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Assessment of similarities and dissimilarities in the essential oils of patchouli and Indian Valerian

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Two chemically alike essential oils derived from two disjunct genera namely patchouli (*Pogostemon cablin* (Blanco) Benth.) and Indian Valerian (*Valeriana jatamansi* Jones) were investigated for identification of marker compounds by gas chromatography–flame ionization detection (GC–FID) and GC–mass spectrometry (GC–MS). Inspite of huge chemical resemblance these two essential oils can be authenticated by their characteristics minor or trace constituents. The marker constituents identified for *P. cablin* oil were pogostone, pogostol, and (*Z*)-thujopsene, whereas marker constituents for *V. jatamansi* oil were 3-methyl valeric acid, thymol methyl ether, carvacrol methyl ether, bornyl acetate, kessane, maaliol, xanthorrhizol, and 8-acetoxy patchouli alcohol. These marker constituents may be utilized as an important tool in oil authentications.

Keywords: *Pogostemon cablin*; *Valeriana jatamansi*; essential oil; chemical differentiation; marker compounds

Introduction

Patchouli (*Pogostemon cablin* (Blanco) Benth.), belonging to the family Lamiaceae is an important aromatic plant, native to tropical Asia and is widely cultivated in India, Malaysia, Philippines, Indonesia, and Singapore. Patchouli oil, extracted from leaves is one of the best fixatives for heavy perfumes which impart strength, alluring notes and lasting characters. Indeed, patchouli oil is a perfume by itself and is highly valued in perfumes, soaps, cosmetics and flavor industries. Tenacity of odor is one of the great intrinsic worth of this oil and is one of the reasons for its versatility (1). Patchouli oil displays a rich, sweet, balsamic, herbaceous aroma with a fine earthy, heavy woody and slightly minty undertone (2, 3). The powerful woody odor of the patchouli oil is mainly due to (–)-patchoulol, (+)-norpatchoulenol, and pogostol (3, 4). Patchouli oil is also known to possess antifungal properties and is being used in skin infections, dandruff and eczema (5–7). Indian Valerian, *Valeriana jatamansi* Jones (Valerianaceae) commonly known as 'Tagar' is a perennial herbaceous plant growing abundantly in the subtemperate and temperate regions of the Himalaya. The roots of the plant are highly aromatic and it is used as an ingredient in Indian herbal medicines, and as a substitute of the European *V. officinalis*. Ancient Indian Ayurvedic and Unani literature describe the medicinal properties of 'Tagar' for curing obesity, nervous disorders, epilepsy, insanity, snake poisoning, eye trouble, and skin diseases. It is still a highly reputed medicinal plant described in many pharmacopoeia monographs (8). The essential oil of Indian Valerian has a warm-woody, balsamic root-like odor with distinct animal undertone of musk like character and great tenacity. A fresh green slightly camphoraceous top note is also typical in odor of good oil (9). Patchouli and Indian Valerian both are reported to possess patchouli alcohol as a main constituent in their essential oil (10, 11). Presence of inter- and intra-specific variations at phenotypic, chemotypic or at genotypic level as well as similarity among different genera/species at chemotypic level in aromatic plant species is very common and it has been pointed out by many researchers (12–19).

Similar compositions of essential oils or aroma chemicals from disjunct genera/species of aromatic plants serve as alternative sources and simultaneously share industrial pressure, but at the same time, they may also create problems of adulterations (mixing the cheaper essential oils to chemically similar highly priced essential oils) (20). However, as a solution to the problem of these adulterations, they can easily be detected by knowing the presence or absence of some minor or trace constituents (marker constituents) in the different essential oils. The modern analytical techniques, gas chromatography (GC) and gas chromatography–mass spectrometry (GC–MS) are capable of detecting this kind of mixing easily. To address this problem our present study attempts to assess the chemical similarities as well as

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dissimilarities in chemically alike essential oils of two industrially important disjunct aromatic plants namely *Pogostemon cablin* and *Valeriana jatamansi*.

