

RESEARCH ARTICLE

Chemical Composition of *Heracleum platytaenium* Boiss. (Apiaceae) essential oil from Turkey

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Abstract

Heracleum platytaenium Boiss. is a monocarpic and strongly aromatic plant that is endemic for Turkey. Many studies on activities, constituents and composition of the essential oil of different parts of this species and other *Heracleum* species have been conducted. However, as far as we are concerned, the essential oil composition of the roots has not been investigated. In this study, we investigated the essential oil composition of the roots of *H. platytaenium* and identified *p*-cymene (33.9%), terpinolene (14.3%), γ -terpinene (7.1%), elemicine (3.1%) and myristicine (2.9%) as the major constituents.

Key words: *Heracleum platytaenium*, Apiaceae, essential oil, GC, GC-MS

Introduction

Heracleum L. belongs to the family Apiaceae and is represented by 23 taxa in Turkey and 7 of these species are endemics (Guner, 2012). The genus has a widespread distribution throughout Europe, therefore some of the species are considered to be invasive (Bahadori et al., 2016). Some species of the genus are being used in traditional medicine in our country and also throughout the world. *Heracleum* species are reported to be used as antipyretic, analgesic, diaphoretic, antiseptic, carminative and digestive agents, for the treatment of rheumatic disease, hypertension, epilepsy and diarrhea, lumbago, gastralgia and injuries from falls, contusions and strains (Akcin et al., 2013) and as wound healing (Tetik et al., 2013). One *Heracleum* species (*H. candicans*) is used as a source for xanthotoxin – a substance used in the composition of suntan lotions for its antileucodermal properties (Guleri et al., 2011) furanocoumarins and essential oil obtained from *H. crenatifolium* is reported to have anticonvulsant activity (Tosun et al., 2008). Some *Heracleum* species are reported to be used as flavoring, spices, carminative and antiseptic, digestive and analgesic in Iranian folk medicine (Sefidkon et al., 2002; Hajhashemi et al., 2009; Torbati et al., 2014; Amanpour et al., 2016); as a carminative herbal drug in Thai folk medicine (Kuljanabhagavad et al., 2010) and for inflammatory condition, as antiarthritic and nerve tonic in Indian folk medicine (Purushothaman and Ravi, 2013). *Heracleum platytaenium* is known with the names havlan, havlanotu, hometi, kekrer, romati, yabanlahanası (Kızılarşlan and Özhatay, 2012) and yavşan otu (Bayrak Özbucak et al., 2007) in Turkish. In addition to its traditional usage as medicine, the species is also used for culinary purposes; e.g. the stems and stalks of the plant are consumed as pickles around İzmit (Kızılarşlan and Özhatay, 2012); the stems of the plant are first pickled and then eaten as raw or as cooked with olive oil in the Black Sea region of Turkey (Bayrak Ozbucak et al., 2006) and leaves of the plant are coked as meal or eaten as fresh in salads around Nigde (Ozdemir and Alpınar, 2010-2011). Since this species (Koçak et Buruk et al., 2006) and different *Heracleum* species are also reported to have antiviral (Tkachenko, 2006), antibacterial and antifungal activities (Benli, et al., 2007; Ergene et al.,

2009; Kuljanabhadgavad et al., 2010; Özçakmak, 2012; Jagannath et al., 2012; Miladinovic, et al., 2013), this species is tested against different strains of bacteria and fungi and the essential oil of the plant was determined to have inhibitory and toxic effect against a fungus called *Penicillium verrucosum* which was isolated from kashar cheese (Özçakmak, 2012), and against *Paenibacillus larvae* (Özkırım et al., 2012).

Though essential oils of different parts of various *Heracleum* species have been studied previously, studies of the composition of the roots are scarce. In this study, we isolated the essential oil of the roots and analyzed it by means of GC and GC-MS and reported the results.

