	0.79	0.52	0.67

Ion source and interface temperatures were 220 °C and 290 °C, respectively. The oven program temperature was the same as GC analysis.

Individual components were identified by comparison of their mass spectra with internal mass spectra library (Wiely 7n.1) and experimental retention indices (RI), which were calculated for all volatile constituents by using a homologous series (C_8 to C_{30}) recorded under the same operating conditions, and comparison with literature data (Table 2) [24].

Results and Discussion

In the present study, the oil content of populations in question varied from 0.86% (Dalahoo) to 2.4% (Azgale), with the average of 1.99% (w/w) (Table 2). In a previous study, the oil content of aerial parts of this plant was reported to be 4.0% (w/w), based on its dry weight [23]. In a previous study [20], the essential oil yield of water distilled aerial parts and seeds of the plant have been reported to be 0.63% and 3.2% (v/w), respectively.

GC and GC-MS analyses resulted in the identification of 37 components in the studied essential oils, constituting about 94 to 98.5% of their composition (Table 2). As shown, monoterpene hydrocarbons constituted considerable portion of all essential oils with the highest (83.55%) and the lowest (71.05%) amounts in the oils from Ghalajeh and Dalahoo populations, respectively. *α*-pinene (7.1-29.8%), (*Z*)-*β*-ocimene (14.7-45.9%), *allo*-ocimene (5.9-16.7%) and *γ*-terpinene (2.0-12.2%) were identified as the main components of this group of metabolites. Oxygen-containing monoterpenes were another major fraction of our oil samples and constituted about 8.7% (Azgale) to 14.8% (Kerend) of their composition with bornyl acetate (3.0-7.3%) as the major compound. On the other hand, essential oils of studied populations contained only negligible amounts of sesquiterpenes, especially oxygenated ones. Of them, *α*-curcumene (1.3-6.0%) was the most abundant component with the highest level in plants collected from Dalahoo.

The dominance of monoterpene hydrocarbons in the essential of aerial parts of *F. carduchorum* with (*Z*)-*β*-ocimene (21.2%), terpinolene (13.1%), *α*-phellandrene (12.7%) and *β*-phellandrene (10.9%) as the main constituents has also been reported [23]. In another study, *cis*-ocimene (27.9%), *α*-pinene (25.7%) and germacrene-D (22.3%) were identified as the major volatile components of aerial parts of this plant, the latest was not detected in our essential oils at all [20].

In a study on the chemical composition of different parts of *F. angulata* subsp. *carduchorum* plant, *α*-phellandrene (18.14-27.24%), *α*-pinene (12.2-21.2%), *β*-phellandrene (15.8-16.6%) and *p*-cymene (10.3-17.7%) constituted the major portion of essential oils from stems, leaves and flowers [25]. In addition, (*Z*)-*β*-ocimene (35.5%), terpinolene (5.7%) and *α*-phellandrene (5.4%) have been reported as the most abundant volatiles of *F. angulata* aerial parts [10].

According to results of our study and those reported on *F. angulata* plants, in addition to oil content, there are significant differences in the chemical profile of analyzed essential oils from the quantitative and qualitative points of view. Differences observed may be resulted from several factors such as type of plant parts tested, harvesting time, seasonal variation, developmental stages, geographical source, climatic conditions, genetic background and type of plant subspecies.

In another research, reported that the major components of the essential oil of *.Ferulago angulata* Boiss. from Iran were Suberosin (12.36%), Spathulenol (10.9%), *trans*-β-Caryophyllene (7.32%), arcurcumene (7.07%), and bicyclogermacrene (6.96%) [26].

In the present study, the squared Euclidean distance matrix resulted from the percentage composition of essential oils was subjected to hierarchical cluster analysis using the ward's method and SPSS 16 software (SPSS Inc., Chicago, IL) to classify our populations chemically. Based on the generated dendrogram, populations in question formed two main groups (Fig. 1). The first group (A) included populations from Kerend and Azgale, whereas the second group (B) comprised of Dalahoo and Ghalaje populations. Members of the first group were characterized by the high amounts of *cis*-*β*-ocimene (28.8-45.9%) and *allo*-ocimene (10.7-16. 7%) and the low levels of *γ*-terpinene (2.0-4.6%). On the other hand, essential oils from Dalahoo and Ghalaje populations contained a high amount of *α*-pinene (29.4-29.8%) and a low level of *cis*-*β*-ocimene (14.7-14.8%).