Experimental

Plant material

Plant materials of *Pogostemon cablin* (leaves) and *Valeriana jatamansi* (roots) were collected during the rainy season from an experimental field of the Central Institute of Medicinal and Aromatic Plants (CIMAP), Research Center, Purara Uttarakhand. In the present study, *P. cablin* var. *CIM-Samarth* developed by CIMAP (21) and *V. jatamansi* (patchouli alcohol rich populations), collection from the wild (Voucher Specimen No. CIMPANT-734) have been used. The experimental site is located at an altitude of 1250 m in the Kattyur valley, western Himalayas. Climatologically, it is categorized as a temperate zone.

The essential oil of both the plants was extracted in replicates (three or more random samples) by hydrodistillation for 4 hours using a Clevenger apparatus. The percentage essential oil content (%*v/w*) was estimated on a fresh weight basis. The oil sample obtained was dehydrated over anhydrous sodium sulfate (Na_2SO_4) and kept in a cool and dark place before analyses.

Analysis of the essential oils

The GC analyses of the oil samples was carried out on Nucon gas chromatograph model 5765 equipped with a flame ionization detector and DB-5 (30 m × 0.32 mm; 0.25 μm film coating) fused silica capillary column. Hydrogen was the carrier gas at 1.0 mL/minute. Oven temperature programming was done from 60°C to 230°C at 3°C/minute. The injector and detector temperatures were 220°C and 230°C, respectively. The injection volume was 0.02 μL neat (syringe: Hamilton 1.0 μL capacity, Alltech, USA) and the split ratio was 1:40.

GC–MS analysis of the essential oil sample was carried out on a Perkin–Elmer AutoSystem XL GC interfaced with a Turbomass Quadrupole Mass Spectrometer fitted with a DB-5 fused silica capillary column (60 m × 0.32 mm i.d., film thickness 0.25 μm). The oven temperature was programmed from 60°C to 210°C at 3°C/minute using helium as the carrier gas at 1.0 mL/minute. The injector temperature was 210°C, injection volume 0.1 μL prepared in *n*-hexane, split ratio 1:40. MS were taken at 70 eV with a mass scan range of 40 to 450 amu.

Constituents were identified on the basis of a arithmetic retention index (RI, determined with reference to homologous series of *n*-alkanes, C_8–C_{30}), under identical experimental conditions, co-injection with standards or known essential oil constituents, MS Library search (NIST and WILEY), by comparing with the MS literature data (22). The relative amounts of individual components were calculated based on the GC peak area (flame ionization detection (FID) response) without using a correction factor.

Results and discussion

Essential oil yield and GC and GC–MS analysis results of *Pogostemon cablin* (leaves) and *Valeriana jatamansi* (roots) are summarized in Table 1. Chromatograms of the *P. cablin* and *V. jatamansi* oils are shown in Figure 1 and 2. The essential oil yield observed in fresh leaves of *P. cablin* and fresh roots of *V. jatamansi* was 0.32% and 0.39%, respectively. Major components identified in the essential oil of *P. cablin* were patchouli alcohol (61.6%), *α*-guaiene (10.0%), *α*-bulnesene (8.7%), seychellene (3.1%), (*E*)-caryophyllene (2.4%), *α*-patchoulene (1.6%), norpatchoulenol (1.6%), *γ*-patchoulene (1.3%), *β*-patchoulene (1.0%), and pogostone (1.0%). Likewise *P. cablin* oil, the essential oil of *V. jatamansi* was also composed of mainly patchouli alcohol (48.5%). However, other major components of *V. jatamansi* oil were *α*-bulnesene (7.6%), guaiol (5.9%), *α*-guaiene (4.3%), seychellene (2.8%), maaliol (1.9%), *α*-patchoulene (1.7%), 7-*epi*-*α*-selinene (1.5%), 8-acetoxy patchouli alcohol (1.4%), bornyl acetate (1.3%), *β*-elemene (1.3%), bulnesol (1.3%), and kessane (1.2%). According to the literature patchouli alcohol (17.5–48.7%), *α*-guaiene (10.9–19.7%), *α*-bulnesene (13.4–20.7%), seychellene (5.3–8.9%), *α*-patchoulene (4.8-7.8%), *γ*-patchoulene (0.2-6.7%), (*E*)-caryophyllene (1.8-5.1%), pogostol (0.2-2.4%), and pogostone (0.1–1.1%) were major constituents of commercial patchouli oil from different countries (23). Diversity in the essential oil composition of *V. jatamansi* has also been assessed earlier and principal components namely, patchouli alcohol (13.4–66.7%), *α*-bulnesene (< 0.05–23.5%), *α*-guaiene (0.2–13.3%), guaiol (< 0.05–12.2%), seychellene (0.2–9.9%), viridiflorol (< 0.05–7.3%), *β*-gurjunene (nil–7.1%), (*E*)-caryophyllene (< 0.05–5.1%), etc. showed considerable variation in their quantitative composition (24).

