Nat. Volatiles & Essent. Oils, 2021; 8(1): 39-48
DOI: 10.37929/nveo.871951

NVEO Natural Volatiles & Essential Oils

RESEARCH ARTICLE

The Essential Oil Profiles of *Chaerophyllum crinitum* and *C. macrospermum* Growing wild in Turkey

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Submitted: 31.01.2021; Accepted: 18.02.2021

Abstract

The current study reports the essential oil compositions of the aerial parts of *Chaerophyllum crinitum* Boiss. and *Chaerophyllum macrospermum* (Willd. ex Spreng.) Fisch. & C.A. Mey. ex Hohen. collected from Bitlis and Hakkari in Turkey, respectively. The essential oils, which were obtained with water distillation were analyzed by GC and GC-MS, simultaneously. The essential oil of *C. crinitum* were characterized with (*E*)-β-ocimene (38.1%) and terpinolene (12.7%), while terpinolene (21.4%), myristicin (18.9%), *p*-cymen-8-ol (11.9%) were identified as major components for *C. macrospermum* essential oil.

Keywords: *Chaerophyllum crinitum, Chaerophyllum macrospermum*, Apiaceae, essential oil, (*E*)-β-ocimene

Introduction

Apiaceae is a large family consisting of 3780 species in 434 genera. Members of Apiaceae are mainly distributed in northern hemisphere. Species belonging to the Apiaceae family are of economic importance and are used in industrial processes such as beverage, perfumery, pharmaceutical, and cosmetic industries (Zengin et al., 2020).

The genus *Chaerophyllum* L. has economic value-added species. In the Flora of Turkey, the genus *Chaerophyllum* L. is represented by 15 species of which four are endemic (Davis, 1972). “Chairo” (to please) and “phylum” (a leaf) words from Greek consist of *Chaerophyllum* and *Chaerophyllum* means the fragrant character of the foliage (Ebrahimabadi et al., 2010). *Chaerophyllum* species are used as aromatizer in food industry. In Turkey, *C. macropodum* Boiss. is used in cheese production for its strong aroma (Demirci et al., 2007, Tarakci et al., 2006). *C. macropodum*, (local name: mendi, mendo), is wildly grown in Van and is used in production of very famous cheese known as “Van Otlu Peynir” (Çelik et al., 2008; Tarakci & Temiz, 2009).

Chaerophyllum species were characterized with lignans, polyacetylenes, essential oils, phenolic acids, and flavone derivatives (Rollinger et al., 2003; Gonnet, 1985; Dall’Acqua et al., 2004; Ayala Garay et al., 2001).

Essential oils of *Chaerophyllum* species have been determined due to their economic values. Previous studies reported that *C. crinitum* was characterized with (*E*)-β-ocimene (50.5%) by Nematollahi et al. (2005); α-terpinolene (20.3%) by Hayta and Celikezen (2016) while *C. macrospermum* seed essential oil was characterized with (*E*)-β-farnesene (27.1%), (*Z*)-β-ocimene (18.8%), *p*-cymene (14.3%), α-fenchyl acetate (12.7%) (Razavi & Nejad-Ebrahimi (2010)). The crushed fruits of *C. aksekiense* an endemic to Turkey were characterized with heptacosane (10.1%) by Başer et al. (2010). β-Phellandrene (17.6%) and limonene (15.9%) were found as major compounds in the essential of *C. libanoticum* growing wild in Turkey (Demirci et al., 2007).

In Turkey, due to the use of *Chaerophyllum* species as food ingredients, these species were evaluated for their potential biological effects, mostly antimicrobial and antioxidant activities (Ağaoğlu et al., 2005; Durmaz et al., 2006; Sagun et al., 2006; Hayta and Celikesen, 2016; Kurkcuoglu et al., 2018).

In the literature, a few studies on *Chaerophyllum* species growing wildly in Turkey were reported until now. Furthermore, the volatile composition of these species is very important because of the uses as food and as foodstuff. The present work reported that the essential oil compositions from aerial parts from both *Chaerophyllum crinitum* and *C. macrospermum* obtained by water distillation.

Materials and Methods

Plant materials

Chaerophyllum crinitum was collected from Bitlis; Adilcevaz, Armutlu Village, Yeşilce Area at an altitude of 2140 m in Turkey. *C. macrospermum* was gathered from Hakkari; Cilo Mountain, between Kırıkdağ Village- and Cehennem Dere Plateau at an altitude of 2500 m in Turkey. Both of the samples are kept in Anadolu University, Pharmacy Faculty, Pharmacognosy Laboratory.

