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The essential oils of *Thymus migricus* and *T. fedtschenkoi* var. *handelii* from Turkey†

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ABSTRACT: The essential oils of *Thymus migricus* Klokov et Des.-Shost. and *T. fedtschenkoi* Ronniger var. *handelii* (Ronniger) Jalas were obtained by hydrodistillation, and analysed by GC–MS. The main components in the essential oils were found to be carvacrol and thymol and linalool, respectively. Copyright © 2001 John Wiley & Sons, Ltd.

KEY WORDS: Lamiaceae; *Thymus migricus*; *Thymus fedtschenkoi* var. *handelii*; essential oil; linalool; thymol; carvacrol

Introduction

The genus *Thymus* (Lamiaceae) is represented in Turkey by 38 species and altogether 64 taxa, 24 of which are endemic.[1,2] In Turkey, members of this genus are called 'kekik' or 'taş kekik' and their dried herbal parts are used in herbal tea, condiments and folk medicine. The essential oils of some *Thymus* spp. are characterized by the presence of high concentrations of the isomeric phenolic monoterpenes thymol and/or carvacrol. However, there are *Thymus* spp. poor in phenolic compounds and some may not even contain any phenolic compounds at all. Previous studies have shown the occurrence of these three types of essential oils on 34 *Thymus* taxa growing in Turkey.[3] In Turkey, other carvacrol or thymol-rich genera, such as *Origanum, Thymbra, Coridothymus* and *Satureja*, are export commodities under the name 'kekik'. *Thymus* spp., however, are not used as a source of essential oil but they are mainly used in herbal tea in the areas where they grow.[4]

Monoterpenic phenol-rich *Thymus* spp. are used in herbal tea against cough, diabetes, stomach and intestinal diseases and as a condiment. Especially monoterpenic phenol-poor or monoterpenic phenol-less *Thymus* spp. are used in herbal tea due to their pleasant aroma. *T. migricus*, of Azerbaijan origin, was reported to yield 0.2–0.6% oil from herbal parts during different stages of vegetation, containing carvacrol (36%) and thymol (13%) as main constituents.[5] In the only previous study on the oil of *T. fedtschenkoi* var. *handelii* from Turkey, linalool (17%) was reported as the main constituent.[6]

In this study, we have investigated the hydrodistilled essential oils of *T. migricus* Klokov et Des.-Shost. and of the endemic *T. fedtschenkoi* var. *handelii* (Ronniger) Jalas. These species are considered similar taxonomically.[1]

Experimental

Plant Material

Information about the plant material is given in Table 1. Voucher specimens are kept at the Herbarium of the Faculty of Pharmacy of Anadolu University in Eskişehir, Turkey (ESSE).

Isolation of the Essential Oils

Air-dried plant material was hydrodistilled for 3 h using a Clevenger-type apparatus to yield an essential oil on a dry weight basis. Oil yields are shown in Table 1.

Analysis of Essential Oils

The essential oils were analysed using a Hewlett-Packard G1800A GCMSD system. An HP-Innowax FSC column (60 m × 0.25 mm i.d., film thickness 0.25 μm) was used

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Table 1. Collection site, dates, plant part, ESSE number and oil yields of *Thymus* spp. studied

Code	*Thymus* spp.	Collection site	Collection date	Plant part	ESSE No.	Oil yield (v/dry weight)
A	*T. migricus*	Ağri	20.8.95	Herb	11950	0.29
B	*T. migricus*	Van	23.6.99	Herb	12272	1.47
C	*T. migricus*	Van	28.6.96	Herb	12174	1.8
D	*T. migricus*	Van	20.7.99	Leaf + flower	13020	*
E	*T. fedtschenkoi* var. *handelii*	Bitlis	17.7.98	Herb	13029	0.23

* Recovered with hexane due to paucity of oil.

