The myrtenol chemotype of essential oil of *Tanacetum vulgare* L. var. *vulgare* (tansy) growing wild in the Vilnius region

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Ten samples of inflorescences and leaves of *Tanacetum vulgare* L. var. *vulgare* were collected in 3 habitats at full flowering. The essential oils were analysed by GC/MS. Four out of 5 inflorescence essential oils were of a myrtenol chemotype and the same quantity of the leaf oils were of a 1,8-cineole type. The amounts of 2 major constituents in the inflorescence and leaf oils were 14.1–24.9% and 8.2–15.8% of myrtenol and 11.6–16.5% and 14.5–27.2% of 1,8-cineole, respectively. The other main components such as camphor made up 8.5–17.8% in 8 oils, α-pinene – 4.9–9.6% in 6 samples, terpinen-4-ol – 6.4–15.0% in 5 oils and borneol – 5.9–14.9% in 3 volatile oils. Trans-pinocarveol (15.9–16.1%) and trans-sabinyl acetate (5.0–9.9%) produced by all areal parts of plants of one habitat were the main constituents. Oxygenated monoterpenes made up 69.1–75.0% of inflorescence oils and 62.5–66.7% of leaf oils. Sixty-five identified constituents made up 80.5–98.6% of oil content. The myrtenol chemotype essential oils produced by tansy plants are very rare.

Key words: *Tanacetum vulgare* L., *Compositae*, myrtenol, 1,8-cineole, trans-pinocarveol, camphor, terpinen-4-ol

INTRODUCTION

Plants of *Tanacetum vulgare* L. grow wild in the northern hemisphere [1]. The bioactivity and main applications of tansy essential oil and plant extracts are presented in [1–10]. Tansy plants are cultivated in gardens and used as an aromatic and spicy herb. *T. vulgare* was traditionally used in culinary, cosmetics, dyes, insecticides, preservatives, acaricides and medicines. It was used as herbal remedy for healing migraine, neuralgia, rheumatism, digestion diseases and loss of appetite without documentation of the effectiveness for these uses. Recent investigations have demonstrated that oils or extracts of tansy exhibit ant-inflammatory, bactericidal, fungicidal, acaricidal and repellent activity. The bioactivity and odour of tansy depends on the chemical composition of the essential oils.

The papers on the composition of tansy volatile oils published up to 1996 were reviewed in [1, 11]. Twenty-three chemotypes were determined according to the first dominant constituent. The β-thujone chemotype was identified in 8 countries (Argentina, Belgium, Canada, Germany, Holland, Hungary, Finland and Italy), while α-thujone – in 3 countries (France, Germany and Italy). The form of thujone isomer has not been determined in some papers from such countries as Brazil, Finland, India and the UK. Plants producing oils of camphor type were collected in 7 (Belgium, Germany, Holland, Hungary, Kazakhstan and USA) and trans-chrysanthenyl acetate in 5 (Belgium, Finland, Holland, Hungary and Italy) countries. The 1,8-cineole and artemisia ketone chemotypes have been found in Holland, Finland and Hungary. Tansy oils from Finland, Hungary and USA were of the bornyl acetate type. The other 16 chemotypes were differentiated according to the major component such as sabinene, thujyl alcohol, umbellulone, thujyl acetate, α-pinene, chrysanthenone, chrysanthenone oxide, isopinocamphone, borneol, γ-terpinene, piperitone, trans-dihydrocarvone, lyratol, lyratyl acetate, davanone and germacrene D. These types of tansy oils were found only in one or two countries. Some further papers on tansy oil have been reported after 1996 [7, 8, 12, 13], but have not found no new chemotypes in them.

*T. vulgar*e plants were most carefully analyzed in Finland (17 chemotypes), Hungary (14 chemotypes)

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and Holland (10 chemotypes). Four types of tansy oils were found in the Vilnius district [13]. Twenty samples of inflorescence and leaf oils were analyzed. Ten oils were of camphor type, 6 samples – of α-thujone, 3 ones– of 1,8-cineole and 1 sample of artemisia ketone chemotype.