Materials and Methods

Plant material

The plant was collected from the below mentioned locality and identified by Prof. Dr. Hayri Duman (Gazi University, Faculty of Science, Department of Biology) and the voucher specimen is kept in AEF (Herbarium of Ankara University Faculty of Pharmacy).

Collection locality: *Heracleum platytaenium*: A7: Trabzon: Maçka-Torul arası, Maçka çıkışında yol kenarları, 3/7/2006 (AEF: 23756)

Isolation of the essential oils

Air dried roots (50g) were subjected hydrodistillation for 3 h using a Clevenger apparatus. Essential oil, obtained with a yield of 4% was dried over anhydrous sodium sulfate and stored in a sealed vial at +4°C in the dark until analyzed and tested.

GC and GC/MS analyses

GC-MS analysis

The GC-MS analysis was carried out with an Agilent 5975 GC-MSD system. Innowax FSC column (60 m x 0.25 mm, 0.25 µm film thickness) was used with helium as carrier gas (0.8 ml/min). GC oven temperature was kept at 60°C for 10 min and programmed to 220°C at a rate of 4°C/min, and kept constant at 220°C for 10 min and then programmed to 240°C at a rate of 1°C/min. Split ratio was adjusted at 40:1. The injector temperature was set at 250°C. Mass spectra were recorded at 70 eV. Mass range was from m/z 35 to 450.

GC analysis

The GC analysis was carried out using an Agilent 6890N GC system. FID detector temperature was 300°C. To obtain the same elution order with GC-MS, simultaneous auto-injection was done on a duplicate of the same column applying the same operational conditions. Relative percentage amounts of the separated compounds were calculated from FID chromatograms.

Identification of the volatile compounds

Identification of the essential oil components were carried out by comparison of their relative retention times with those of authentic samples or by comparison of their relative retention index (RRI) to series of *n*-alkanes. Computer matching against commercial (Wiley GC/MS Library, Adams Library, MassFinder 3 Library), and in-house "Başer Library of Essential Oil Constituents" built up by genuine compounds and components of known oils, as well as MS literature data (Joulain and Koenig, 1998; ESO 2000, 1999) was used for the identification.

Results and Discussion

Analysis of the essential oil resulted in the characterization of 70 components, corresponding to the 89.4% of the total oil. The composition of the essential oil is given in Table 1 with major components in written in bold.

Table1. Composition of the Essential Oil of *Heracleum platytaenium* roots

RRI	Compounds	%
1032	α -Pinene	2.5
1035	α -Thujene	2.6
1076	Camphene	0.5
1093	Hexanal	0.2
1118	β -Pinene	0.3
1132	Sabinene	1.2
1159	δ -3-Carene	0.1
1174	Myrcene	1.2
1176	α -Phellandrene	tr
1188	α -Terpinene	0.2
1194	Heptanal	0.4
1203	Limonene	2.6
1218	β -Phellandrene	tr
1244	2-Pentyl furan	0.3
1246	(Z)- β -Ocimene	0.2
1255	γ-Terpinene	7.1
1266	(E)- β -Ocimene	0.3
1280	<i>p</i>-Cymene	33.9
1290	Terpinolene	14.3
1296	Octanal	1.3
1483	Octyl acetate	0.1
1492	Cyclosativene	0.1
1497	α -Copaene	0.3
1548	(E)-2-Nonenal	0.2
1562	Octanol	0.3
1591	Bornyl acetate	0.2
1595	Isothymol methyl ether	tr
1596	α -Guaiene	0.2
1597	β -Copaene	0.3
1604	Thymol methyl ether (=methyl thymol)	0.3
1611	Terpinen-4-ol	0.2
1614	Carvacrol methyl ether (=methyl carvacrol)	1.1
1655	(E)-2-Decenal	0.7
1683	<i>trans</i> -Verbenol	tr
1704	γ -Muurolene	tr
1709	α -Terpinyl acetate	tr
1715	(E,E)-2,4-Nonadienal	tr

