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Essential oil composition of aerial parts of *Angelica glauca* growing wild in North-West Himalaya (India)

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Abstract

Fresh aerial parts of *Angelica glauca*, growing wild in Kashmir valley in higher Himalaya (Jammu and Kashmir, India), collected at flowering stage from different locations, on hydro-distillation provided a refreshing light pale coloured essential oil with characteristic floral woody flavour. The oil was found to be a complex mixture of mono- and sesquiterpenes and 34 compounds accounting for nearly 97.4% of the oil were characterized with the help of capillary GC, GC-MS, and NMR. Major compounds of the oil were characterized as α -phellandrene (13.5%), *trans*-carveol (12.0%), β -pinene (11.7%), thujene (7.5%), β -caryophyllene oxide (7.2%), β -caryophyllene (7.0%), γ -terpinene (6.7%), nerolidol (6.5%), β -bisabolene (5.2%) and germacrene D (4.5%). It is the first report to exploit the essential oil from Himalayan *A. glauca* herb collected at flowering stage.

Keywords: Angelica glauca; North-West Himalaya; Essential oil composition; β-Pinene; α-Phellandrene; trans-Carveol

1. Introduction

Angelica (Apiaceae), a genus of tall, ornamental, perennial herbs, native to the North-Temperate region and New Zealand, are propagated from seeds and also vegetatively. In India three species viz. Angelica glauca, Angelica archangelica and Angelica cyclocarpa are found. A. glauca Edgew occurs wild in North-West Himalayas from Kashmir to Garhwal at an altitude of 1500–3700 m. The whole herb is reported to be useful as stimulant, cordial, appetizer, dyspepsia, cardioactive, carminative, expectorant, diaphoretic and also in stomach troubles, billious complaints, infantile atrophy, menorrhiza, for treating rinderpest and constipation, etc. (The Wealth of India, 1985; Polunin and Stainton,

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1984; Agarwal, 1986; Uniyal and Chauhan, 1971; Kirtikar and Basu, 1988; Bal, 1932; Jain and Tarafdar, 1970; Sharma et al., 1990). The roots of the plant are being used as a spice and condiment. Efforts are being made by some tribal peoples to cultivate it in farms (Satyal et al., 2002). This work is in continuation of the screening programme and chemical investigation of unexploited aromatic flora of N-W Himalaya for new sources of aromatic oils or aroma chemicals. The present work being appears to be the first report in literature on the chemical composition of the essential oil extracted from the flowering herb of *A. glauca*.

2. Results and discussion

A. glauca is found sparsely distributed in North-West Himalayas (India) and it is for the first time that

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the essential oil obtained from aerial parts of A. glauca at flowering stage is being investigated for its chemical components. The plant material collected from ten different populations from Uri, Khillanmarg areas on hydrodistillation yielded about 0.12% refreshing light pale coloured oil on moisture free basis (mfb) with characteristic floral woody flavour. Qualitatively and quantitatively GC pattern of all the oil samples was found to be nearly similar, therefore, a representative sample of herb from entire area was collected and chemically investigated. The oil was found to be a complex mixture of mono- and sesquiterpenes and 34 components were identified by RI, GC-MS and with the help of other spectroscopic methods, which accounted for 97.4% of the oil. Out of 34 identified compounds, ten accounted for nearly 81.8% of the oil vide GC (Table 1).

The flavour and essential oil composition of aerial parts was found to be widely different from the root essential oil collected from the same area. The *A. glauca*

root oil has been found to be rich in ligustilide and butylidene phthalides (Thappa et al., 2004). The characteristic major flavour compounds of A. glauca root oil were characterized as pthalides (ligustilides and butylidene phthalides), while these compounds were absent in A. glauca herb essential oil. Further, herb essential oil was found to be rich in monoterpenoids and 16 of these were common to root oil. The major compounds of A. glauca herb essential oil were characterized as terpene hydrocarbons like α -phellandrene (13.5%), β -pinene (11.7%), thujene (7.5%), β -caryophyllene (7.0%), γ -terpinene (6.7%), β -bisabolene (5.2%), germacrene D (4.5%) with oxygenated terpenes like trans-carveol (12.0%), β -caryophyllene oxide (7.2%) and nerolidol (6.5%).

The quality of essential oils extracted from different areas of Kashmir valley, indicated that there is no significant variation in chemical composition, and therefore oil of uniform quality may be produced from wild populations for its commercial exploitation.