Only one species of the genus *Tanacetum*, *Tanacetum vulgare* L. (syn. *Chrysanthenum vulgare* L.), is growing wild in Lithuania [14]. The plant is widespread all over the country. Two varieties of the plant, var. *vulgare* and var. *crispum*, were found.

Investigations of some new habitats of *T. vulgare* in the Vilnius region have demonstrated the existence of the fifth myrtenol chemotype of essential oils in Lithuania.

MATERIALS AND METHODS

The aerial parts (up to ~50 cm) of plants (0.1–0.5 kg) growing wild were collected in August 2000 (A, B, C), 2001 (D) and 2002 (E). The plants were gathered in 3 localities: A – Rudninkai (Šalčininkai district), B – Mikailiškės (Vilnius district), C, D, E – Gumbas (Šalčininkai district).

The voucher specimens were deposited in the Herbarium of the Institute of Botany (BILAS), Vilnius, Lithuania (numbers: A-59532, B-59526, C, D, E-59529).

All samples were collected at full flowering stage (inflorescence was indicated by the letter F and leaf by L). The plants were dried at room temperature (20–25 °C) and flowers were separated from stems and leaves before drying. The oils (0.4–1.1%) were prepared by hydrodistillation of 15–25 g of air-dried plants for 2.5 h. Analyses by GC/MS were performed using an HP 5890 chromatograph interfaced to a HP 5971 mass spectrometer (ionization voltage 70 eV) and equipped with a CP-Sil 8 CB capillary column (50 m × 0.32 mm i. d., film thickness 0.25 μm). The oven temperature was kept at 70 °C for 5 min, then programmed from 70 ° to 100 °C at a rate 3 °C/min, kept for 1 min, then programmed to 100–250 °C at a rate 25 °C/min and finally to isothermal 250 °C for 10 min, using He as the carrier gas (2.0 ml/min). Injector and detector temperatures were 250 °C and 280 °C, respectively. GC analyses were performed using a HP 5890 gas chromatograph equipped with FID. The column and analysis conditions were the same as in GC/MS.

The percentage composition of the essential oils was computed from GC peak areas without correction factors. Qualitative analysis was based on a comparison of retention times and mass spectra with the corresponding data in the literature [15] and computer mass spectra libraries (Wiley and NBS 54K).

RESULTS AND DISCUSSION

Myrtenol (tr. – 0.2%) was found in 2 out of 76 essential oils of *Tanacetum vulgare* L. presented in 25 papers reviewed by Lawrence [11]. The quantity of myrtenol exceeded 10% only in one paper from 16 reviewed by Keskitalo et al. [1]. The essential oils under study contained myrtenol as the predominant constituent (Table). The plants were collected from 3 habitats in 2000 (A, B and C) and from one locality in 2001 (D) and in 2002 (E). The volatile oils of myrtenol (15.5–24.9%) chemotype were determined in 4 inflorescence samples (AF, BF, CF, DF). The second main component of the above oils was camphor in the AF sample, trans-pinocarvone in BF, borneol in CF and 1,8-cineole in DF. The third dominant compound was 1,8-cineole in 3 inflorescence oils (AF, BF, CF) of plants collected in 2000, while in the oil of 2001 (DF) it was terpinen-4-ol. Myrtenol was the second major component in the oil (EF) of plants of 2002. The first main compound of the above oils was camphor and the third one 1,8-cineole.

The quantity of 1,8-cineole in the all essential oils under study was 11.6–27.2%. Four leaf oils (AL, CL, DL, EL) were of the 1,8-cineole (16.2–27.2%) chemotype. Camphor was the second component in 2 leaf oils (AL, EL), myrtenol in 1 sample (CL) and terpinen-4-ol in one oil (DL).