Table 2 Chemical composition of essential oils from inflorescences of different *F. angulata* subsp. *carduchorum* populations

No.	Component	RI*	Percentage			
			Kerend	Dalahoo	Ghalaje	Azgale
1	*α*-pinene	932	7.1	29.8	29.4	25.0
2	Camphene	943	0.3	2.3	1.2	2.7
3	Sabinene	969	0.5	tr	Tr	0.2
4	*β*-pinene	972	0.9	2.6	2.6	2.0
5	Myrcene	989	2.1	2.9	4.3	2.8
6	*α*-phellandrene	1002	0.9	-	-	0.6
7	*p*-cymene	1022	tr	2.9	3.7	1.9
8	Limonene	1026	0.1	2.0	5.2	2.1
9	*cis*-*β*- ocimene	1043	45.9	14.7	14.8	28.8
10	*trans*-β-ocimene	1049	tr	2.1	3.0	1.5
11	*γ*-terpinene	1058	2.0	5.3	12.2	4.6
12	*α*-terpinolene	1086	2.1	0.5	0.9	0.8
13	Linalool	1099	0.3	0.2	tr	0.4
14	*allo*-ocimene	1129	16.7	5.9	6.4	10.7
15	*cis*-verbenol	1140	1.6	0.6	0.8	0.4
16	*trans*-verbenol	1150	4.7	1.2	5.1	1.8
17	Borneol	1169	0.9	0.3	1.0	0.4
18	*p*-cymene-8-ol	1187	0.4	0.1	0.3	0.1
19	(*E*,*E*)-2,6-Dimethyl-3,5,7-octatriene-2-ol	1203	tr	tr	0.2	tr
20	Verbenone	1208	0.4	tr	0.1	0.3
21	*β*-cyclocitral	1215	0.1	0.2	0.1	tr
22	bornyl acetate	1290	6.2	7.3	3.0	4.8
23	*trans*-Pinocarvyl acetate	1297	0.2	0.3	0.7	tr
24	*trans*-carvyl acetate	1336	tr	0.1	0.2	tr
25	*β*-elemene	1391	0.2	0.4	0.2	0.3
26	*α*-cedrene	1411	0.3	1.0	0.4	Tr
27	*trans*-*β*-farnesene	1455	0.2	0.3	tr	0.2
28	*α*-curcumene	1481	1.3	6.0	1.8	2.4
29	Germacrene B	1556	tr	0.2	0.2	0.1
30	Geranyl butanoate	1574	tr	0.1	0.4	-
31	*α*-copaene-8-ol	1577	0.1	0.3	-	-
32	Geranyl isovalerate	1600	tr	1.2	0.6	0.6
33	*α*-cadinol	1652	0.4	3.0	-	0.2
34	*epi*-*β*-bisabolol	1668	tr	-	-	0.2
35	*α*-bisabolol	1681	-	0.2	-	-
36	Palmitic acid	1961	-	tr	-	-
37	Suberosin	2207	-	0.1	-	0.4
-	Monoterpene hydrocarbons	-	78.5	71.1	83.6	83.5
-	Oxygenated monoterpenes	-	14.8	11.6	12.4	8.7
-	Sesquiterpene hydrocarbons	-	2.0	7.8	2.6	3.0
-	Oxygenated sesquiterpenes	-	0.6	3.5	-	0.4
-	Other compounds	-	-	0.1	-	0.4
-	Total identified	-	95.8	94.1	98.5	95.9
-	Oil content (%, w/w)	-	1.7	0.9	3.01	2.4

* Retention indices relative to C_8-C_{30} n-alkanes on the HP-5 column; tr, trace (<0.1%)

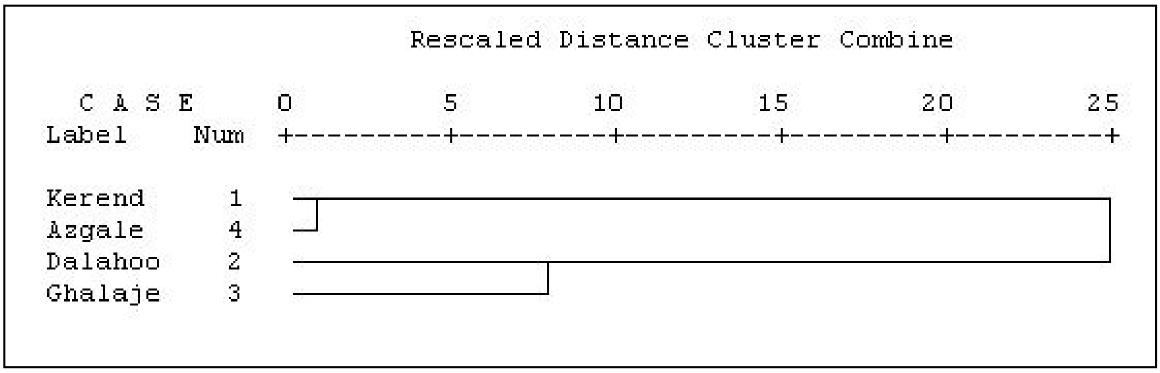

Fig. 1 Dendrogram constructed by cluster analysis of data from chemical composition of five different *F. angulata* subsp. *carduchorum* populations

Conclusion

There were significant differences in the chemical composition of studied populations, which, considering constituents of an essential oil determine its flavor and biological activities, enabled selection of favored populations for use in different industries.

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