Isolation of essential oils of the samples

The essential oils from aerial parts of *Chaerophyllum crinitum* and *C. macrospermum* were isolated by water distillation for 3 h using a Clevenger-type apparatus.

GC and GC-MS analysis

The methods for GC (Agilent 6890N GC system) and GC-MS analysis (Agilent 5975 GC-MSD system) were given by our previous study (Duymuş et al., 2014).

Identification of components

Individual components were identified by computer matching with commercial libraries (Wiley GC-MS Library, MassFinder 3 Library) and in-house "Baser Library of Essential Oil Constituents", which includes over 3200 authentic compounds with Mass Spectra and retention data from pure standard compounds. In addition, components of known oils as well as MS literature data, were also used for the identification (Duymuş et al., 2014).

Result and Discussion

The aerial parts of *Chaerophyllum crinitum* and *C. macrospermum* were subjected to water distillation using Clevenger apparatus for 3 h. Each essential oil was analyzed by GC and GC-MS systems, simultaneously.

The essential oil of *C. crinitum* obtained by lower than 0.1% yielding was characterized with sixty compounds comprising 95.8% of the oil. *C. macrospermum* essential oil were obtained by 0.1% yielding from the aerial parts. Thirty-eight components were identified comprising 94.1% of total oil. The essential oil compositions of both samples were given at Table 1.

The main constituents of the essential oil of *C. crinitum* were found as (*E*)-β-ocimene (38.1%), terpinolene (12.7%), α-pinene (5.5%), *p*-cymene (5.3%), limonene (5.3%) and *p*-cymen-8-ol (2.2%). Terpinolene (21.4%), myristicin (18.9%), *p*-cymen-8-ol (11.9%), limonene (6.1%), α-pinene (4.6%), spathulenol (4.4%), and α-*p*-dimethyl-styrene (3.6%) were the major compounds for *C. macrospermum* essential oil.

Table 1. The essential oil compositions of *Chaerophyllum crinitum* and *C. macrospermum*

RRI	**Compounds**	***C. crinitum* (%)**	***C. macrospermum* (%)**	**IM**
1032	α-Pinene	5.5	4.6	t_R, MS
1035	α-Thujene	0.1	-	t_R, MS
1076	Camphene	0.4	-	t_R, MS
1093	Hexanal	0.1	-	t_R, MS
1118	β-Pinene	1.7	0.4	t_R, MS
1132	Sabinene	1.8	-	t_R, MS
1174	Myrcene	0.5	0.5	t_R, MS
1176	α-Phellandrene	0.1	0.2	t_R, MS
1188	α-Terpinene	0.4	-	t_R, MS
1194	Heptanal	tr	-	t_R, MS
1203	Limonene	5.3	6.1	t_R, MS
1218	β-Phellandrene	1.2	1.7	t_R, MS
1246	(*Z*)-β-Ocimene	0.4	0.7	MS
1255	γ-Terpinene	1.1	0.2	t_R, MS
1266	(*E*)-β-Ocimene	38.1	2.4	MS
1280	*p*-Cymene	5.3	1.4	t_R, MS
1290	Terpinolene	12.7	21.4	t_R, MS
1296	Octanal	1.7	0.2	t_R, MS
1398	2-Nonanone	0.3	-	MS
1400	Nonanal	0.2	-	t_R, MS
1413	Rose furane	0.3	-	MS
1452	α-*p*-Dimethylstyrene	0.9	3.6	MS
1477	4,8-Epoxyterpinolene	0.8	1.6	MS
1482	Fencylacetate	0.3	-	t_R, MS
1497	α-Copaene	0.2	-	MS
1498	(*E*)-β-Ocimene epoxide	0.1	-	MS
1548	(*E*)-2-Nonenal	0.2	-	MS
1586	Pinocarvone	0.2	-	t_R, MS
1590	Bornyl acetate	1.3	-	t_R, MS
1611	Terpinen-4-ol	0.2	-	t_R, MS
1612	β-Caryophyllene		0.8	t_R, MS
1648	Myrtenal	0.2	-	MS
1655	(*E*)-2-decenal	0.8	-	MS
1668	(*Z*)-β-Farnesene	0.3	-	MS
1670	*trans*-Pinocarveol	0.2	-	t_R, MS
1684	*trans*-Verbenol	0.2	-	t_R, MS
1700	Limonen-4-ol	-	1.7	MS
1706	α-Terpineol	-	0.2	t_R, MS
1726	Germacrene D	0.3	1.3	t_R, MS
1751	Carvone	0.2	-	t_R, MS
1773	δ-Cadinene	0.2	-	MS
1755	Bicyclogermacrene	-	0.8	MS

