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ESSENTIAL OIL COMPOSITION OF *Origanum majorana* AND *Origanum vulgare* ssp. *hirtum* GROWING IN INDIA

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Origanum species (Family Labiatae) have been used in medicine and as spices since antiquity mainly because of their essential oils, which consist of a considerable amount of carvacrol and thymol [1]. Both *O. majorana* L. and *O. vulgare* L. ssp. *hirtum* are perennial and are found in temperate regions of the Himalayas from Kashmir to Sikkim at altitudes from 1500 to 3700. The plant possesses a thyme- like flavor and is used for flavoring foods and beverages [2].

Origanum species contain chiefly carvacrol and/or thymol together with considerable amounts of γ-terpinene and *p*-cymene and have been used for their essential oils in medicine and the food industry [3, 4]. During the past few years there has been a market increase in the interest shown on many herbal spices, and the most popular of them is oregano, which is used to enhance many kinds of foods [2]. In this study, we report on the chemical composition of the essential oil obtained from the air-dried parts of *O. majorana* and *O. vulgare* plants growing in India.

The essential oil content of *O. majorana* and *O. vulgare* ssp. *hirtum* on a dry weight basis constitutes 0.37% and 0.25%, respectively. The chemical compositions of the essential oils obtained from *O. majorana* and *O. vulgare* ssp. *hirtum* are listed in Table 1. In this study, a total of 34 and 28 compounds, which account for about 90.85% and 94.64% of the essential oils of *O. majorana* and *O. vulgare* plants, respectively, was identified. The major components of *O. vulgare* were thymol (33.92%), carvacrol (6.90%), γ-terpinene (17.67%), and *p*-cymene (11.99%). Both thymol and carvacrol constituted 40.82% for the phenolic character in *O. vulgare*, which was completely absent in *O. majorana*. In the case of phenolic compounds, the metabolic pathway is through the autooxidative conversion of γ-terpinene to *p*-cymene followed by hydroxylation of *p*-cymene to thymol, carvacrol, and *p*-cymen-8-ol. The last one was not detected in our oil. A sum of the two phenols and their precursors constituted the bulk of *O. vulgare* oil (70.48%). The observed increase in thymol percentage with decreasing carvacrol content indicates a biosynthetic correlation between the two compounds. Reports suggest that the thymol biosynthetic pathway is favored with altitude and becomes more efficient than the carvacrol biosynthetic pathway [5]. A thymol content of 49.3% was reported in the oil of plants from the Mediterranean region along with 24.6% carvacrol [6]. Several chemotypes of *O. vulgare* were reported from different places [7, 8]. The chemical composition of oregano essential oils is of great importance because of its high biological activity as well as antioxidant activity.

The oil composition of *O. majorana* showed terpinen-4-ol (31.15%), *cis*-sabinene hydrate (15.76%), *p*-cymene (6.83%), sabinene (6.91%), *trans*-sabinene hydrate (3.86%), and α-terpineol (3.71%) as the main constituents. Precursors of phenolic components like *p*-cymene and γ-terpinene were much higher in *O. vulgare* compared to *O. majorana*, whereas sabinene, *cis*- and *trans*-sabinene hydrate, and α-terpineole were much higher in *O. majorana.* The amounts of oxygenated monoterpenes were higher in *O. majorana. cis*-Sabinene hydrate is a compound of intense, spicy "marjoramy" aroma, whereas *trans*-sabinene hydrate has no typical "marjoramy" properties. The reported high content of terpinen-4-ol in the essential oil is due to component rearrangements occurring during the process of distillation. The chemical compositions of wild marjoram and marjoram have been studied by several authors. The most prominent components of *O. majorana* were carvacrol (65%) and thymol (4%) [1]. The main components of the oil from *O. majorana* collected from Greece were terpinen-4-ol (37.1%), *p*-cymene (12.05%), α-terpineol (7.15%), carvacrol (3.60%), *trans*-sabinene hydrate (2.41%), *cis*-sabinene hydrate (1.43%), and thymol (0.7%) [9]. Oil with linalool (32.68%), terpinen-4-ol (32.30%), *p*-cymene (8.0%), sabinene (4.57%), and α-terpeneol (3.26%) as the major constituents were also reported [10]. Our *O. majorana* oil composition was found to be close to that reported by [10] except for some minor variations. The oil quality was close to oil produced in Europe and southern India.