Table 2. Percentages and composition of the essential oils of *Thymus migricus* and *T. fedtschenkoi* var. *handelii*

		Thymus migricus				*Thymus* fedtschenkoi var. *handelii*
RRI	Compound	A Ağr1	B Van	C Van	D Van	E Bitlis
1032	α-Pinene	0.1	1.4	0.1	—	1.7
1035	α-Thujene	0.1	0.7	tr	—	0.1
1076	Camphene	0.2	0.7	tr	—	0.9
1118	β-Pinene	0.1	0.4	0.1	—	0.4
1132	Sabinene	0.1	tr	tr	—	0.3
1174	Myrcene	0.2	1.7	0.4	—	0.3
1188	α-Terpinene	0.3	2.8	0.7	—	0.4
1195	Dehydro-1,8-cineole	—	—	—	—	0.3
1203	Limonene	0.3	0.4	0.2	—	1.3
1213	1,8-Cineole	2.8	3.1	2.0	2.5	5.9
1218	β-Phellandrene	0.1	0.3	0.1	—	tr
1246	(*Z*)-β-Ocimene	—	tr	—	—	0.1
1255	γ-Terpinene	0.9	11.6	4.6	0.5	1.7
1265	(*E*)-β-ocimene	—	tr	tr	—	0.9
1266	5-Methyl-3-heptanone	0.8	1.1	0.7	—	0.2
1280	*p*-Cymene	5.6	12.9	5.5	0.9	3.9
1290	Terpinolene	tr	0.2	0.1	—	0.3
1345	3-Octyl acetate	—	—	—	—	0.1
1367	3-Nonanone	0.1	tr	tr	—	tr
1386	Octenyl acetate	—	—	—	—	0.1
1393	3-Octanol	0.4	0.1	0.1	—	0.2
1400	Nonanal	—	tr	—	—	0.1
1408	1,3,8-*p*-Menthatriene	—	—	—	—	tr
1415	Rosefuran	—	—	—	—	tr
1429	Perillen	—	—	—	—	tr
1435	γ-Campholene aldehyde	—	—	—	—	0.1
1446	2,6-Dimethyl-1,3(*E*),5(*Z*),7-octatetraene	—	—	—	—	tr
1450	*trans*-Linalool oxide (furanoid)	—	—	—	—	0.7
1451	β-Thujone	—	—	—	—	tr
1452	1-Octen-3-ol	0.2	0.2	0.3	—	0.1
1460	2,6-Dimethyl-1,3(*E*),5(*E*),7-octatetraene	—	—	—	—	tr
1463	Heptanol	—	—	—	—	tr
1465	Eucarvone	—	—	—	—	tr
1474	*trans*-Sabinene hydrate	0.7	0.8	1.1	0.9	5.8
1475	Menthone	4.7	—	—	—	—
1478	*cis*-Linalool oxide (furanoid)	—	—	—	—	0.5
1495	Bicycloelemene	—	—	—	0.1	tr
1496	3-Nonanol	tr	—	—	—	—
1497	α-Copane	—	0.3	0.1	0.1	—
1498	(*E*)-β-Ocimene epoxide	—	—	—	—	tr
1503	Isomenthone	4.6	—	—	—	—
1506	Decanal	—	—	—	—	0.1
1507	(*E,E*)-2,4-Heptadienal	—	tr	0.1	—	—
1532	Camphor	0.1	—	—	2.7	5.7
1535	β-Bourbonene	0.3	0.4	0.3	—	0.1
1553	Linalool	0.1	0.1	0.1	—	**12.9**
1556	*cis*-Sabinene hydrate	0.4	0.4	0.4	0.3	—
1562	Octanol	—	—	tr	—	0.1

Table 2. (*Continued*)