DOI: 10.37929/nveo.871951

1797	*p*-Menthylacetophenone	0.3	0.9	MS
1827	Cuminaldehyde	tr	tr	t_R, MS
1802	(*E*, *E*)-2,4-decadienal	0.2	-	MS
1845	*trans*-Carveol	0.4	-	t_R, MS
1864	*p*-Cymen-8-ol	2.2	11.9	MS
1868	(*E*)-Geranyl acetone	tr	-	MS
2008	Caryophyllene oxide	1.6	2.7	t_R, MS
2037	Salvial-4(14)-en-1-one	-	tr	MS
2050	(*E*)-Nerolidol	-	tr	t_R, MS
2071	Humulene epoxide 2	0.3	-	MS
2131	Hexahydrofarnesyl acetone	0.1	-	MS
2144	Spathulenol	0.7	4.4	MS
2245	Elemicine	-	0.4	MS
2243	Torilenol	-	0.4	MS
2247	*trans*-α-Bergamotol	-	0.3	MS
2255	α-Cadinol	-	0.4	t_R, MS
2296	Myristicin	0.3	18.9	t_R, MS
2298	Decanoic acid	0.2	-	t_R, MS
2369	Eudesma-4(15)-7-dien-1-betaol	0.2	0.6	MS
2392	Caryophyllenol-II	0.3	0.4	MS
2404	*trans*-Isoelemicine	-	0.5	MS
2500	Pentacosane	tr	0.3	t_R, MS
2503	Dodecanoic acid	0.2	-	t_R, MS
2655	Benzyl benzoate	0.3	-	t_R, MS
2670	Tetradecanoic acid	1.8	-	t_R, MS
2700	Heptacosane	tr	0.4	t_R, MS
2900	Nonacosane	0.9	0.7	t_R, MS
2931	Hexadecanoic acid	2.0	1.1	t_R, MS
	TOTAL	**95.8**	**94.1**	

RRI, Relative retention indices calculated against *n*-alkanes % calculated from FID data. IM, Identification methods. t_R, identification based on the retention times of genuine compounds on the HP Innowax column. MS, identified on the basis of computer matching of the mass spectra with those of the Wiley and MassFinder libraries and comparison with literature data; tr, trace amount <0.1%.

Essential oil compositions of *Chaerophyllum* species were determined by several studies. Table 2 shows previous reports on different *Chaerophyllum* species. According to Table 2, *C. aksekiense* (Turkey), *C. aromaticum* (Turkey), *C. aureum* (Serbia), *C. azoricum* (Portugal), *C. bulbosum* (Iran), *C. bulbosum* ssp. *Bulbosum* (Greece), *C. byzantinum* (Turkey), *C. coloratum* (Montenegro), *C. crinitum* (Iran, Turkey), *C. hirsutum* (Serbia), *C. libanoticum* (Turkey), *C. macropodum* (Iran, Turkey), *C. macrospermum* (Iran, Nakhichevan), *C. prescotti* (Siberia), *C. temulum (Serbia), C. villosum* (India) were analyzed for their essential oil compositions by different researchers. Different techniques such as water distillation, steam distillation, microwave assisted hydrodistillation, microdistillation, simultaneous distillation-extraction were used to isolate volatile compounds. Different parts of the plants (fruits, seeds, umbels, inflorescences, roots, shoots, stems, and aerial parts) were subjected to distillation procedures. In general, monoterpenes and diterpenes were the main groups in essential oils. Recent data have shown that factors such as seasonal variation, edaphic and genetic factors, harvest time, vegetative stage affected the composition of essential oil (Evergetis and Haroutounian, 2020).