1726	Germacrene D	tr
1730	δ -Guaiene	0.4
1740	α -Muurolene	0.4
1758	(<i>E,E</i>)- α -Farnesene	0.1
1763	Naphthalene	0.2
1771	γ -Bisabolene	0.3
1773	δ -Cadinene	0.8
1779	(<i>E,Z</i>)-2,4-Decadienal	0.5
1827	(<i>E,E</i>)-2,4-Decadienal	0.8
1864	<i>p</i> -Cymen-8-ol	0.7
1868	(<i>E</i>)-Geranyl acetone	tr
1878	2,5-Dimethoxy- <i>p</i> -cymene	0.4
1900	<i>epi</i> -Cubebol	tr
1941	α -Calacorene	0.3
1957	Cubebol	0.4
2008	Caryophyllene oxide	0.3
2050	(<i>E</i>)-Nerolidol	0.4
2071	Humulene epoxide-II	0.5
2080	Cubenol	tr
2088	1- <i>epi</i> -Cubenol	tr
2144	Spathulenol	1.0
2187	T-Cadinol	0.6
2209	T-Muurolol	tr
2219	δ -Cadinol (= α -muurolol)	0.5
2239	Carvacrol	0.3
2245	Elemicine	3.1
2247	<i>trans</i> - α -Bergamotol	tr
2255	α -Cadinol	tr
2257	β -Eudesmol	tr
2296	Myristicine	2.9
2384	Hexadecanol	0.5
2822	Pentadecanoic acid	tr
2931	Hexadecanoic acid	0.8
Total		89.4

RRI Relative retention indices calculated against *n*-alkanes; % calculated from FID data; tr Trace (< 0.1 %)

As it can be seen from the Table 1.; *p*-cymene (33.9%), terpinolene (14.3%), γ -terpinene (7.1%), elemicine (3.1%) and myristicine (2.9%) were identified as the major constituents of the essential oil of the roots of *H. platytaenium*. When we compare these results with the literature, we can see that myristicin was found to be present in two other species as the major component, however with a percentage much higher than the one we have in our study [(2.9%) vs. 95.15% for *H. anisactis*, and 96.87% for *H. transcaucasicum*,].

Table 2. Composition of the Essential Oil of roots of other *Heracleum* spp. found in the literature

Species	Used part	Compound	%	Ref.
<i>H. anisactis</i>	Roots	Myristicin	95.15	Torbati et.al. (2014)
<i>H. candolleianum</i>	Rhizomes	α -pinene	18.9	George et al. (2001)
		bornylene	18.6	
		octyl acetate	11.9	
<i>H. hemsleyanum</i>	Radix	(1S)-6,6-dimethyl-2-methylenebicyclo[3,1,1]heptane	12.60	Zhang et al. (2005)
		8-isopropenyl-1,5-dimethyl-cyclodeca-15-diene	10.42	
<i>H. persicum</i>	Roots	Viridiflorol	23.05	Mojab and Nickavar (2003).
<i>H. sprengelianum</i>	Rhizomes	1,8-cineole	23.10	Karuppusamy and Muthuraja (2011)
		β -pinene	21.84	
		β -phellandrene	15.19	
<i>H. transcaucasicum</i>	Roots	Myristicin	96.87	Torbati et.al., (2014)

When we searched the literature for the components of essential oils obtained from other parts of *Heracleum* spp., we have seen that myristicin was present in the aerial parts of *H. anisactis* and *H. transcaucasicum* species (2.9% vs. 93.54% and 70.12%, respectively) (Torbati et.al., 2013). Our other major compounds were found to be present in different *Heracleum* species are as follows:

***p*-cymene (33.9%)** was present in the essential oil of the fruits of *H. persicum* (37.3%), (Scheffer et al., 1984) and the stem of *H. rechingeri* (4.5%) (Habibi et al., 2010);

terpinolene (14.3%) was present in the fruits of *H. antasiaticum* (10.75%) (Ibadullaeva, 2000), in the leaves of *H. persicum* (9.86%) (Mojab et al., 2002), in the stems of *H. persicum* (11.3%) (Sefidkon et al., 2004), in the aerial parts of *H. thomsonii* (22.24%) (Guleria et al., 2011);

γ -Terpinene (7.1%) was present in the fruits of *H. persicum* (27.8%) (Scheffer et al., 1984), in the flowers of *H. persicum* (17.8%), (Sefidkon et al., 2002); in the stems of *H. persicum* (7.1%) (Sefidkon et al., 2004), in the stems of *H. rechingeri* (8.8%) (Habibi et al., 2010);

elemicine (23.1%) was not found to be present in the *Heracleum* species according to our literature search.

Though the major compounds that we have found in the essential oil of *H. platytaenium* roots may be present in different parts of some other *Heracleum* species (major components of the volatile oil isolated from different parts of various *Heracleum* species is given in Table. 3 for further information) , we can not conclude that these can be considered as chemotaxonomic markers for the genus. In order to make this assumption, essential oils of all *Heracleum* species should be analyzed and compared since the studies on the composition of the roots of *Heracleum* species are scarce in the literature. However, we can conclude that elemicine has been found to be present for the first time in a *Heracleum* species, though with no chemotaxonomic value.

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Table 3. Composition of volatile oils obtained from different parts of different *Heracleum* species

Species name	Organ	Major compounds (%)	Reference
<i>H. afghanicum</i>	Fruits	Hexyl butyrate	34.3
		Octyl acetate	21.1
<i>H. anisactis</i>	Aerial	Myristicin	93.5

<i>H. anisactis</i>	Fruits	Octyl acetate	48.1	Radjabian et al., 2014
<i>H. anisactis</i>	Roots	Myristicin	95.2	Torbati et al., 2014
<i>H. antiasiaticum</i>	Flower	Octyl-4-methylvalerate Octyl acetate	86.0 5.9	Tkachenko and Zenkevich 1993
<i>H. antiasiaticum</i>	Fruits	Ethylcaprylate Octyl acetate terpinolene	29.8 17.4 10.8	Ibadullaeva, 2000
<i>H. argaeum</i>	Fruits	Hexyl butyrate	39.1	Başer et al., 1998
<i>H. candicans</i>	Fruits	Bornyl acetate Amyl acetate Octyl acetate	21.5 19.6 6.0	Ashraf and Bhattay, 1978
<i>H. candolleanum</i>	Rhizomes	α -pinene bornylene octyl acetate	18.9 18.6 11.9	George et al., 2001
<i>H. candolleanum</i>	Fruits	Methyl cinnamate n-hexyl hexanoate octyl alcohol	22.4 21.7 11.8	Prushothaman and Ravi, 2013
<i>H. crenatifolium</i>	Fruits	Octyl acetate	93.7	İscan et al., 2004
<i>H. crenatifolium</i>	Fruits	Octyl acetate	93.7	Özek et al., 2005
<i>H. crenatifolium</i>	Fruits	Octyl acetate	88.4	Tosun et al., 2008
<i>H. dissectum</i>	Aerial	α -pinene myrcene kessan humulene	22.2 10.9 8.8 8.3	Papageorgiou et al., 1985
<i>H. dulce</i>	Fruits	Octyl acetate Octyl butyrate	40.0 26.0 20.0	Tkachenko, 1993
<i>H. gorganicum</i>	Fruits	Hexyl butanoate Octyl acetate	33.3 18.4	Radjabian et al., 2014
<i>H. grandiflorum</i>	Fruits	Octyl acetate Octyl hexanoate Hexyl butyrate	39.0 13.0 5.7	Tkachenko, 1993
<i>H. hemsleyanum</i>	Radix	(1S)-6-6,dimethyl-2- methylenebicyclo[3,1,1]heptane 8-isopropenyl-1,5-dimethyl-cyclodeca- 15-diene	12.6 10.4	Zhang et al., 2005
<i>H. leskovii</i>	Fruits	Octyl acetate Octyl butyrate Octyl octanoate	65.0 9.3 6.1	Tkachenko, 1993
<i>H. nanum</i>	Fruits	Octyl acetate Octyl butyrate Hexyl butyrate	41.0 20.0 7.4	Tkachenko, 1993
<i>H. mantegazzianum</i>	Fruits	Octyl butyrate Octyl acetate	32.0 18.0	Tkachenko, 1993
<i>H. maximum</i>	Fruit	Octyl acetate Octyl butyrate	65.6 7.9	St-Gelais et al., 2016
	Stem	Limonene sabinene	45.2 9.5	
<i>H. moellendorffii</i>	Fruits	Octyl acetate Octyl butyrate Octyl octanoate octanal	51.0 13.4 10.2 6.0	Tkachenko, 1993
<i>H. paphlagonicum</i>	Fruits	Octyl acetate Hexyl butyrate Octyl hexanoate	31.5 17.0 10.2	Başer et al., 2000
<i>H. pastinacifolium</i>	Aerial	Myristicin (Z)-trans- α -bergamotene	53.6 10.6	Firuzi et al., 2010
<i>H. pastinacifolium</i>	Fruits	Octyl acetate	59.5	Radjabian et al., 2014
<i>H. persicum</i>	Fruit	p-cymene γ -terpinene	37.3 27.8	Scheffer et al., 1984