		Thymus migricus				*Thymus* fedtschenkoi var. *handelii*
RRI	Compound	A Ağr1	B Van	C Van	D Van	E Bitlis
1565	Linalyl acetate	—	—	—	—	tr
1571	*trans*-*p*-Menth-2-en-1-ol	—	tr	tr	—	0.3
1586	Pinocarvone	—	—	—	—	0.2
1588	Bornyl formate	—		—	—	0.5
1597	Bornyl acetate	0.9	0.1	tr	3.4	2.7
1598	Thymol methyl ether	—	0.2	0.1	—	3.0
1598	*trans*-Isopulegone	0.1	—	—	—	—
1611	Terpinen-4-ol	0.6	—	—	—	3.7
1612	β-Caryophyllene	0.3	1.9	1.8	5.3	—
1614	Carvacrol methyl ether	2.6	0.5	0.5	4.2	0.4
1624	*cis*-Dihydrocarvone	0.3	—	—	0.5	tr
1628	Aromadendrene	—	0.5	0.4	—	—
1638	*cis*-*p*-Menth-2-en-1-ol	—	tr	tr	—	0.1
1645	*trans*-Dihydrocarvone	0.2	tr	tr	—	0.2
1661	Alloaromadendrene	—	tr	tr	0.4	0.3
1662	Pulegone	9.5	—	—	0.4	—
1663	*cis*-Verbenol	—	—	—	—	0.2
1665	*trans*-Pinocarveol	0.1	—	tr	—	0.6
1671	(*E*)-β-Farnesene	0.1	—		—	—
1677	*epi*-Zonarene	—	0.1	0.1	—	—
1683	δ-Terpineol	0.1	0.1	0.1	0.3	0.5
1687	α-Humulene	—	0.1	0.1	0.4	—
1691	*trans*-Verbenol	0.1	tr	tr	—	1.8
1694	*p*-Vinylanisole	—	—	—	—	0.1
1697	Carvotanacetone	—	—	tr	—	—
1704	γ-Muurolene	0.2	0.6	0.4	0.3	—
1707	α-Terpineol	0.4	0.4	0.5	0.6	6.0
1707	Ledene	—	0.3	0.3	—	—
1709	α-Terpinyl acetate	—	—	—	—	4.9
1719	Borneol	6.9	2.2	1.1	8.4	5.1
1725	Verbenone	—	—	—	—	0.2
1726	Germacrene-D	0.9	—	—	4.2	0.8
1737	Carvenone	—	tr	tr	—	—
1740	*trans*-*p*-Menth-2-en-1,8-diol	—	—	—	—	0.2
1740	Valencene	0.1	0.2	—	0.2	—
1741	β-Bisabolene	0.7	1.0	1.1	1.3	0.2
1744	α-Selinene	—	tr	—	—	—
1748	Piperitone	3.7	—	—	—	0.1
1751	Carvone	0.1	tr	tr	—	—
1755	Bicyclogermacrene	—	—	—	6.8	
1758	*cis*-Piperitol	—	tr	tr	—	0.2
1766	Decanol	tr	—	0.1	—	—
1773	δ-Cadinene	0.4	0.7	0.6	0.7	0.1
1776	γ-Cadinene	0.2	0.3	0.3	0.4	0.1
1783	β-Sesquiphellandrene	—	tr	tr	0.1	—
1797	*cis*-*p*-Ment-2-en-1,8-diol	—	—	—	—	0.2
1798	Methyl salicylate	—	tr	tr	—	—
1799	Cadina-1,4-diene	—	0.1	0.1	—	—
1802	Cumin aldehyde	0.1	—	—	—	0.1
1804	Myrtenol	—	—	tr	—	0.2
1811	*trans*-*p*-Mentha-1(7),8-dien-2-ol	—	tr	—	—	0.1
1819	Geranyl isobutyrate	—	—	tr	0.1	0.1
1830	2,6-Dimethyl-3(*E*),5(*E*),7-octatriene-2-ol	—	—	—	—	0.2
1838	β-Damascenone	—	—	—	—	tr
1845	*trans*-Carveol	—	—	—	—	0.6
1853	*cis*-Calamenene	0.1	0.1	tr	—	—
1857	Geraniol	—	—	—	—	0.1
1864	*p*-Cymen-8-ol	0.1	0.1	0.1	—	0.3
1867	Thymyl acetate	0.1	0.1	—	—	—
1882	*cis*-Carveol	—	—	—	—	0.1
1889	Ascaridole	—	tr	0.1	—	—
1900	Epicubebol	tr	tr	tr	—	0.1

(*continued overleaf*)

Table 2. (*Continued*)