When our findings compared with previous data, *C. crinitum* essential oils were characterized with (*E*)-β-ocimene (50.5%) in Iran (Nematollahi et al., 2005), and characterized with α-terpinolene (20.3%), β-cubebene (9.3%), α-terpineol (7.2%) and limonene (5.8%) in Turkey (Hayta and Celikezen, 2016). Our results indicated (*E*)-β-ocimene (38.1%), terpinolene (12.7%), α-pinene (5.5%), *p*-cymene (5.3%), limonene (5.3%) and *p*-cymen-8-ol (2.2%) as main constituents. *C. macrospermum* essential oils were obtained from seeds (Iran), aerial parts (Iran) and inflorescences (Nakhichevan). The seed essential oil consisted of (*E*)-β-farnesene (27.1 %), (*Z*)-β-ocimene (18.8%), *p*-cymene (14.3%), α-fenchyl acetate (12.7 %) and spathulenol (8.8%) (Razavi and Nejad-Ebrahimi, 2010), the essential oil from aerial parts contained as main constituents (*E*)-β-ocimene (55.9%), terpinolene (9.8%), α-pinene (7.5%), β-phellandrene (4.3%) and β-pinene (4.2%) (Sefidkon and Abdoli, 2005), while the inflorescence essential oil was characterized with 1,8-cineole (7.2%), linalool (6.7%), δ-3-carene (4.4%), α-terpineol (4.7%), farnesol (4.0%) (Mamedova, 1995). Our study reported that terpinolene (21.4%), myristicin (18.9%), *p*-cymen-8-ol (11.9%), limonene (6.1%), α-pinene (4.6%), spathulenol (4.4%), and α-*p*-dimethyl-styrene (3.6%) were major compounds in the essential oil.

Table 2. Previous studies on essential oils of *Chaerophyllum species*

Plant / Origin	**Method/ Results**	**Reference**
C. aksekiense A. Duran et Duman Crushed fruits (Turkey)	Water distillation, GC-MS analysis 77 components, representing 82.0% of the oil; Heptacosane (10.1%), humulene epoxide II (7.8%), (*E*)-β-farnesene (6.2%), caryophyllene oxide (6.0%), α-humulene (5.5%), terpinolene (5.5%), nonacosane (5.3%) and terpinen-4-ol (4.6%)	Başer et al., 2000
C. aromaticum L. Aerial parts (Turkey)	Water distillation, GC and GC-MS analysis 18 compounds, 99.2% of the oil; Sabinene (28.1%), terpinolene (16.7%) and γ-terpinene (16.1%)	Kurkcuoglu et al., 2018
C. aureum L. Aerial parts and fruits (Serbia)	Water distillation, GC and GC-MS analysis 50 compounds, representing 97.7% of aerial parts (Suva mountains), 97.0% of aerial parts (Kopaonik mountains) and 98.5% of fruits (Kopaonik mountains); Sabinene (18.5-31.6%), *p*-cymene (7.9-25.4%) and limonene (1.9-10.9%)	Lakusic et al., 2009
C. azoricum Trel. Leaves+stems (Portugal)	Simultaneous Distillation-Extraction, GC and GC-MS analysis 34-37 compounds, 98-99% of the oil; Terpinolene (44-62%) and γ-terpinene (9-31%) of the oils	Pedro et al., 1999
C. bulbosum L. Aerial parts (Iran)	Water distillation, GC and GC-MS analysis 29 compounds, 92.2% of the oil; (*E*)-β-Farnesene (22.3%), (*Z*)-β-ocimene (18.8%), and myristicin (17.1%), caryophyllene oxide (6.6%), allo-ocimene (5.1%), and (*E*)-β-ocimene (4.0%)	Masoudi et al., 2011
C. bulbosum L. ssp. *bulbosum* (Greece)	GC and GC-MS analysis 29 compounds, 95% of the oil; Apiol (37%), trimethyl-3,7,11-dodecatrien-1,6,10-ol-3 (8.5%), linalool (7.7%), myristicin (6.9%) and eugenol (5.8%)	Kokkalou and Stefanou, 1989
C. byzantinum Boiss. Aerial parts (Turkey)	Water distillation, GC and GC-MS analysis 65 compounds, 94.6% of the oil; Sabinene (30.0%), *p*-cymen-8-ol (16.0%)	Kürkçüoğlu et al., 2006
C. coloratum L. Root and stem (Montenegro)	Water distillation, GC and GC-MS analysis Root: 56 components, 99.2% of the oil; Myrcene (72.2%), β-phellandrene (5.5%) Stem: 18 compounds, 94.4% of the oil; (*E*)-β-ocimene (33.6%), (*Z*)-β-ocimene (20.4%) and terpinolene (10.8%)	Stesevic et al., 2016
C. coloratum	Water distillation, GC and GC-MS analysis	Vajs et al., 1995