The inflorescence and leaf oils of the same sample of plants contained the same main constituents. Myrtenol, camphor, 1,8-cineole and α-pinene were the major components of both oils from the first locality (A) and myrtenol, trans-pinocarveol, 1,8-cineole and trans-sabinyl acetate – from the second habitat (B). Myrtenol, 1,8-cineole, and camphor were dominant constituents in 6 oils from the third vicinity. α-Pinene, borneol and terpinen-4-ol were the main compounds only in 4 oils. One of the reasons explaining the variability of the essential oil composition might be the ability of *T. vulgare* plants to produce different oils at the same locality (Table). This phenomenon was observed in *Thymus pulegioides* L. plants [16, 17]. The differences of soil chemical composition in the same locality might influence the oil production. Different quantity of sunny days in spring and summer might change the synthesis of the oil components. In 2002, spring and summer were warmer and dryer in comparison with 2000 and 2001. All above effects and/or some other reasons could influence the chemical composition of the volatile oils.

The largest part of the investigated oils consisted of oxygenated monoterpenes: in inflorescence oils – 69.1–75.0% and in the leaf oils – 62.5–66.7%. Four inflorescence oils contained 35.1–39.7% of

Table. **Chemical composition of essential oils of *Tanacetum vulgare* L. containing myrtenol as the major component**[a]

Compound	R I	AF	AL	BF	BL	CF	CL	DF	DL	EF	EL
1	*2*	*3*	*4*	*5*	*6*	*7*	*8*	*9*	*10*	*11*	*12*
Tricyclene	926	tr.	0.1	0.9	tr.	0.1	0.1	0.1	tr.	0.1	0.3
α-Thujene	931	tr.	0.2	0.5	tr.	0.1	0.1	tr.	0.1	tr.	
α-Pinene*	939	5.5	7.3	0.8	0.5	9.6	6.4	6.8	4.9	4.3	1.9
Camphene	953	1.5	2.0	0.9	1.1	1.5	0.6	0.6	0.8	2.0	1.6
Sabinene	976	1.7	3.6	0.9	1.5	1.0	0.9	1.1	1.7	2.4	2.8
β-Pinene*	980	2.2	3.1	0.1	tr.	2.4	1.9	2.4	2.3	tr.	0.7
Myrcene	991	0.4	0.4				tr.	tr.	0.2	tr.	
α-Terpinene	1018	0.5	0.6	0.3	0.5	1.4	1.8	3.3	3.1	1.2	0.6
o-Cymene	1025	0.7	1.3	0.1	0.3	0.9	1.9	1.3	3.4	0.7	1.5
1,8-Cineole	1033	**16.0**	**23.6**	12.3	14.5	**14.3**	**19.2**	**16.5**	**27.2**	**11.6**	**16.2**
γ-Terpinene	1062	0.9	1.0	0.2	0.3	2.4	3.2	5.7	5.6	3.9	1.6
Artemisia ketone	1062					tr.	0.1	0.1	tr.	tr.	
cis-Sabinene hydrate	1068		tr.	tr.	tr.			tr.			4.0
Terpinolene	1088	0.1		0.3	0.5	1.7	1.5	1.0	0.9	0.9	0.7
trans-Sabinene hydrate	1097							tr.	0.3	0.2	1.6
Camphenol-6	1109		tr.	tr.		0.1	tr.	tr.	tr.	0.1	
cis-Pinene hydrate *	1121	tr.			tr.						0.4
Chrysanthenone *	1123			0.1		0.2		tr.	tr.	0.3	0.5
trans-Pinocarveol *	1139		tr.	**16.1**	**15.9**	1.4	1.0	1.2	1.3	0.4	tr.
Camphor	1143	**16.2**	**14.7**	tr.	0.5	8.5	**13.4**	8.7	9.8	**17.8**	**11.0**
cis-Chrysanthenol *	1143		tr.			3.9	tr.	tr.	tr.	tr.	
Pinocarvone *	1162		tr.	0.9	0.9	2.4	0.2	1.6		2.2	0.9
Borneol	1165	1.8	3.4	2.8	2.3	14.9	7.5	tr.	0.1	5.9	4.1
Thujanol	1166							0.1	tr.	0.4	2.2
Pinocampheol *	1170			0.1	0.5	tr.	0.5	0.1	tr.	tr.	
cis-Pinocamphone *	1173		2.0	0.2	0.8	tr.	1.7	1.4	0.1		tr.
Terpinen-4-ol	1177	2.4	2.0	1.3	1.9	6.4	1.0	**15.0**	**13.8**	7.0	6.8
α-Terpineol	1189	1.6	3.2	0.1	0.2	1.8	1.1	0.9	0.1	1.8	1.1
Dihydrocarveol	1192		tr.	0.2	0.2	1.4		1.7	tr.	0.1	tr.
Myrtenal *	1193	3.1	1.1								
Myrtenol *	1194	**24.9**	**14.1**	**20.8**	**15.8**	**15.5**	**14.1**	**18.0**	**10.6**	**14.1**	**8.2**
trans-Piperitol	1205		tr.		tr.	0.8	tr.	0.1	tr.	0.3	0.3
iso-Dihydro carveol	1212			0.1	0.2	0.3		tr.	tr.	0.1	
trans-Dihydro carvone	1200	tr.	tr.		0.1	0.2	0.1	tr.	tr.		tr.
trans-Carveol	1217			0.2	0.2	0.1	tr.			0.2	
Myrtenyl acetate *	1235	tr.	tr.	0.5	3.8	1.5	0.1	3.6	1.7	tr.	0.6
Carvotanacetone	1246		tr.	0.2	tr.	0.9	tr.			0.5	0.4
Isobornyl acetate	1285	0.5	tr.	3.2	3.6		0.7	0.5	0.1	1.8	0.7
Thymol	1290									0.2	
trans-Sabinyl acetate	1291	2.6	0.9	9.9	5.0	0.1	0.9	tr.	tr.	2.1	0.1
iso-Thujyl acetate	1291					tr.	tr.			0.2	0.4
Dihydro neo-carveol acetate	1303				0.1		0.1	0.6	tr.		0.2
cis-Pinocarvyl acetate *	1312		tr.	0.1	0.2	0.3	0.1			2.6	0.6
Verbenol acetate *	1321				tr.		tr.		tr.	0.1	0.4
trans-Piperitol acetate	1346	tr.								0.4	
cis-Terpenyl acetate	1350		tr.		tr.		0.7	tr.	tr.		1.6
Eugenol	1356			tr.	tr.	tr.	tr.			0.2	0.2
α-Copaene	1376	0.3	tr.							0.1	0.1
Caryophyllene	1418	0.3	0.7	1.2	1.6	0.3	0.2	0.1	0.1		1.4
β-Gurjunene	1432			0.1	tr.	0.3	tr.				0.2