As far as the similarity of both oils was concerned, out of fifty-one characterized components a total of twenty-three components (90.3% of *Pogostemon cablin* and 78.5% of *Valeriana jatamansi*) were found to be common in both oils. The characteristic components of *P. cablin* oil which was not noticed in *V. jatamansi* oil were pogostone (1.0%), pogostol (0.5%), (*Z*)-thujopsadiene (0.3%), and (*Z*)-thujopsene (trace). Furthermore, characteristic components for *V. jatamansi* oil which were not observed in *P. cablin* oil were isovaleric acid (0.5%), 3-methyl valeric acid (0.9%), thymol methyl ether (0.1%), carvacrol methyl ether (0.1%), bornyl acetate (1.3%), kessane (1.2%), maaliol (1.9%),

Table 1. Similarities and differences in the chemical profiles of *Pogostemon cablin* and *Valeriana jatamansi* essential oils.

Compound	RI[a]	RI[b]	Content (%)[c]	
			Pogostemon cablin	*Valeriana jatamansi*
Isovaleric acid [MV]	830	827	—	0.5 (0.41)
α-Pinene	935	932	t	0.3 (0.30)
3-Methyl valeric acid [MV]	944	939	—	0.9 (0.72)
1-Octen-3-ol	979	974	0.4 (0.15)	—
β-Pinene	979	974	—	0.1 (0.08)
Myrcene	989	988	t	—
p-Cymene	1023	1020	t	0.1 (0.05)
Limonene	1025	1024	—	0.1 (0.05)
γ-Terpinene	1050	1044	t	t
Borneol	1161	1165	t	t
α-Terpineol	1184	1186	t	t
Thymol methyl ether [MV]	1232	1232	—	0.1 (0.13)
Carvacrol methyl ether [MV]	1243	1241	—	0.1 (0.04)
Bornyl acetate [MV]	1277	1284	—	1.3 (0.62)
α-Copaene	1373	1374	0.1 (0.11)	0.3 (0.28)
β-Patchoulene	1377	1379	1.0 (0.12)	0.2 (0.17)
β-Elemene	1392	1389	0.8 (0.25)	1.3 (0.52)
β-Longipinene	1398	1400	t	0.7 (0.77)
Cycloseychellene	1401	1406	0.4 (0.06)	—
α-Santalene	1414	1416	—	0.1 (0.12)
(*E*)-Caryophyllene	1416	1417	2.4 (0.29)	0.1 (0.08)
(*Z*)-Thujopsene [MP]	1428	1429	t	—
α-Guaiene	1442	1437	10.0 (1.97)	4.3 (1.61)
Seychellene	1448	1444	3.1 (0.35)	2.8 (1.13)
α-Patchoulene	1453	1454	1.6 (0.00)	1.7 (1.71)
(*Z*)-Thujopsadiene[#]	1466	1465	0.3 (0.06)	—
γ-Gurjunene	1473	1475	0.2 (0.06)	0.1 (0.04)
γ-Muurolene	1477	1478	0.2 (0.23)	0.4 (0.42)
Germacrene-D	1478	1484	0.1 (0.04)	—
(*Z*)-β-Guaiene	1489	1492	—	0.2 (0.05)
Valencene	1492	1496	0.3 (0.12)	0.6 (0.40)
α-Selinene	1494	1498	t	0.7 (0.57)
γ-Patchoulene	1497	1502	1.3 (0.25)	—
α-Bulnesene (=δ-Guaiene)	1516	1509	8.7 (2.59)	7.6 (2.33)
7-*epi*-α-Selinene	1518	1520	0.1 (0.04)	1.5 (0.24)
δ-Cadinene	1526	1522	0.1 (0.11)	0.1 (0.05)
Kessane [MV]	1528	1529	—	1.2 (0.67)
α-Cadinene	1537	1537	t	—
Norpatchoulenol	1549	1553	1.6 (0.10)	—
Maaliol [MV]	1566	1566	—	1.9 (2.67)
Longipinanol	1567	1567	t	—
Caryophyllene oxide	1585	1582	0.3 (0.21)	—
Viridiflorol	1594	1592	—	0.6 (0.50)
Guaiol	1596	1600	t	5.9 (5.47)
Pogostol [MP]	1650	1651	0.5 (0.10)	—
Patchouli alcohol	1665	1656	61.6 (7.32)	48.5 (7.46)
Bulnesol	1676	1670	0.1 (0.06)	1.3 (0.50)
Pogostone [MP]	1720	—	1.0 (0.84)	—
Xanthorrhizol [MV]	1752	1751	—	0.3 (0.22)
Xanthorrhizol isomer [MV]	1868	—	—	0.1 (0.13)
8-Acetoxy patchouli alcohol [MV]	2004	—	—	1.4 (2.68)
Total identified			96.2 (154)	87.4 (6.57)
Essential oil yield (%)			0.32 (0.01)	0.39 (0.04)