Ripe fruits, umbels (Montenegro)	Ripe fruit: 8 compounds, 95.1% of the oil; (*E*)-β-Farnesene (79.2%), β-pinene (7.0%), (*Z*)-β-ocimene (4.3%) Umbel: 8 compounds, 80.2% of the oil; (*E*)-β-Farnesene (68.5%), (*Z*)-β-ocimene (5.5%)	
C. crinitum Boiss. Aerial parts (Iran)	Water distillation, GC and GC-MS analysis 11 components, 84.3% of the oil; (*E*)-β-ocimene (50.5%)	Nematollahi et al., 2005
C. crinitum Aerial parts (Turkey)	Water distillation, GC and GC-MS analysis 64 compounds, 85.5% of the oil; α-Terpinolene (20.3%), β-cubebene (9.3%), α-terpineol (7.2%) and limonene (5.8%)	Hayta and Celikezen, 2016
C. hirsutum L. Roots, shoots, inflorescences (Serbia)	Water distillation, GC and GC-MS analysis Root: 70 compounds, 97.3% of the oil; γ-Terpinene (15.8%), acorenone B (14.2%), (*Z*)-Falcarinol (7.9%), isothymol methyl ether (6.5%) Shoot: 55 compounds, 95.2% of the oil; Acorenone B (57.0%), *endo*-Fenchyl acetate (9.8%), 5-neo-cedranol (6.0%) Inflorescence: 69 compounds, 98.8% of the oil; Acorenone B (44.6%), *endo*-Fenchyl acetate (19.1%), γ-terpinene (6.4%)	Petrovic et al., 2017
C. libanoticum Boiss. et Kotschy Crushed fruits (Turkey)	Water distillation, GC and GC-MS analysis 73 components, 98.3% of the total oil; β-Phellandrene (17.6%), limonene (15.9%), β-pinene (8.8%), and sabinene (8.5%)	Demirci et al., 2007
C. macropodum Boiss. Aerial parts (Iran)	Water distillation (WD) and Microwave assisted hydrodistillation (MAHD), GC-MS analysis (*E,Z*)-β-Ocimene (29.3 and 29.3%), myrcene (14.5 and 14.6%), and terpinolene (14.4 and 14.3%) in WD and MAHD, resp.	Khajehie et al., 2017
C. macropodum Leaves and flowers (Iran)	Water distillation, GC and GC-MS analysis Leaf: 37 compounds, 98.3% of the oil; Spathulenol (10.4%), myrcene (10.0%), germacrene D (8.7%), limonene (6.7%), β-caryophyllene (6.2%) Flower: 31 compounds, 99.4% of the oil; *trans*-β-Farnesene (27.5%), *trans*-β-ocimene (20.9%), limonene (12.0%), *cis*-β-ocimene (4.3%)	Ebrahimabadi et al., 2010
C. macropodum Flowers, leaf, stem (Iran)	Water distillation, GC and GC-MS analysis Flower: 10 compounds, 98.5% of the oil; Myristicin (42.5%) and *trans*-β-ocimene (41.0%) Leaf: 18 compounds, 99.3% of the oil; *trans*-β-Ocimene (24.9%), myristicin (15.7%), terpinolene (14.5%), fenchyl acetate (13.9%), *cis*-β-ocimene (6.3%) and sabinene (6.1%) Stem: 11 compounds, 97.1% of the oil; *trans*-β-Ocimene (54.2%), myristicin (22.4%) and sabinene (8.9%)	Shafaghat, 2009
C. macropodum Fruits (Turkey)	Microdistillation, GC and GC-MS analysis 41 compounds, 92% of the sample; *p*-Cymene (39.3%), spathulenol (7.3%), *p*-cymen-8-ol (5.9%), octanal (5.2%), (*E*)-β-ocimene (4.5%)	Başer et al., 2006
C. macropodum Aerial parts (Iran)	Water distillation, GC and GC-MS analysis 28 compounds, 98.5% of the oil; α-Pinene (23.0%), β-pinene (17.3%) and fenchyl acetate (13.8%)	Nematollahi et al., 2005
C. macrospermum (Spreng.) Fisch et C.A. Mey. Seeds (Iran)	Water distillation, GC-MS analysis 15 compounds, 94.6% of the oil; (*E*)-β-Farnesene (27.1%), (*Z*)-β-ocimene (18.8%), *p*-cymene (14.3 %), α-fenchyl acetate (12.7%) and spathulenol (8.8%)	Razavi and Nejad-Ebrahimi, 2010
C. macrospermum Aerial parts (Iran)	Water and Steam distillation, GC and GC-MS analysis 27 compounds; (*E*)-β-Ocimene (55.9%), terpinolene (9.8%), α-pinene (7.5%), β-phellandrene (4.3%) and β-pinene (4.2%)	Sefidkon and Abdoli, 2005
C. macrospermum	Water distillation, GC and GC-MS analysis	Rustaiyan et al., 2002