Table (continued)

1	2	3	4	5	6	7	8	9	10	11	12
β-Copaene	1435					0.2	0.1			0.7	
α-Humulene	1454	tr.	tr.	0.1	0.3	0.1	tr.	0.1	tr.	0.1	0.4
Germacrene D	1480	tr.	tr.		0.1		tr.	0.1	0.1	0.9	0.2
Bicyclogermacrene	1494			tr.		tr.		0.1	0.1	0.2	0.4
γ-Cadinene	1513			tr.				0.2	tr.	tr.	0.1
E-Nerolidol	1564	tr.	tr.	0.4	0.6	0.2	0.2	1.1	1.7		
Spathulenol	1576	1.3	1.0	1.2	1.8	0.2	0.1	0.8	1.7	0.8	2.3
Caryophyllene oxide	1581	1.1	3.0	1.2	1.9	0.2	0.3	1.1	3.1	1.2	2.0
Unknown 1	1586	1.3	1.3						0.9		
Salvial-4(14)-en-1-one	1595	tr.	tr.	tr.	0.6	0.5		0.1	0.1	0.5	0.4
trans-Arteannuic alcohol	1613			0.3	0.6	0.3	0.4	0.1	tr.	0.1	0.3
γ-Eudesmol	1630	tr.		0.5	0.9	0.2		0.1	tr.		0.3
α-epi-Muurolol	1641		tr.	0.4	0.4			0.3	0.3	0.2	
Caryophyllene-4(14)-8(15)-dien	1641	tr.		0.6	0.3	tr.	0.9			0.7	0.2
Vulgarone B	1647	2.0	0.9			tr.		tr.	tr.		
Vulgarol	1869	1.2	1.7								
Eicosane	2000	tr.	tr.	0.4	0.2	tr.	1.1	0.1	0.1	0.3	0.1
Total		90.1	93.2	80.5	80.7	98.6	84.2	96.7	96.3	92.0	82.6
Monoterpenes hydrocarbons		13.5	19.6	5.0	4.7	21.1	18.4	22.3	23.0	15.5	11.7
Oxygenated monoterpenes		69.1	65.0	69.1	66.7	75.0	62.5	70.1	65.1	70.6	62.5
Sesquiterpenes hydrocarbons		0.6	0.7	1.4	2.0	0.9	0.3	0.6	0.3	2.0	2.8
Oxygenated sesquiterpenes		6.9	7.9	5.0	7.3	1.6	3.0	3.7	7.9	3.8	5.6
Pinane skeleton *		35.7	27.6	39.7	38.4	37.2	26.0	35.1	20.9	24.0	14.2