		Thymus migricus				*Thymus* fedtschenkoi var. *handelii*
RRI	Compound	A Ağr1	B Van	C Van	D Van	E Bitlis
1901	Geranyl butyrate	—	—	tr	0.2	0.2
1904	Geranyl 2-methylbutyrate	—	—	tr	0.2	0.1
1940	α-Calacorene	—	0.1	0.1	—	—
1941	4-Isopropyl salicylaldehyde	—	—	tr	—	—
1949	Piperitenone	5.3	—	—	—	—
1953	Shyobunol	—	—	—	0.6	
1957	Cubebol	0.1	tr	tr	—	tr
1958	β-Ionone	—	tr	—	—	tr
1984	γ-Calacorene	—	tr	tr	—	—
2001	Isocaryophyllene oxide	—	—	—	—	0.3
2008	Caryophyllene oxide	—	0.2	0.2	0.8	4.5
2029	Perilla alcohol	—	—	—	—	0.1
2037	Salvial-4(14)-en-1-one	—	—	—	—	tr
2046	Norbourbonone	—	—	—	—	0.1
2050	(*E*)-Nerolidol	—	—	—	—	1.1
2057	Ledol	—	—	—	0.2	0.1
2069	1(10),5-Germacradien-4β-ol	—	—	—	1.1	0.6
2098	Globulol	—	tr	tr	0.1	—
2104	Viridiflorol	—	—	—	—	0.2
2105	Geranyl hexanoate	—	—	—	0.1	—
2113	Cumin alcohol	—	tr	tr	—	—
2131	Hexahydrofarnesylacetone	0.1	—	tr	—	0.1
2144	Spathulenol	—	0.3	0.3	2.2	1.8
2181	Isothymol (=2-Isopropyl-4-methylphenol)	—	tr	0.1	—	0.1
2186	Eugenol	0.1	—	—	—	—
2187	T-Cadinol	0.1	—	—	—	0.2
2198	Thymol	2.6	**44.2**	36.3	**38.9**	2.9
2239	Carvacrol	**36.3**	4.1	**36.5**	7.9	0.8
2247	*trans*-α-Bergamotol	—	tr	—	—	0.1
2255	α-Cadinol	—	0.1	—	0.2	0.2
2256	Cadalene	—	tr	—	—	—
2300	Tricosane	—	—	—	—	0.1
2324	Caryophylladienol-II (=caryophylla-2(12),6(13)-diene-5α-ol)	—	—	—	0.1	0.4
2392	Caryophyllenol-II (=*Caryophylla-2(12), 6-diene-5β-ol*)	—	—	—	—	0.7
2500	Pentacosane	—	—	tr	—	0.1
	Total	96.7	98.2	98.3	98.6	94.7

RRI Relative retention indices calculated against *n*-alkanes.
%, calculated from TIC data.
tr, trace (<0.1%).

with helium (1 ml/min) as carrier gas. GC oven temperature was kept at 60 °C for 10 min, programmed at 4 °C/min to 220 °C, kept constant at this temperature for 10 min, and subsequently programmed at 1 °C/min to 240 °C. Mass range was recorded at m/z 35–425. Alkanes were used as reference points in the calculation of relative retention indices (RRIs). The split ratio was adjusted to 50 : 1. The injection port temperature was 250 °C. MSs were taken at 70 eV. A library search was carried out using the Wiley GC–MS Library and the TBAM Library of Essential Oil Constituents. Relative percentage amounts of the separated compounds were calculated automatically from peak areas of the total ion chromatograms. The MSs were also compared with reference compounds and confirmed with the aid of retention index sources.

Results and Discussion

In the GC–MS analysis of the oil (see Table 2) of *T. migricus* from Ağri (A), 60 compounds, representing 97% of the total oil, were characterized, carvacrol (36%) being the major component. In the other oils obtained from *T. migricus* samples collected from Van province (B–D), 40–75 compounds, representing 98–99% of the oils, were characterized. Thymol (36–44%) was found as the major component. However, in one sample (C) carvacrol (37%) was the main constituent. In the only previous work, carvacrol and thymol were reported as the main constituents.[5] In the same work, best oil yields (0.4–0.6%) were obtained with flowering herbs and the worst yields (0.2–0.3%) in materials collected during late flowering and fruiting stage.[5] During the present

study, although the plants were collected from different localities, somewhat similar results were obtained. Flowering plant samples gave the best yields (1.5% and 1.8% for B and C, respectively) and much lower yields (trace and 0.3% for D and A, respectively) were observed with samples collected at late flowering or fruiting stage.

In the essential oil of *T. fedtschenkoi* var. *handelii* (E), 107 compounds, representing 95% of the total oil, were characterized. Linalool (13%) was the major component of this oil, as reported previously.[6]

The oils studied here fall into the first group of Stahl–Biskup's classification of *Thymus* oils, which is characterized by the high percentage of thymol, carvacrol, linalool, linalyl acetate, borneol, *p*-cymene, 1,8-cineole, γ-terpinene and camphor.[7]

According to a recent treatise published by our group, 80% of the Turkish *Thymus* spp. were found to fall into this group and the two species currently studied are no exception.[3] *Thymus migricus* is characterized by high concentrations of thymol and carvacrol in its oil. Although only one sample of *T. fedtschenkoi* var. *handelii* was studied, a clear distinction could be made due to the presence of a high percentage of linalool and low amounts of phenolic monoterpenes in its oil. As the two species are morphologically quite similar, the possible diagnostic value of this finding is clear, since linalool was previously reported also as main constituent of the oil of *T. fedtschenkoi* var. *handelii*.[6]

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