Aerial parts (Iran)	16 compounds, 99.6% of the oil; (*E*)-β-Ocimene (40.0%), tricyclene (19.4%), δ-3-carene (18.3%) and mycrene (10.1%)	
C. macrospermum Inflorescences (Nakhichevan)	Steam distillation, GC analysis 33 compounds; Cineole (7.2%), linalool (6.7%), δ-3-carene (4.4%), a-terpineol (4.7%), farnesol (4.0%)	Mamedova, 1994
C. prescotti DC Flowering tops (Siberia)	Steam distillation, GC and GC-MS analysis 18 compounds, 99% of the oil; (*E*)-β-Ocimene (35.6%), (*Z*)-β-ocimene (19.4%), γ-terpinene (18.8%), myrcene (10.6%) and terpinolene (4.6%)	Letchamo et al., 2005
C. temulum L. Root, stem, inflorescence, fruit (Serbia)	Water distillation, GC and GC-MS analysis Root at the flowering stage: 35 compounds, 90.4% of the oil; (*Z*)-Falcarinol (61.7%) Root at the fruiting stage: 31 compounds, 91.2% of the oil; (*Z*)-Falcarinol 62.3% Stem at the flowering stage: 43 compounds, 94.8% of the oil; Germacrene D (38.4), phytol (12.4%), α-humulene (10.1%) Stem at the fruiting stage: 45 compounds, 92.3% of the oil; Germacrene D (32.5%), phytol (11.6%), α-humulene (8.1%) Inflorescence: 74 compounds, 96.8% of the oil; (*Z, E*)-α-Farnesene (23.4%), germacren-D-4-ol (9.0%), (*E*)-β-farnesene (9.0%), (*E*)-β-ocimene (7.4%), germacrene D (6.6%), β-phellandrene (6.1%), Fruit: 41 compounds, 95.6% of the oil; Germacren-D-4-ol (27.6%), (*Z, E*)-α-farnesene (13.4%), α-zingiberene (8.6%), (*E*)-β-farnesene (7.8%)	Stamenkovic et al., 2015
C. villosum Wall. ex DC. Root, leaf (India)	Water distillation (for root), Steam distillation (for leaf), GC and GC-MS analysis Root: 31 compounds, 91.5% of the oil; Carvacrol methyl ether (31.1%), myristicin (19.1%), thymol methyl ether (18.6 %), γ-terpinene (11.7%) Leaf: γ-Terpinene (74.9%), *p*-cymene (10.0%), terpinolene (2.9%) and β-pinene (2.5%)	Joshi, 2013a; Joshi, 2013b

As conclusion, significant differences between the essential oils obtained by our study and the previous studies which analyzed *Chaerophyllum* species gathered from different countries (geographic regions), originated from different parts of the plants (fruit, stem, flower, seed, *etc*) and plant parts at different phases of growth were observed. It may be concluded that large differences in the environmental conditions have contributed to the large variability in the composition and major compounds of *Chaerophyllum* essential oils. Due to the rather heterogeneous data on the chemistry of the different *Chaerophyllum* species and the insufficient number of taxa studies, it may not be sufficient to make detailed comparison within the genus and obtain possible chemical diagnostic properties. Hence, detailed further studies on evaluation of essential oils of these species are needed.

ACKNOWLEDGEMENT

This study presented as a poster presentation at International Symposium of Essential Oils held on September 11-14, 2011 in Turkey was published as an abstract in JEOR's 23th volume (sup 1).

CONFLICTS OF INTEREST

The authors have no conflicts of interest to declare.

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