		α -pinene	13.8	
		β -pinene	5.8	
<i>H. persicum</i>	Leaves	B-springene	37.7	Mojab et al., 2002
		Spathulenol	23.8	
		α -farnesene	22.0	
		zingiberene	21.5	
		α -bergamotene	20.3	
		α -caryophyllene	19.9	
		trans-anethole	15.6	
		cis-anethole	14.4	
		stragole	12.7	
		terpinolene	9.9	
<i>H. persicum</i>	Leaves (at flowering)	(<i>E</i>)-anethole	47.5	Sefidkon et al., 2002
		1-(4-methoxy phenyl)-2-propanone	18.1	
		anisaldehyde	8.9	
	Flowers	(<i>E</i>)-anethole	38.6	
		γ -terpinene	17.8	
		myrcene	13.5	
<i>H. persicum</i>	Roots	Viridiflorol	23.1	Mojab and Nickavar, 2003.
<i>H. persicum</i>	Stem	(<i>E</i>)-anethole	60.2	Sefidkon et al., 2004
		Terpinolene	11.3	
		γ -terpinene	7.1	
	Seed	Hexyl butyrate	35.5	Sefidkon et al., 2004
		Octyl acetate	27	
		Hexyl isobutyrate	3.2	
<i>H. persicum</i>	Fruits	Hexyl butyrate	56.5	Hajhashemi et al., 2009
		Octyl acetate	16.5	
<i>H. persicum</i>	Aerial	(<i>E</i>)-anethole	25.0	Firuzi et al., 2010
		Octyl-2-methyl butanoate	14.2	
		Hexyl butanoate	10.0	
<i>H. persicum</i>	Fruits	Hexyl butanoate	17.7	Radjabian et al., 2014
		Octyl acetate	20.5	
<i>H. persicum</i>	Fruits	Hexyl butyrate	65.6	Amanpour et al., 2016
		Octyl acetate	18.2	
<i>H. platytaenium</i>	Fruits	Octyl acetate	72.3-76.7	Kürkçüoğlu et al., 1995
		Octyl butyrate	11.3-16.7	
<i>H. platytaenium</i>	Fruits	Carvacrol	29.9	Akcin et al., 2013
		Thymol	29.4	
		Octyl octanoate	26.9	
		Octyl hexanoate	24.4	
		Decanol	23.6	
<i>H. platytaenium</i>	Fruits	Octyl acetate	87.6	İscan et al., 2004
<i>H. platytaenium</i>	Fruits	Octyl acetate	87.6	Özek et al., 2005
<i>H. rawianum</i>	Fruits	Octyl acetate	75.4	Radjabian et al., 2014
<i>H. rechingeri</i>	Aerial	Hexyl butanoate	29.7	Firuzi et al., 2010
		Octyl butanoate	10.1	
<i>H. rechingeri</i>	Flower	Elemicin	39.5	Habibi et al., 2010
		Octyl acetate	25.1	
		(<i>E</i>)-caryophyllene	10.0	
		(<i>E</i>)- β -ocimene	5.2	
	Fruit	Octyl acetate	95.3	
Stem	Elemicin	37.7		
	Octyl acetate	26.5		
	γ -terpinene	8.8		
	(<i>E</i>)-caryophyllene	6.6		
	<i>p</i> -cymene	4.5		
<i>H. rechingeri</i>	Fruits	Hexyl butanoate	38.4	Radjabian et al., 2014
		Octyl acetate	13.9	
<i>H. rigens</i>	Fruits	Bornyl acetate	51.2	Jagannath et al., 2012
		α -pinene	22.6	