Notes: [a]RI, calculated retention index on DB-5 column; [b]RI, literature value of retention index (16); [c]values in parentheses are standard deviations; [MV]marker compound characteristic of *V. jatamansi* root oil; [MP]marker compound characteristic of *P. cablin* leaf oil; [#]tentative identification.

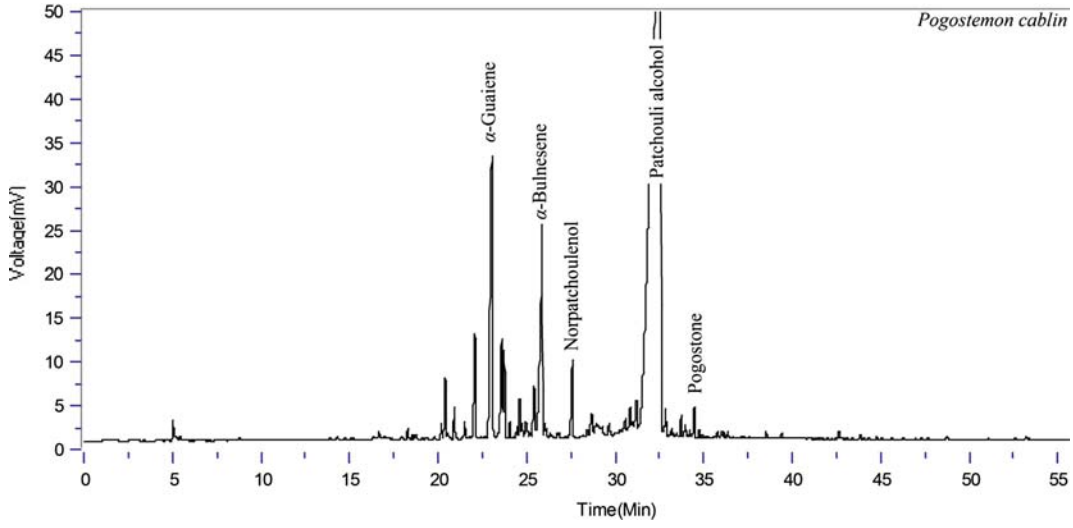

Figure 1. Chromatogram (GC) of *Pogostemon cablin* leaf volatile oil.