Table 1 Chemical composition of *Angelica glauca* essential oil from flowering herb

Compounds	Retention index		%	Method of identification
	DB-5	DB-Wax		
Thujene	923	1052	7.5	MS, RI, ¹³ C NMR
α-Pinene	931	1046	0.5	MS, RI, ¹³ C NMR
Sabinene	964	1146	1.8	MS, RI, ¹³ C NMR
β-Pinene	970	1137	11.7	MS, RI, ¹³ C NMR
Myrcene	984	1181	0.1	RI, ¹³ C NMR
α-Phellandrene	995	1195	13.5	MS, RI, ¹³ C NMR
Δ^3 -Carene	1006	1162	0.7	MS, RI, Co-GLC
<i>p</i> -Cymene	1012	1303	0.1	MS, RI, Co-GLC
Limonene	1019	1210	0.2	MS, RI, Co-GLC
β-Phellandrene	1023	1250	0.2	MS, RI, Co-GLC
cis-Ocimene	1028	1264	0.5	MS, RI, ¹³ C NMR
trans-Ocimene	1037	1290	0.2	MS, RI, Co-GLC
γ-Terpinene	1049	1273	6.7	MS, RI, ¹³ C NMR
Linalool	1085	1584	0.2	MS, RI, Co-GLC
Camphor	1130	1552	0.6	MS, RI, Co-GLC
Borneol	1152	1746	0.1	MS, RI
Terpinen-4-ol	1161	1643	0.1	MS, RI, Co-GLC
α-Terpineol	1172	1741	0.2	MS, RI, Co-GLC
Menthol	1174	1628	0.2	MS, RI, Co-GLC
cis-Piperitol	1189	1770	0.6	MS, RI, ¹³ C NMR
trans-Carveol	1196	1853	12.0	MS, RI, ¹³ C NMR
Citronellol	1212	1803	0.3	MS, RI, Co-GLC
L-Carvone	1242	1790	0.2	MS, RI, Co-GLC
p-Cymene-8-acetate	1328	1680	0.7	MS, RI, ¹ H and ¹³ C NMR
trans-Carvyl acetate	1332	1687	0.4	MS, RI, ¹³ C NMR
β-Elemene	1390	1622	0.7	MS, RI
β-Caryophyllene	1413	1658	7.0	MS, RI, ¹³ C NMR
Alloaromadendrene	1464	1717	1.0	MS, RI, Co-GLC
Germacrene D	1471	1771	4.5	MS, RI, ¹³ C NMR
β-Bisabolene	1496	1786	5.2	MS, RI, ¹³ C NMR
trans-Nerolidol	1550	2056	6.5	MS, RI, ¹³ C NMR
β-Caryophyllene oxide	1565	2036	7.2	MS, RI, ¹³ C NMR
β-Eudesmol	1635	2290	2.5	MS, RI, ¹³ C NMR
(E,E)-Farnesol	1674	2415	3.5	MS, RI, ¹³ C NMR

3. Experimental

3.1. Plant material

The fresh aerial parts of *A. glauca* Edgew were collected at flowering stage in July 2002–2003 from Uri, and Khillanmarg areas of Kashmir (India, elevation, 1500–2100 m). The plants were identified and authenticated by the taxonomist and voucher specimen deposited at Regional Research Laboratory Herbarium, Jammu, India (Accession No. RRL-H 20094).

3.2. Extraction and isolation procedure

The essential oils were obtained by the hydrodistillation of fresh plant material in a Cleavenger type apparatus for three hrs. Each sample afforded refreshing light pale coloured oil with characteristic floral woody flavour (yield 0.06%; mfb 0.12%). The oils were dried over anhydrous sodium sulphate and placed at low temperature until further analysis. The oil was also prefractionated and fractions examined with the help of ¹H NMR (200 MHz) and ¹³C NMR (50.3 MHz), which were recorded on a Brucker model DPX-200 NMR spectrometer in deuterated benzene solution with TMS as an internal standard.

3.3. GC analysis

The composition of the oil was carried out by GC on two different gas chromatographs equipped with different capillary columns. The first was a Nucon 5765 gas chromatograph equipped with a FID and an AIMIL Chromatography Data processor. The separation was achieved using a DB-Wax (J&W Scientific, Folsom, CA, USA) fused-silica capillary column (30 m×0.25 mm i.d.; 0.25 µm film thickness). Column temperature, 90 °C (2 min) to 220 °C (5 min) with programming at 7 °C/min. "Injector temperature, 240 °C"; "detector temperature, 260 °C"; injection mode, split. Carrier gas was helium at column flow rate of 1.05 ml/min (100 kPa).

The second GC was on a Shimadzu GC-17 equipped with a DB-5 (J&W Scientific, Folsom, CA, USA) fused-silica capillary column (30 m \times 0.25 mm i.d.; 0.25 µm film thickness), using the same chromatographic conditions. Retention indices (RI) of the sample components and authentic compounds were determined on the basis of homologous *n*-alkane hydrocarbons under the same conditions.

3.4. GC/MS analysis and identification

GC/MS analyses were conducted using a Shimadzu QP 2000 using a DB-5 (J&W Scientific, Folsom, CA,

USA) capillary column (30 m \times 0.25 mm i.d.; 0.25 µm film thickness). Column temperature, 70 °C (2 min) to 220 °C (5 min) at 4 °C/min. "Injector temperature, 240 °C"; "Interface temperature, 250 °C"; acquisition mass range, 700–10 amu; ionization energy, 70 eV. Helium was used as carrier gas, 100 kPa (27.38 cm/s).

Peak identification was accomplished by comparison of their mass spectra with those reported in the literature (Adams, 1989). The identification of the oil components was also possible by comparison of their linear RI with those from literature and comparison of ¹³C NMR spectra obtained from the oil with those from literature (Kubeczka and Formacek, 2002).

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