		limonene	9.6	
<i>H. siamicum</i>	Fruits	n-octyl acetate o-cymene limonene δ -2-carene	65.3 10.4 7.5 6.9	Kuljanabagavad et al., 2010
<i>H. sibiricum</i>	Aerial	Octyl butanoate Hexyl butanoate 1-octanol Octyl hexanoate	36.8 16.1 13.6 8.1	Miladinovic et al., 2013
<i>H. sphondylium</i>	Fruits	Octyl butyrate Octyl acetate	37.7 31.6	İscan et al., 2004
<i>H. sphondylium</i> ssp. <i>ternatum</i>	Fruits	Octanol Octyl butyrate Octyl acetate	39.2 27.4 10.6	Ozek et al., 2002
<i>H. sphondylium</i> ssp. <i>ternatum</i>	Seeds	1-octanol Octyl butyrate	50.3 24.6	İşcan et al., 2003
<i>H. sphondylium</i> ssp. <i>ternatum</i>	Fruits	Octyl butyrate Octyl acetate	37.7 31.6	Özek et al., 2005
<i>H. sphondylium</i> ssp. <i>ternatum</i>	Fruits	Octyl acetate Octyl butyrate	54.9-60.2 10.1.-13.4	Maggi et al., 2014
<i>H. sprengelianum</i>	Leaves	1,8-cineole β -pinene β -phellandrene	21.2 16.2 11.4	Karuppusamy and Muthuraja, 2011
	Seeds	β -pinene 1,8-cineole β -phellandrene	22.3 20.3 12.4	
	Rhizomes	1,8-cineole β -pinene β -phellandrene	23.1 21.9 15.2	
<i>H. stevenii</i>	Buds	Octyl acetate Octyl hexanoate	49.0 8.2	Tkachenko, 1994
	Flowers	Octyl acetate Myrcene	73.0 5.1	
	Stems	Octyl acetate Octyl hexanoate Octyl octanoate	28.0 22.0 10.4	
	Leaves	Octyl acetate Octyl octanoate Octyl 4-methylvalerate	35.0 15.0 12.0	
	Roots	Octyl acetate limonene Octyl octanoate Octyl hexanoate	35.0 20.0 18.0 12.0	
<i>H. transcaucasicum</i>	Aerial	Elemicin	41.1	Firuzi et al., 2010
<i>H. transcaucasicum</i>	Aerial	Myristicin <i>n</i> -octanol	70.1 14.3	Torbati et al., 2013
<i>H. transcaucasicum</i>	Roots	Myristicin	96.9	Torbati et al., 2014
<i>H. thomsonii</i>	Aerial	Neryl acetate Terpinolene limonene	36.2 22.2 4.3	Guleria et al., 2011