[a] RI – retention index on nonpolar column, A–E letters indicate habitats, F – inflorescences, L – leaves. * Compounds with pinane skeleton.

compounds with a pinane skeleton (Table, marked compounds). Leaves produced a lower quantity (14.2–27.6%) of such components, except leaves from the second habitat (38.4%). Plants collected in 2002 synthesized lower amounts of compounds with a pinane skeleton than in previous years. A large part of oxygenated monoterpenes in earlier investigated plants of the Vilnius region consisted of ketones (camphor, α-thujone, artemisia ketone, etc.) [13]. In this study, the essential oils contained more monoterpene alcohols than ketones (Table). Further investigations of a large number of plant oil samples from different localities during several years will help to clarify the effect of seasonal conditions and other reasons influencing the essential oil composition.

CONCLUSIONS

In the essential oils under study, the main components were myrtenol, 1,8-cineole, camphor, trans-pinocarveol and terpinen-4-ol. Four out of 5 inflorescence essential oils were of myrtenol (15.5–24.9%) chemotype and 4 leaf oils were of 1,8-cineole (16.2–27.2%) type. The quantity of myrtenol was higher in the inflorescence than in leaf oils of the same plant samples. The opposite ratio was found for 1,8-cineole.

Oxygenated monoterpenes made up 69.1–75.0% of the inflorescence oils and 62.5–66.7% of leaf oils. Sixty-five identified constituents made up 80.5–98.6% of oil content.

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PAPRASTOSIOS BITKRĖSLĖS *Tanacetun vulgare* L., AUGANČIOS VILNIAUS APSKRITYJE, MIRTENOLIO CHEMOTIPO ETERINIAI ALIEJAI

S a n t r a u k a

Tirti paprastosios bitkrėslės, surinktos 2000–2002 m. Vilniaus apskrityje, žiedų ir lapų eteriniai aliejai. Pateikta 10 bandinių, kuriuose mirtenolis sudarė 14,1–24,9% žiedynų ir 11,6–16,5% lapų eterinio aliejaus. Kiti pagrindiniai komponentai: kamparas (8,5–17,8%) – 8 aliejuose, α-pinenas (4,9–9,6%) – 6 bandiniuose, terpinen-4-olis (6,4–15,0%) – 5 aliejuose ir borneolis (5,9–14,9%) –3 bandiniuose. Identifikuoti 65 junginiai sudarė 80,5–98,6% eterinio aliejaus, didžioji jų dalis – oksiduoti monoterpenai (62,5–75,0%).

Kaip rodo literatūros apžvalga, pasaulyje bitkrėslės eteriniuose aliejuose mirtenolio randama retai ir dažniausiai tik mažai (iš daugybės straipsnių tik viename jų užsiminta, kad mirtenolio rasta šiek tiek daugiau nei 10%).

Šiame straipsnyje aprašytas labai retas bitkrėslės aliejų mirtenolio chemotipas.