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TABLE 1. Essential Oil Composition of *Origanum majorana* (1) and *Origanum vulgare* (2) ssp. *hirtum*

Compound	RI	Area, %		Compound	RI	Area, %	
		1	2			1	2
α-Thujene	924	0.34	0.11	*trans*-*p*-Menth-2-en-1-ol	1136	1.88	–
α-Pinene	932	0.53	2.00	(*Z*)-β-Terpineol	1159	0.39	–
Camphene	949	0.11	1.55	Borneol	1170	0.63	1.69
Sabinene	969	**6.91**	**0.21**	**Terpinen-4-ol**	1174	**31.15**	**0.37**
β-Pinene	976	0.34	0.20	*p*-Cymen-8-ol	1179	0.04	–
3-Octanol	984	–	0.65	α-Terpineol	1186	3.71	1.10
Myrcene	989	1.63	3.40	*cis*-Sabinene hydrate acetate	1219	0.20	–
δ-3-Carene	1008	–	0.90	Carvone	1239	–	1.25
α-Terpinene	1014	–	0.51	*trans*-Sabinene hydrate acetate	1253	1.18	–
***p*-Cymene**	1020	**6.83**	**11.99**	Linalyl acetate	1254	2.41	1.09
Methyl heptenone	1021	0.16	–	Bornyl acetate	1284	0.32	–
β-Phellandrene	1025	0.34	–	**Thymol**	1300	**0.33**	**33.92**
Limonene	1030	2.41	–	**Carvacrol**	1311	–	**6.90**
(*Z*)-β-Ocimene	1032	0.04	0.17	β-Caryophyllene	1429	2.49	2.06
(*E*)-β-Ocimene	1044	0.05	–	α-Humulene	1452	0.11	0.80
1,8-Cineole	1035	0.20	0.49	Bicyclogermacrene	1500	1.90	–
γ-Terpinene	1054	**5.37**	**17.67**	β-Bisabolene	1505	–	1.36
***cis*-Sabinene hydrate**	1065	**15.76**	**0.54**	Spathulenol	1577	–	0.46
Terpinolene	1086	1.63	–	Caryophyllene oxide	1582	0.88	0.20
trans-Sabinene hydrate	1098	3.86	2.01	(*Z,Z*)-Farnesol	1698	0.05	–
Linalool	1102	0.88	1.04	(*E,E*)-Farnesol	1742	0.02	–
cis-*p*-Menth-2-en-1-ol	1118	1.10	–	Total identified compounds		**95.85**	**94.64**

The present study indicated that the oil of *O. vulgare* from the Uttranchal region of India is a thymol-rich chemotype which has great importance because of its high biological activity as well as antioxidant activity. It also indicated that the thymol biosynthetic pathway is favored with altitude and becomes more efficient than the carvacrol biosynthetic pathway. The oil of *O. majorana* was a mixed chemotype rich in *cis*-sabinene hydrate and terpinen-4-ol.

Plant Material and Isolation Procedure. Samples were collected from plants of *O. majorana* and *O. vulgare* ssp. *hirtum* growing in NBPGR Regional Station, Bhowali (1600 m altitude, 79°30′ N latitude and 29°20′ E longitude) of the rain-fed subtemperate hills of Uttaranchal. Aerial parts of the plant were harvested during the flowering period in April–May 2007 and 2008. The aerial parts were dried in the shade at room temperature.

Dried aerial parts of the plants were ground and subjected to hydrodistillation for 4 h using a Clevenger-type apparatus. The oils extracted were dried over anhydrous sodium sulfate. The oil was stored in a sealed glass vial in the dark at 4°C until further analysis.

The chemical composition of the essential oils isolated by hydrodistillation of the aerial parts of *Origanum majorana* L. and *O. vulgare* L. from India were analyzed by gas chromatography and gas chromatography-mass spectrometry [11, 12].

GC and GC-MS Analysis. Gas chromatographic analyses were carried out using Perkin–Elmer Autosystem 8500 gas chromatograph, fitted with a FID and a Carbowax 20M capillary column (50 m × 0.25 mm × 0.25 μm). Nitrogen was used as carrier gas (1.0 mL/min). The initial temperature of column was 60°C for 10 min; then it was raised to 180°C at 3°C/ min, and held there for 10 min. The injector and detector temperature was maintained at 200°C and 250°C, respectively. The sample (0.1 μL) was injected neat in split ratio (1:40) at 200°C. Gas chromatography/mass spectrometry (GC-MS) analyses were carried out under similar conditions using DB-5 fused silica capillary column (30 m × 0.25 mm × 0.25 μm) and Helium was used as carrier gas (1.0 mL/min). The acquisition mass range used was *m/z* 40 to 450 [11, 12].

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