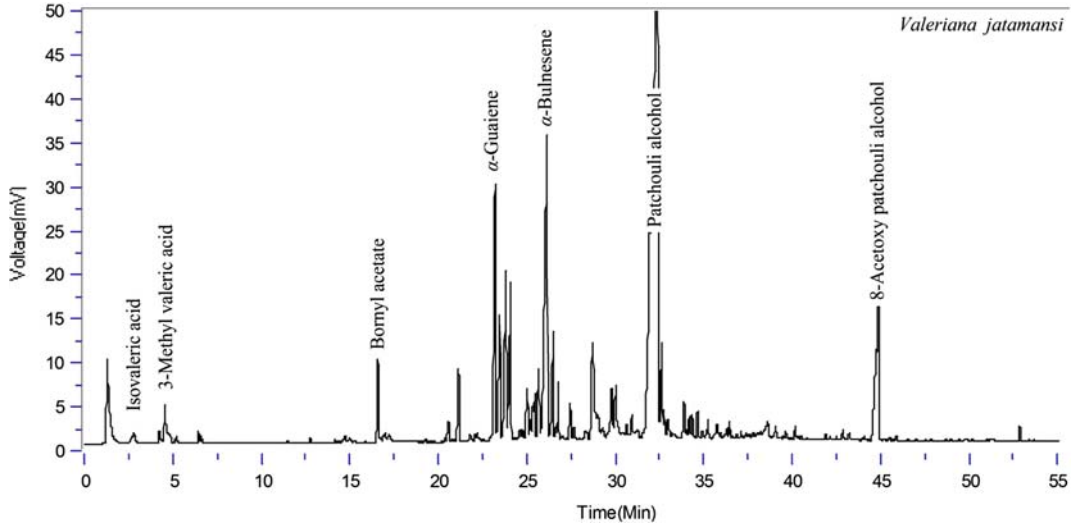

Figure 2. Chromatogram (GC) of *Valeriana jatamansi* root volatile oil.

xanthorrhizol (0.3%), xanthorrhizol isomer (0.1%), and 8-acetoxy patchouli alcohol (1.4%). However, recently Cornwell (25) has identified isovaleric acid in trace amount in commercial patchouli oil from Asian countries except Indian oil.

The use of secondary metabolites as chemical markers plays an important role in determining the relationship within the species of a genus and also outside of a particular genus, especially in evolution studies focusing on phylogenic and inter-taxonomic variations (26). Biosynthesis of similar molecules in different plants has been reported earlier (12, 14, 15, 18). This is probably due to the presence of similar enzyme systems and the compounds which they produce may indicate a relationship between the plants.

The essential oils obtained from *Pogostemon cablin* and *Valeriana jatamansi* are in great demand in perfumery, flavor, and pharmaceutical industries. Further, patchouli alcohol had shown interesting pharmacological activities such as antimicrobial and anti-emetic (27, 28). On the basis of present results; pogostone, pogostol, and (*Z*)-thujopsene may be identified as marker constituents of *P. cablin* oil, whereas 3-methyl valeric acid, thymol methyl ether, carvacrol methyl ether, bornyl acetate, kessane, maaliol, xanthorrhizol, and 8-acetoxy patchouli alcohol can be used as marker constituents for *V. jatamansi* oil. These marker constituents in their essential oils are not only responsible for the characteristic olfactory note of the oils; but they can also be utilized as an important tool in oil authentication. Presently, only

P. cablin is being used as a commercial source of patchouli alcohol, whereas *V. jatamansi* is basically known for its reputation in traditional medicines, but its potential to deliver essential oil as a source of patchouli alcohol is still overlooked. Thus, *V. jatamansi* could be a potent source of patchouli alcohol rich essential oil apart from being used in traditional medicines.

On the basis of the present study it can be said that inspite of huge chemical resemblance, *Pogostemon cablin* and *Valeriana jatamansi* essential oils differed considerably with each other. Thus, any mixing or substitution of these oils with one another could be noticeable by the presence or absence of respective marker constituents.

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